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James Russell Morris
(Ph. D. thesis)

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OPTICAL DIFFERENCE FREQUENCY GENERATION
OF FAR INFRARED RADIATION

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ABSTRACT

Three investigations of difference frequency generation (DFG) of far-infrared radiation by optical mixing are described: a theory of DFG by monochromatic, focused Gaussian pump laser beams, a theory of DFG by a picosecond pump laser pulse, and an experiment using ruby-pumped dye lasers. First, the theory of far-infrared generation by optical mixing of monochromatic, focused Gaussian beams in a uniaxial crystal is developed, taking into account the effects of diffraction, absorption, double refraction, and multiple reflections and total reflection at the boundary surfaces. (Reflection and transmission coefficients of a uniaxial crystal slab are derived by a new matrix technique.) Results of numerical calculations are presented. Focusing the pump beams appreciably enhances the far-infrared output despite the strong far-infrared diffraction. In a 1-cm long crystal, the optimum focal spot size is approximately equal to or smaller than the far-infrared wavelength for output frequencies less than 100 cm$^{-1}$. Double refraction of the pump beams is relatively unimportant. Both far-infrared absorption and boundary reflections have major effects on the far-infrared output and its angular distribution. The former is often the factor
which limits the output power. We show that a simple model treating
the nonlinear polarization as a constant 1/e-radius Gaussian distribution
of radiating dipoles adequately describes the effect of pump-beam
focusing. We also compare the results of our calculations with those
for second-harmonic generation. Second, a theoretical calculation of
far-infrared power spectra generated by picosecond pulses in a nonlinear
crystal is developed. The results are illustrated with two practical
examples: LiNbO$_3$ slabs oriented for rectification of the optical e-ray
and for beating of the optical o-ray with the optical e-ray. The
former is phase matched at 0 cm$^{-1}$; the latter, at both the forward-(FCPM)
and backward-collinear phase-matching frequencies. The one-dimensional,
time-dependent electric field is discussed and then used to explain
the origin of the oscillation periods in the power spectra. Finally,
a series of experiments using a pair of ruby-pumped dye lasers and a
novel dual-frequency dye laser system is described. With these two
laser systems, continuously tunable far-infrared radiation in the
frequency range 20 to 190 cm$^{-1}$ was generated. Forward-, backward-, and non-collinear phasematching in LiNbO$_3$ at frequencies between 20
and 160 cm$^{-1}$ were investigated; 90° noncritical FCPM was also observed
in ZnO at 190 cm$^{-1}$, CdS at 180 cm$^{-1}$, and ZnS at 91 cm$^{-1}$. The highest
peak power (~200 mW) and the broadest tuning range using a single
crystal sample (40 to 160 cm$^{-1}$) were obtained with noncollinear phase
matching in LiNbO$_3$. 
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CHAPTER I. DIFFERENCE FREQUENCY GENERATION OF FAR-INFRARED RADIATION AS A NONLINEAR OPTICS RESEARCH TOPIC

I. Introduction

For the purposes of this thesis, optical difference frequency generation is defined as a process in which two optical waves mix in an electro-optic crystal to create a third electromagnetic wave at their difference frequency. There are three fundamental reasons why investigation of this process has been an active area of nonlinear optics research for over a decade. First, the generation process itself differs from second harmonic generation (SHG) enough to require a separate analysis. When the pump frequencies are a factor of ten or more larger than the difference frequency as in typical far-infrared generation experiments, the large diffraction angle of the difference frequency wave compared to the pump waves can make the approximations used to describe SHG inapplicable. Second, the dispersion of the DFG susceptibility provides data on the low frequency excitations of the nonlinear crystal (e.g., magnons and phonons) which do not contribute to the optical or near-infrared SHG susceptibilities. Finally, DFG is a means of transferring the tunability of optical lasers or parametric oscillators into lower frequency regions where tunable bright coherent sources have not yet been developed.

Since the 1920s, radio engineers have known how to generate coherent radiation from a few kHz to 100 MHz or more. In the 1930s, the split-anode magnetron extended radio frequency techniques to cm wavelengths, and in response to the need for cm wavelength radar, the klystron,
the multicavity magnetron, and the traveling wave tube - invented during the World War II years - extended the wavelength range to the millimeter regime. After the war, these techniques were pushed into sub-millimeter wavelengths so that now carcinotrons can reach 350 μm.\(^1\) At the other end of the spectrum from the near-ultraviolet down through the near-infrared, organic dye lasers have provided bright tunable sources while parametric oscillators and spin-flip Raman lasers cover a region that extends down into the medium-infrared.

The 30 to 500 cm\(^{-1}\) (333 to 20 μm wavelength) part of the far-infrared region has proved more resistant to the development of bright tunable sources using either radio frequency or optical techniques. For decades one of the best radiation sources for far-infrared spectroscopy has been the quartz envelope high-pressure mercury arc lamp; when used in Fourier transform spectroscopy, it is still the most versatile far-infrared source. An ideal 5000\(°\)K blackbody of 1 cm\(^2\) surface area is a convenient source to compare with, since between 30 and 70 cm\(^{-1}\) the high-pressure mercury arc radiates like a 5000 to 6000°K blackbody. (By 150 cm\(^{-1}\) its effective temperature has dropped to 1000°K.\(^2\)) Although its total radiated power is 3.5 kW, only 0.54 W or 0.015% of this power is in the 30 to 500 cm\(^{-1}\) spectral region and only 0.13 mW is in a 1 cm\(^{-1}\) bandwidth at 100 cm\(^{-1}\).

Many quantum electronic systems generate coherent far-infrared radiation without using difference frequency generation. Glow discharge pumped HCN, H\(_2\)O, H\(_2\)S, and SO\(_2\) molecular gas lasers\(^3\) provide bright coherent continuous wave (cw) or repetitively pulsed sources. Since
they are not tunable, they are of limited use for spectroscopy. Optically pumped molecular gas lasers offer more promise as a spectroscopic source. With a single TEA CO₂ pump laser, many organic molecules can be made to lase - each at its own far-infrared wavelength. Although these sources are not tunable either, the sheer number of such wavelengths should make such a system useful for low resolution spectroscopy of broad band transitions and high resolution spectroscopy of line transitions (by means of a Stark shift, etc.). PbSe and Pb_{1-x}Sn_x Te semiconductor p-n junction lasers have been operated in the far-infrared. The PbSe laser has been pressure tuned from 455 to 1370 cm⁻¹; so a Pb_{1-x}Sn_x Te laser system might be pressure tunable over a significant part of the far-infrared. Stimulated Raman and stimulated polariton scattering have also provided tunable far-infrared sources. Very recently, far-infrared radiation has also been observed in the output of the InSb spin-flip Raman laser.

Relativistic electron beams can also generate far-infrared radiation at harmonics of the accelerator bunching frequency or at a wavelength approximately equal to the Doppler-shifted period of a spatially periodic magnetic field. The harmonics of the bunching frequency can be generated, for example, as Cerenkov radiation in a waveguide loaded with a periodic slow wave structure. The Doppler shifted schemes have been of more interest lately due to the possibility of free-electron laser action and a potentially much larger tuning range. Elias et al. have measured a gain of 0.07 per pass at 10.6 μm using a 70 mA instantaneous peak electron current, 24 MeV electron beam...
in a 5.2 m long, 3.2 cm pitch, 2.4 kG helical magnetic field. Using the same magnet with a 1 amp, 8 MeV electron beam, the output wavelength and the gain per pass would be 95 µm (105 cm⁻¹ frequency) and e²⁷, respectively. Unfortunately, electron accelerators capable of reaching relativistic energies are expensive to build and operate; thus, far-infrared sources based on relativistic electron beams will probably be feasible, if at all, only at laboratories that possess the necessary accelerator for other purposes.

The experimental observations of far-infrared generation by nonlinear optical difference frequency mixing can be sorted into two groups by the spectral characteristics of the pump source. In the first group of experiments, the Fourier components of a single broad bandwidth laser form the pump source. With such a laser Zernike and Berman¹⁴ observed the first reported nonlinear optical difference frequency generation of far-infrared radiation in 1965. They illuminated a quartz crystal that was cut for collinear phase matching at ~100 cm⁻¹ with the broad bandwidth output of a free-running Nd³⁺:glass laser and observed random difference frequency spikes. Yajima and Takeuchi¹⁵ reported observing the far-infrared radiation generated by a Q-switched Nd³⁺:glass laser in LiNbO₃ in their paper on DFG by picosecond pulses. Yajima and Takeuchi,¹⁵,¹⁶ Yang et al.,¹⁷ and Takeuchi et al.¹⁸ have observed the self-beating of the frequency components of a picosecond mode-locked Nd³⁺:glass laser pulse in various crystals: LiNbO₃,¹⁵-¹⁷ ZnTe,¹⁵,¹⁶ ZnSe,¹⁵ CdS,¹⁵ quartz,¹⁵ and LiIO₃;¹⁸ these experiments have demonstrated a means of generating tunable far-infrared pulses
a few tens of picoseconds in duration. More recently, Matsumoto and Yajima\(^{19}\) have reported far-infrared generation in ZnTe, LiNb\(_3\)O, and ZnSe using the output of broad bandwidth ruby-pumped dye lasers.

The second group, which includes the majority of far-infrared generation by difference frequency mixing experiments, uses two nearly monochromatic pump beams. Such experiments have been reported by over half a dozen research groups under a wide variety of experimental conditions in the twelve years since Zernike and Berman's experiment. Many different combinations of laser systems and nonlinear crystals have been used to investigate the DFG process: CO\(_2\) laser lines in InSb,\(^{20-25}\) GaAs,\(^{26-42}\) and ZnGeP\(_2\);\(^{43}\) a CO\(_2\) laser and a spin-flip Raman laser beam in InSb;\(^{44-49}\) R\(_1\) and R\(_2\) lines from a specially constructed ruby laser in ZnTe;\(^{50-52}\) temperature tuned ruby laser lines from a pair of simultaneously Q-switched ruby lasers in LiNb\(_3\)O\(_3\);\(^{53-57}\) and quartz;\(^{53}\) ruby-pumped Raman lasers in GaP;\(^{58-60}\) and ruby-pumped dye lasers in LiNb\(_3\)O, ZnO, ZnS, and CdS\(^{61-63}\) and in reduced (black) LiNb\(_3\)O.\(^{64}\)

The most common experimental scheme uses collinear pump beams and generates a far-infrared wave that propagates in the same direction as the pump waves. Usually the mixing process is phase matched for efficient far-infrared generation, but in a few experiments\(^{20,25,28,50-52}\) phase matching was unnecessary; in most of these cases the coherence length for far-infrared generation was longer than the effective sample length due to either a fortunate coincidence\(^{20,28,50-52}\) or to a large optical absorption coefficient.\(^{64}\) Usually collinear phase matching is attained through the dependence of the birefringence of uniaxial crystals.
on their orientation. This mechanism doesn't exist in isotropic crystals, so various ingenious methods have been employed to achieve collinear phase matching: Nguyen and Patel\textsuperscript{22,23} used a static magnetic field to induce birefringence in InSb through the Voigt\textsuperscript{22} or Faraday\textsuperscript{23} effects; Zernike\textsuperscript{21} temperature tuned the 9.6 and 10.6 $\mu$m refractive indices of InSb; Nguyen and Bridges\textsuperscript{44-48} used the free carrier concentration (by selecting a properly doped sample) to set the plasma frequency and hence the 9.6 and 10.6 $\mu$m refractive indices of InSb; finally, Thompson and Coleman,\textsuperscript{29-31} the Bell Labs groups,\textsuperscript{26,27} and Bogatkin et al.\textsuperscript{32} have used the waveguide dispersion of GaAs (or GaAs filled) waveguides.

In isotropic materials the most generally useful phase matching technique is noncollinear phase matching (NCPM) in which a slight angle between the pump beams is used to compensate for the higher far-infrared refractive index. Of course, NCPM can also be used in birefringent crystals. DeMartini,\textsuperscript{58-60} the Berkeley group,\textsuperscript{62} and the National Magnet Lab group\textsuperscript{34-42} have successfully employed this technique; the latter two groups have demonstrated that far-infrared generation in the NCPM geometry can be very efficient. In particular, the Magnet Lab group has developed a folded noncollinear geometry,\textsuperscript{39-42} a modification of the multiple internal reflection scheme first suggested by Armstrong et al.,\textsuperscript{65} in which the pump beams propagate in a zig-zag pattern down a long crystal with a rectangular cross section while the far-infrared beam propagates parallel to the long axis of the sample. This scheme has increased the far-infrared power that can be obtained from mixing CO\textsubscript{2} lasers in GaAs by several orders of magnitude while
preserving the convenient "one crystal works for all difference frequencies" feature of noncollinear phase matching in an isotropic crystal. With this efficient folded geometry Aggarwal et al.\textsuperscript{41,42} have generated continuous wave far-infrared radiation that is step-tunable from 10 to 140 cm\textsuperscript{-1} with less than a 100 kHz bandwidth at significant power levels; at 100 cm\textsuperscript{-1} two 25W CO\textsubscript{2} lasers generated \(\sim\) 2 \(\mu\)W of far-infrared power.

The largest group of theoretical papers on DFG of far-infrared radiation have treated the two pump beams and the far-infrared wave as diffractionless apertured plane waves. De Martini\textsuperscript{60} discussed DFG in an isotropic medium for a pair of plane pump waves with a common plane of incidence when the nonlinear polarization vector is perpendicular to this plane. He has included the far-infrared absorption and the polariton dispersion of both the linear and nonlinear susceptibilities as well as the single surface boundary conditions.\textsuperscript{66} Shen\textsuperscript{10} has discussed the connection between DFG and stimulated polariton scattering due to the spin-flip Raman transition in InSb and has shown that the interference between the resonant magnetic dipole and the non-resonant electric dipole terms of the nonlinear difference frequency susceptibility explains the asymmetry of the experimentally measured\textsuperscript{25} dependence of the difference frequency power on the static magnetic field strength. In an earlier paper, Brown and Wolff\textsuperscript{67} have discussed the resonant magnetic dipole DFG process by itself. Paraire \textit{et al.}\textsuperscript{56} have generalized the plane wave solution to treat a wedge-shaped sample of a uniaxial crystal in the special case when
the c-axis is normal to the input surface and the pump wave vectors, the surface normal vectors, and the c-axis are all coplanar. They discuss generation of both far-infrared o- and e-rays, single surface reflection and transmission at the output surface of the wedge, and the dispersion of the nonlinear susceptibility.

Prior to the research discussed in chapter II, several models which include diffraction effects have appeared in the literature. In their experimental papers, Zernike and Berman and Faries et al. used the far-field diffraction pattern of the nonlinear polarization created by a uniformly illuminated cylinder with plane wave fronts normal to the axis of the cylinder and included the effect of total reflection at the exit surface. Our results show that this model or its generalization to Gaussian beam illumination is a good description of far-infrared generation when walk-off effects are not too large. Boyd and Kleinman and Faries have given the nonlinear polarization for DFG by interacting Gaussian beams, but they have not given any results using this polarization. Abdullin et al. have also described far-infrared generation by a pair of interacting Gaussian beams. They restricted their treatment to isotropic media and ignored boundary reflections, total reflection, and far-infrared absorption to emphasize the physical explanation of the phase matched cone seen in our calculations as Cerenkov radiation. (Note: Abdullin et al.'s power formula is an asymptotic expression that is valid only when the Cerenkov cone is significantly narrower than its opening angle.)
The theory of DFG of far-infrared radiation by picosecond optical pulses has been developed independently by Yajima and Takeuchi\textsuperscript{15,16} and by Morris and Shen.\textsuperscript{70} While we have emphasized the intensity variation in the far-infrared radiation by considering the case of a single coherent picosecond pump pulse, Yajima and Takeuchi have expanded the pump beam as a sum of laser cavity longitudinal modes to compare the power generated by a mode-locked (coherent case) pulse to that generated by the random fluctuations of a Q-switched Nd\textsuperscript{3+} :glass laser or a broad bandwidth dye laser.\textsuperscript{19} They have also included the effect of optical dispersion which we have omitted for simplicity; for LiNbO\textsubscript{3} this merely displaces the phase matched frequencies or changes the coherence length slightly, but for crystals like ZnTe in which the difference between the optical and far-infrared refractive indices is small, it can change the coherence length appreciably. Yajima and Takeuchi have neglected the backward phase matched signal\textsuperscript{70} without which the experimental results\textsuperscript{15,17} for LiNbO\textsubscript{3} cannot be understood.

There are two review articles that deal with DFG. Shen's\textsuperscript{71} review covers the status of far-infrared generation by optical mixing research up to 1974; this paper is an excellent introduction to the DFG literature. Warner's\textsuperscript{72} review article emphasizes up-conversion and includes only a very sketchy treatment of DFG; it does, however, discuss DFG at frequencies above the far-infrared.

In the subsequent chapters of this thesis, we describe three investigations of far-infrared generation by difference frequency mixing of optical or near-infrared laser beams in nonlinear crystals -
two theoretical and one experimental. In chapter II we present a theory of far-infrared generation by difference frequency mixing of two monochromatic TEM$_{oo}$ mode laser beams and a series of computer calculations based on this theory. We discuss the important effects of a far-infrared diffraction angle that is much larger than the optical diffraction angles: loss of far-infrared power to totally reflected modes and variation of the phase mismatch from one far-infrared plane wave mode to another. We also discuss the effects of far-infrared absorption and optical walk-off, and we present the solution to the crystal-vacuum boundary conditions for a uniaxial crystal in a matrix form that focuses attention on the physical reflection processes rather than on the many simultaneous equations. In chapter III we describe a theory of DFG from the beating of the Fourier components of a single picosecond pulse (or a mode-locked train of such pulses) using a quasi-plane wave approximation. We discuss two types of DFG of far-infrared radiation with picosecond pulses: (1) the propagating wave analog of optical rectification in which an optical $o$-ray or $e$-ray beats with itself and (2) the beating of an optical $o$-ray with an optical $e$-ray in which spectral narrowing due to phase matching can dominate. Both forward and backward propagating far-infrared waves are generated by this process, and reflection off the surfaces of the nonlinear crystal mixes the two waves. The theory predicts that, with an appropriately chosen backward collinear phase matching frequency, generation of the backward propagating wave should be easily observable in LiNbO$_3$ slabs less than several mm thick; this prediction and the general features of our theory have been experimentally verified by
In chapter IV, we discuss the experimental observation of far-infrared radiation from 20 to 160 cm\(^{-1}\) in \(\text{LiNbO}_3\) and at the maximum forward collinear phase matching frequency in \(\text{ZnO}, \text{CdS},\) and \(\text{ZnS}\) using two nearly monochromatic dye laser beams. Substantial signal-to-noise ratios are observed for forward collinear, backward collinear, and noncollinear phase matched DFG in \(\text{LiNbO}_3\) and for forward collinear phase matched DFG in \(\text{ZnO}\); of these, noncollinear phase matched DFG in \(\text{LiNbO}_3\) is the most efficient process. The direct observation of backward collinear phase matched generation of a far-infrared wave with nearly monochromatic laser beams in \(\text{LiNbO}_3\) is indisputable verification of the origin of the low frequency peak in the mixed polarization mode picosecond pulse experiment of Yang, Richards, and Shen. \(^{17}\) We also describe a novel dual frequency dye laser that was developed for and used in some of these experiments. Appendices A and B describe some of the mathematical details of chapter II. Appendix C describes the numerical methods used for the computer calculations presented in chapter II and contains a listing of the computer code. Appendix D gives the far-infrared field for the square optical pulse case discussed in chapter III. Appendix E describes the nonlinear least-squares fitting procedure used to obtain the dispersion of the \(\sigma\)-ray far-infrared absorption coefficient from our experimental data. The remainder of this chapter describes the connection between DFG and other nonlinear optical processes.
II. Relationship Between Difference Frequency Generation of Far-Infrared Radiation and Other Nonlinear Optical Processes

Difference frequency generation, sum frequency generation, second harmonic generation, optical rectification, up-conversion, parametric amplification or oscillation, and the linear electro-optic effect are all described by the second order nonlinear susceptibility tensor and Maxwell's equations. These processes are all manifestations of a single nonlinear optical interaction in different frequency regimes and under different initial or boundary conditions. Parametric amplification, parametric oscillation, and optical rectification have especially close connections to DFG. When both parametric amplification (or oscillation) and DFG involve the same set of three frequencies, they differ only in their initial and boundary conditions. Drastically reducing the intensity of the low frequency pump (signal) beam, lengthening the nonlinear crystal sample, and enclosing the crystal in an optical resonator for the signal or idler (difference frequency) waves converts a phase matched DFG experiment into a parametric amplifier experiment. In the parametric oscillator, the signal beam builds up from spontaneous emission noise at the set of cavity modes that are most nearly phase matched. Optical rectification, when observed through propagating waves, is the zero frequency limit of difference frequency generation; however, in the more common experimental arrangement optical rectification is observed as an electrical pulse across parallel capacitor plates and is thus the longitudinal or
electro-static solution to Maxwell's equations rather than the transverse, propagating wave solution observed in DFG experiments.

The electro-optic effect is the DC field limit of the combined sum- and difference-frequency processes which are degenerate when one mixing frequency is zero (i.e. when the optical input and output waves have the same frequency). When absorption at the pump and at very low far-infrared frequencies is negligible, the second order susceptibility (discussed below) obeys a permutation symmetry and the optical rectification and electro-optic effect susceptibilities are equal.

A brief description of the pertinent part of nonlinear optical susceptibility theory will clarify these interrelationships.

Since nonlinear optical effects are observed with high photon flux laser beams, we can use the semi-classical theory of radiation to describe the growth and propagation of the nonlinearly generated electric fields. Furthermore, because we are only concerned with effects induced by external fields, the electromagnetic field is described by Maxwell's equations in which the induced current density, \( \mathbf{J} \), can be expanded in a multipole series

\[
\mathbf{J} = \frac{\partial \mathbf{P}}{\partial t} + c \mathbf{v} \times \mathbf{M} - \frac{\partial}{\partial t} (\mathbf{V} \cdot \mathbf{Q}) + \ldots
\]  

(1)

where \( \mathbf{P} \), \( \mathbf{M} \), and \( \mathbf{Q} \) are the electric dipole, magnetic dipole, and electric quadrupole polarizations, respectively. Although each of these terms can have both linear and nonlinear contributions, I shall discuss
only the electric dipole polarization term, $\partial \mathbf{P} / \partial t$, as this is sufficient to illustrate the phenomena that are related to difference frequency generation. To further simplify the discussion, I shall make the customary dipole or local field approximation in which $\mathbf{P}$ depends only on the history of $\mathbf{E}$ at the same point in space.

When the applied fields are much smaller than the atomic fields seen by electrons in the medium, the polarization $\mathbf{P}(\mathbf{r}, t)$ can be expanded into a power series in the applied electric field $\mathbf{E}(\mathbf{r}, t)$. Each term of this series has associated with it a response function tensor, $R^{(n)}$, such that

$$p_i(\mathbf{r}, t) = \sum_n p_i^{(n)}(\mathbf{r}, t)$$  \hspace{1cm} (2)

with

$$p_i^{(n)}(\mathbf{r}, t) = \sum_{j_1} \ldots \sum_{j_n} \int_{-\infty}^{0} d\tau_1 \ldots \int_{-\infty}^{0} d\tau_n R^{(n)}_{i,j_1,j_1',\ldots,j_n,j_n'}(\mathbf{r}, t) .$$  \hspace{1cm} (3)

Since the part of $R^{(n)}$ that is antisymmetric in the exchange of any two of the pairs $(j_1, \tau_1), \ldots, (j_n, \tau_n)$ does not contribute to $\mathbf{P}^{(n)}$, the tensor $R^{(n)}$ can be rendered unique by requiring that it be invariant under all the permutations of the pairs $(j_1, \tau_1), \ldots, (j_n, \tau_n)$; this property is called the intrinsic permutation symmetry.

The dipole susceptibility tensors are simply the Fourier transforms of these dipole response tensors:
Then, the Fourier transform of \( \phi^{(n)}(t) \) is given by:

\[
\phi^{(n)}(w; \omega_1, \ldots, \omega_n) = \int_{-\infty}^{0} dt_1 \cdots \int_{-\infty}^{0} dt_n \phi^{(n)}(t_1, \ldots, t_n) \\
\times \exp[-i(\omega_1 t_1 + \ldots + \omega_n t_n)]
\]

Then, the Fourier transform of \( \hat{\phi}^{(n)}(t) \) is given by:

\[
\hat{\phi}^{(n)}(w) = \int dw_1 \cdots \int dw_n \delta(w - \omega_1 - \ldots - \omega_n) \phi^{(n)}(w; \omega_1, \ldots, \omega_n):
\]

\[
\hat{\phi}(w_1) \cdots \hat{\phi}(w_n)
\]

with

\[
\hat{\phi}(t) = \int \hat{\phi}(\omega) \exp(-i\omega t) \, d\omega
\]

and

\[
\hat{\bar{\phi}}(t) = \int \hat{\phi}(\omega) \exp(-i\omega t) \, d\omega.
\]

(The dependence of \( \hat{\phi} \) and \( \hat{\bar{\phi}} \) on \( \tau \) has been omitted to simplify the notation.) The tensor \( \phi^{(n)}\)

\( (i, j_1, \ldots, j_n) (\omega; \omega_1, \ldots, \omega_n) \) is invariant under any permutation of the pairs \( (j_1, \omega_1) \cdots (j_n, \omega_n) \) due to the intrinsic permutation symmetry of \( \phi^{(n)} \), and it also obeys the additional permutation symmetry:

\[
\phi^{(n)}(i, j_1, \ldots, j_n) (\omega; \omega_1, \ldots, \omega_n) = \phi^{(n)*} (j_1, \ldots, j_n, i) (-\omega_1; \omega_2, \ldots, -\omega_n).
\]

Furthermore, since the fields \( \hat{\phi}(t) \) and \( \hat{\bar{\phi}}(t) \) are real valued, \( \phi^{(n)} \) satisfies the further symmetry relation
Although Eq. (5) also describes the mixing of the Fourier components of a mode-locked or other broad bandwidth laser, the second order nonlinear optical frequency mixing processes are most simply discussed using monochromatic laser fields. For a single monochromatic laser with 
\[ \tilde{E}(\omega) = \tilde{E}_1 \delta(\omega - \omega_1) + \tilde{E}_1^* \delta(\omega + \omega_1), \]
only second harmonic generation (SHG) and optical rectification (OR) are caused by \( \phi^{(2)} \) and

\[ \tilde{p}(\omega) = \phi^{(2)}(2\omega_1; \omega_1, \omega_1) \tilde{E}_1 \tilde{E}_1^* \delta(\omega - 2\omega_1) \]

\[ + 2\phi^{(2)}(0; \omega_1, -\omega_1) \tilde{E}_1 \tilde{E}_1^* \delta(\omega) \]  \( \text{(8)} \)

where from Eq. (8) on I shall omit those terms which are merely the negative frequency counterparts of some explicitly displayed term.

With two monochromatic laser beams, \( \tilde{p}^{(2)} \) becomes

\[ \tilde{p}^{(2)}(\omega) = 2\phi^{(2)}(\omega_1 - \omega_2; \omega_1, -\omega_2) \tilde{E}_1 \tilde{E}_1^* \delta(\omega - \omega_1 + \omega_2) \]

\[ + 2\phi^{(2)}(\omega_1 + \omega_2; \omega_1, \omega_2) \tilde{E}_1 \tilde{E}_2 \delta(\omega - \omega_1 - \omega_2) \]  \( \text{(9)} \)

\[ + \text{SHG and OR terms.} \]

The linear electro-optic effect is described by the mixing of a laser field, \( \tilde{E}_1 \), with a DC field, \( \tilde{E}_dc \), which can be obtained from Eq. (9) by setting \( \omega_2 \) to zero and replacing \( (\tilde{E}_2 + \tilde{E}_2^*) \) with \( \tilde{E}_dc \):

\[ \phi^{(n)}(\omega; \omega_1, \ldots, \omega_n)^* = \phi^{(n)}(-\omega; -\omega_1, \ldots, -\omega_n). \]  \( \text{(7)} \)
The tabulated electro-optic coefficients, $r_{ijk}$, are expressed in terms of the induced change in the inverse of the linear dielectric tensor $\varepsilon'(E_{dc})$ so that in the principal axis coordinate system of a crystal:

$$r_{ijk} = \frac{\partial}{\partial E_{dc,k}} \left[ \varepsilon'(E_{dc}) \right]^{-1}_{ij}$$

$$= \frac{\partial}{\partial E_{dc,k}} \left[ \varepsilon'(0) + 8\pi \phi'(2)(\omega_1;\omega_1,0)E_{dc} \right]^{-1}_{ij}$$

$$= -8\pi E_{dc0}(0)E_{dc0}^{-1}(0)\phi'(2)(\omega_1;\omega_1,0).$$

Throughout the remainder of this thesis, I shall use Bloembergen's notation for the susceptibility $\chi^{(2)}$ for historical reasons and because it eliminates the miscellaneous factors of two in Eqs. (8), (9), and (10); in this notation

$$P_{ij}(\omega) = \sum_{jk} \chi_{ijk}^{(2)}(\omega_1;\omega_2)\delta_{j\omega_1}\delta_{k\omega_2}\delta(\omega-\omega_1-\omega_2).$$

and the peak electric field at $\omega_1 \neq 0$ is $2|E_0(\omega)|$. The relationship between the two notations and between them and the $d$ tensor of Boyd and Kleinman[68] which is used in the SHG tabulations of the Landoelt-Boernstein series[79] are summarized below:
When the optical frequencies are far from the electronic resonances of the material, Raman scattering from its simultaneously infrared and Raman active far-infrared elementary excitations is responsible for the dispersion of $\chi^{(2)}$ for DFG. Although spin-flip transitions, magnons, plasmons, etc. can also lead to dispersion of $\chi^{(2)}$, TO phonons cause the dispersion of $\chi^{(2)}$ in all of our experiments. In section IV of his review paper, Shen has given a clear, concise derivation of the dispersion of $\chi^{(2)}$ due to Raman scattering from polaritons associated with a TO phonon mode. He starts with Maxwell's equations for the high frequency pump ($\omega_1$), low frequency pump or Raman Stokes ($\omega_2$), and difference frequency ($\omega_3 = \omega_1 - \omega_2$) electromagnetic fields, a damped simple harmonic oscillator (SHO) equation for the TO phonon mode, and nonlinear coupling terms that can be derived from the phenomenological energy density:

$$ F = \dot{Q}^{*}(\omega_3) \times \hat{E}(\omega_1) \hat{E}^{*}(\omega_2) + \hat{E}^{*}(\omega_3) \times \chi^{(2)}_{\text{mix}} \times \hat{E}(\omega_1) \hat{E}^{*}(\omega_2) $$

$$ + Q^{*}(\omega_3) \times \hat{A} \times \hat{E}(\omega_3) + c.c. \quad (14) $$

\[
\begin{align*}
\chi_{\text{SHG}}^{(2)} &= 2d_{\text{SHG}}^{(2)} = \psi_{\text{SHG}}^{(2)} \\
\chi_{\text{OR}}^{(2)} &= 4d_{\text{OR}}^{(2)} = 2\psi_{\text{OR}}^{(2)} \\
\chi_{\text{eo}}^{(2)} &= 4d_{\text{eo}}^{(2)} = 2\psi_{\text{eo}}^{(2)} \\
\chi_{\text{mix}}^{(2)} &= 2d_{\text{mix}}^{(2)} = 2\psi_{\text{mix}}^{(2)}
\end{align*}
\]
The nonlinear polarization at the three frequencies $\omega_i$ (i=1,2,3) and
the force on the SHO are given by the equations $\mathbf{P}^{(2)}(\omega_i) = 3F/3E^*(\omega_i)$
and $\mathbf{F}(\omega_3) = 3F/3Q^*(\omega_3)$, respectively. Solving the SHO equation for
$Q(\omega_3)$ and eliminating $Q(\omega_3)$ from the three Maxwell's equations. Shen
obtains the Raman Stokes and difference frequency wave equations:

\[ \{\nabla x \nabla x - (\omega_2/c)^2 [\epsilon^{(2)}(\omega_2) + 4\pi \chi^{(3)}_R] \nabla (\omega_1) \nabla (\omega_1) \} \epsilon^{(2)}(\omega_2) \]

\[ = (4\pi \omega_2^2/c^2) \chi^{(2)} : \epsilon^{(2)}(\omega_1) \epsilon^*(\omega_3) \]  

\[ [\nabla x \nabla x - (\omega_3/c)^2 \epsilon^{(2)}(\omega_3)] \epsilon^{(2)}(\omega_3) = (4\pi \omega_3^2/c^2) \chi^{(2)} : \epsilon^{(2)}(\omega_1) \epsilon^*(\omega_2) \]

where \( ^{80} \)

\[ \epsilon(\omega_3) = \epsilon_\infty(\omega_3) + 4\pi A^* A/(\omega_0^2 - \omega_3^2 - i\omega_3 \Gamma) \]

\[ \chi^{(2)} = \chi^{(2)} + A^* A/(\omega_0^2 - \omega_3^2 - i\omega_3 \Gamma) \]

\[ \chi^{(3)}_R = f^* f^* / (\omega_0^2 - \omega_3^2 + i\omega_3 \Gamma) \]

For fixed $\omega_1 \gg \omega_3$, the tensors $A$, $f$, $\epsilon_\infty$, and $\chi^{(2)}$ are all nearly
independent of $\omega_3$ unless $\omega_1$ is close to an electronic transition of
the crystal. As can be seen from Eqs. (15) and (16), far-infrared
reflection experiments determine $A$ through $\epsilon(0)$ and $\epsilon_\infty$, and Raman
scattering experiments measurements determine $f$ through $\chi^{(3)}_R$. The
linear electro-optic coefficient determines $\chi^{(2)}(\omega_1,\omega_2,\omega_1,\omega_2)$ through
Eqs. (11), (13c), and the permutation symmetry of $\chi^{(2)}$; together with
and if this determines $\chi^{(2)}$. (If its dispersion can be neglected, $\chi^{(2)}$ can also be determined from the results of SHG experiments.)
REFERENCES


12. L. R. Elias, W. M. Fairbank, J. M. J. Madey, H. A. Schwettman, and
    or F. A. Hopf, P. Meystre, M. O. Scully, and W. H. Louisell,
    320 (1971).
    Phys. 11, 268 (1972).
24. V. T. Nguyen, A. R. Strand, A. M. Jean-Louis, and G. Duraffourg,
    in The Physics of Semimetals and Narrow Gap Semiconductors,
    p. 231.
    13, 357 (1968).
45. V. T. Nguyen and T. J. Bridges, in International Conference on Submillimeter Waves and their Applications, Atlanta, 1974, Conference Digest, p. 41.
66. See, for example, N. Bloembergen, *Nonlinear Optics*, (Benjamin, New York, 1965).


80. In the definitions of $\varepsilon^{(\omega_2)}, \chi^{(2)},$ and $\chi_R^{(3)}$, the "*" means sum over the index that corresponds to $\omega_2$. Also, the index of $\chi_R^{(3)}$ which is not summed over in Eq. (15) is the index of the first $f$ that corresponds to $E^*(\omega_2)$. 

CHAPTER II. THEORY OF FAR-INFRARED GENERATION BY DIFFERENCE
FREQUENCY MIXING OF MONOCHROMATIC LASER BEAMS

I. Introduction

Far-infrared generation by optical mixing has recently received increasing attention.\(^1\) It has the potential of providing a coherent tunable far-infrared source which compliments far-infrared molecular lasers. The most commonly used scheme is that of difference-frequency generation (DFG) by mixing of two laser beams in a non-centrosymmetric crystal. With dye lasers,\(^2,3\) CO\(_2\) lasers,\(^4-16\) or spin-flip Raman lasers\(^17-20\) as the pump beams, DFG can provide a far-infrared source discretely or continuously tunable from 1 cm\(^{-1}\) to 200 cm\(^{-1}\) or more. The output linewidth can easily be less than 0.1 cm\(^{-1}\) as determined by the pump laser linewidths. In most cases, the output is in pulses with pulsewidths between 10 nsec and 10 \(\mu\)sec, but CW operation has recently been achieved.

A serious limitation of far-infrared generation by optical mixing has been the attainable average power, although so far as spectral power per unit solid angle is concerned it is already better than a blackbody source at 5000\(^{\circ}\)K.\(^1\) While focusing of the pump beams may increase the far-infrared output, it is not clear how tight the focusing can be before the detrimental effect of far-infrared diffraction sets in. No adequate theoretical calculation of nonlinear far-infrared generation with focusing and diffraction properly taken into account has been reported. Experimentally, on the other hand, a tight focusing geometry has so far been avoided. As a result, the full potential of
nonlinear far infrared generation has not been assessed.

In the literature, the plane-wave theory was often used to interpret the results of far-infrared generation experiments.\(^{2,6,7,11,12,21-25}\) The theory assumes a single spatial Fourier component for each monochromatic wave so that the nonlinear process is characterized by a single phase matching relation. However, when the pump beams are focused to a spot comparable in size to the far-infrared wavelength, far-infrared diffraction is important and the spatial Fourier components of the output extend over a large cone. Each Fourier component now has its own phase matching relation with respect to the pump beams. Since it is not possible to phase match all the Fourier components simultaneously, focusing of the pump beams does not improve the far-infrared output power as much as the plane-wave theory predicts.

The plane-wave theory also assumes a single transmission coefficient for the far-infrared output across the boundary surface. Actually, with the far-infrared output extending over a large cone, the transmission coefficient is different for each Fourier component and falls to zero at the total reflection angle. Thus, the real output can be considerably less than what the plane wave theory predicts. Finally, the plane wave theory often ignores the reduction in output power due to double refraction which can be significant for small spot sizes in crystalline media.

Improvement in the calculations of far-infrared generation by optical mixing has been achieved by Faries et al.\(^{26}\) using the far-field diffraction theory for a distribution of oscillating dipoles induced.
by the pump beams. They used an average transmission coefficient for the far-infrared output across the boundary and excluded the contribution from the totally reflected modes. The effect of double refraction was, however, ignored. As we shall see later, in the absence of double refraction, this approach in fact gives a remarkably good estimate of the far-infrared output.

In this paper, we present a more rigorous calculation of far-infrared generation by optical mixing. It proceeds by first calculating separately each Fourier component of the output field and then evaluating the output power by summing over the Fourier components. The effects of focusing, absorption, phase matching, and double refraction can all be properly taken into account. For the sake of simplicity, the pump beams are assumed to be of single mode with Gaussian profiles. Our approach is essentially the same as that used by Bjorkholm and by Kleinman et al. for second-harmonic generation by focused beams.

The main difference between second-harmonic (or sum-frequency) generation in the visible or near infrared and difference-frequency generation in the far-infrared is diffraction. Validity of the scalar Fresnel approximation for the pump beams guarantees its validity for the sum frequency but not for the difference frequency. Because of its much longer wavelength and hence stronger diffraction, the far-infrared output extends over a much broader cone. Thus, the phase matching condition varies much more appreciably among the output Fourier components in difference-frequency generation (DFG) than in
sum-frequency generation (SFG). All the Fourier components can often be nearly simultaneously phase matched for SFG but not for DFG. An accurate description of DFG also requires knowledge of the difference-frequency transmission coefficients over a very broad output cone.

The body of the paper is organized into the following sections: Section II describes the theory of DFG by monochromatic Gaussian laser beams which is valid even when the pump focal spot size is smaller than a far-infrared wavelength. This theory is developed from a generalization of the non-linear polarization used by Boyd and Kleinman\textsuperscript{31} and by Faries.\textsuperscript{28} Section III contains the results of numerical calculations obtained from this theory. First, we present the results for the ideal case of no double refraction. Then, we discuss briefly the reductions in attainable power due to far-infrared absorption and double refraction. Finally, in Section IV, we compare our results with the results of three other calculations: a simple plane wave calculation, a far-field diffraction calculation assuming a constant 1/e radius Gaussian distribution of induced dipoles, and the second harmonic generation calculations of Boyd and Kleinman.\textsuperscript{31}

II. Theory

A. Nonlinear Polarization

We assume that the pump beams are monochromatic with Gaussian TEM\textsubscript{00} mode. If focusing and diffraction of the pump beams are not too strong, the focused pump fields in a slab medium can be written as\textsuperscript{28,31}
\[ \tilde{E}_i(r,t) = \frac{\tilde{E}_i}{1 + i\xi_i} \exp \left[ - \frac{(x-a_i - \xi_i z)^2 + y^2}{w_i^2(1 + i\xi_i)} + ik_iz - \omega_it \right] \] 

(17)

for \( 0 \leq z \leq \ell \), where the subindex \( i \) denotes the \( i \)-th beam; \( w_i \) is the \( e^{-2} \) beam radius in the focal plane which is located at \( z = z_{0i} \); the beam axis intersects the front surface of the medium at \( x = a_i \) and \( y = 0 \); the quantity \( \xi_i \) is defined by \( \xi_i = \frac{2(z-z_{0i})}{k_iw_i^2} \) with \( k_i = \omega_i n_i/c \), \( n_i \) being the refractive index; finally, \( \zeta_i \) is the walk-off angle given by \( \zeta_i = \frac{1}{2} \sin(2\theta_i) \) \( n_i^2(n_{em,i}^{-2} - n_{o,i}^{-2}) \) if the beam is an extraordinary ray propagating in a uniaxial medium along a direction at an angle \( \theta \) with respect to the optical axis where \( n_{o,i} \) and \( n_{em,i} \) are respectively the ordinary and extraordinary refractive indices at \( \theta = 90^\circ \). The derivation of Eq. (17) involves some approximations which can easily be justified as shown in Appendix A. In the following, to simplify the calculations in practical cases, we can assume that the largely overlapping pump beams are focused to the same spot size at the same point with \( w_i = w, \xi_i = \xi \) and \( z_{0i} = z_0 \). This is a good approximation when the refractive indices of the pump beams are not very different, as is true in all practical cases which have been investigated.

The pump fields now induce a nonlinear polarization at the far-infrared frequency in the medium. We consider here only the case of DFG in a uniaxial crystal as an example although the formalism can be easily extended to more general cases of optical mixing. The nonlinear polarization at the difference frequency \( \omega \) is then given by
\[ P^{(2)}(\mathbf{r},\omega) = \chi^{(2)}(\omega = \omega_1 - \omega_2): \mathbf{E}_1(\mathbf{r},\omega_1) \mathbf{E}_2^*(\mathbf{r},\omega_2) \]  

where \( \chi^{(2)} \) is the second-order nonlinear susceptibility tensor. We assume that \( \mathbf{E}_1 \) is an ordinary ray and \( \mathbf{E}_2 \) is extraordinary. The nonlinear polarization \( \mathbf{P}^{(2)}(\mathbf{r}) \) can be readily found by substituting the expression of \( \mathbf{E}_1 \) of Eq. (17) into Eq. (18). For convenience of solving the wave equation later, we are however interested in the transverse Fourier components of \( \mathbf{P}^{(2)}(\mathbf{r}) \). The transverse Fourier transform gives

\[ P^{(2)}(k_T, z) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} P^{(2)}(\mathbf{r}) \exp(-ik_x x - ik_y y) \]

\[ = \chi^{(2)}: \mathbf{E}_1^x \mathbf{E}_2^* \frac{1}{4} w^2 \exp\{i[(k_1 - k_2^x) z - k_x (a_1 - \zeta z)/2]\} \]

\[ \times \exp\left[-\frac{(a_1 - \zeta z)^2}{2w^2} - \frac{1}{8} k_T^2 w^2 (1+\zeta^2) - \frac{1}{2} k_x (a_1 - \zeta z) \zeta \right] \]

where \( \mathbf{k}_T = \mathbf{\hat{x}} k_x + \mathbf{\hat{y}} k_y \) and we set \( a_2 = 0 \). For economy of notation, we omit explicit mention of the argument \( \omega \).

B. Solution of Wave Equations

Far-infrared generation by optical mixing is described by the set of wave equations

\[ [\nabla \times (\nabla \times - (\omega^2/c^2)\varepsilon^*)] \mathbf{E}(\mathbf{r},\omega) = 4\pi(\omega^2/c^2) P^{NL}(\mathbf{r},\omega) \]  

\[ \nabla \cdot [\varepsilon^* \mathbf{E}(\mathbf{r},\omega)] = -4\pi \nabla \cdot \mathbf{P}^{NL}(\mathbf{r},\omega) \]  

(20a)

(20b)
where the nonlinear polarization \( P_{NL} \) acts as a driving source for the nonlinear process. For DFG in a uniaxial medium \( P_{NL} = P^{(2)} \) given by Eq. (18). Since the normal to slab boundary planes is \( \hat{z} \), the easiest method is to Fourier transform the \( x \) and \( y \) variables in Eq. (20) and to solve for each Fourier component \( \hat{E}(\hat{\mathbf{k}},z) \) separately. The corresponding source term for \( \hat{E}(\hat{\mathbf{k}},z) \) is \( 4\pi(\omega^2/c^2) \hat{P}^{(2)}(\hat{\mathbf{k}},z) \) with \( \hat{P}^{(2)}(\hat{\mathbf{k}},z) \) given by Eq. (19).

The general solution for \( \hat{E}(\hat{\mathbf{k}},z) \) consists of two parts, the homogeneous solution and the particular solution respectively. The homogeneous solution is well known. For ordinary and extraordinary polarizations respectively, it can be written as

\[
\hat{E}_{o\pm}^{h}(\hat{\mathbf{k}},z) = \hat{\mathbf{e}}_{o\pm} \exp(ik_{o\pm}z) \\
\hat{E}_{e\pm}^{h}(\hat{\mathbf{k}},z) = \hat{\mathbf{e}}_{e\pm} \exp(ik_{e\pm}z) \tag{21}
\]

where the subindices \( \pm \) denote forward and backward propagating waves respectively with the same \( \hat{\mathbf{k}}_T \), and \( k_{o\pm},z = \pm[\omega n_o/c^2 - k_T^2]^{1/2} \) with a similar expression for \( k_{e\pm},z \). To find the particular solution, let us first assume that the nonlinear slab is imbedded in a linear medium with an equal linear dielectric constant. Thus, reflection and refraction at the crystal boundaries can be ignored. The boundary effects will be taken into account later. As shown in Appendix B, the particular solution for \( E(\mathbf{k},z) \) is then given by
\[ \hat{E}^{p}(k_T, z) = E_{o+}^{p}(k_T, z) \hat{e}_+ + E_{o-}^{p}(k_T, z) \hat{e}_- + E_{e+}^{p}(k_T, z) \hat{e}_+^{e+} + E_{e-}^{p}(k_T, z) \hat{e}_-^{e-} \]

\[ + E_{e-}^{p}(k_T, z) \hat{e}_- - \left(4\pi n^2 e / n_o^2 n_{em}^2 \right) z P_z^{(2)}(k_T, z) \]

where \( n_{em} \) is the refractive index for extraordinary ray propagating perpendicular to the optical axis,

\[ E_{o-}^{p}(k_T, z) = \frac{2\pi i \omega^2}{c^2 k_o z} \int_0^z \hat{e}_+ \cdot \hat{E}^{(2)}(k_T, z') e^{-ik_{o z}(z-z')} dz' \]

\[ E_{o-}^{p}(k_T, z) = \frac{2\pi i \omega^2}{c^2 k_o z} \int_z^\ell \hat{e}_- \cdot \hat{E}^{(2)}(k_T, z') e^{-ik_{o z}(z-z')} dz' \]

\[ E_{e+}^{p}(k_T, z) = \frac{2\pi i \omega^2}{c^2 (k_{ez})_{eff}} \int_0^{z} \hat{e}_+ \cdot \hat{E}^{(2)}(k_T, z') e^{ik_{e+}(z-z')} dz' \]

\[ E_{e-}^{p}(k_T, z) = \frac{2\pi i \omega^2}{c^2 (k_{ez})_{eff}} \int_z^{\ell} \hat{e}_- \cdot \hat{E}^{(2)}(k_T, z') e^{ik_{e-}(z-z')} dz' \]

\[ (k_{ez})_{eff, +} = (k_{e+}, z - k_{e-}, z) n_{em}^2 / \left\{ 2n_e^2 \left[ 1 - \left( \frac{n_o^2 - n_{em}^2}{n_o^2} \right) \frac{(\hat{c} e^e)^2}{k_o^2} \right] \right\} \]

\( \hat{c} \) is the optical axis of the crystal. The last term in Eq. (22) is a longitudinal field which leads to optical rectification when \( \omega = \omega_1 - \omega_2 = 0 \). It is, however, a non-radiating term and we shall neglect it in the following discussion.
The solution in Eqs. (22) and (23) appear in the form normally obtained for nonlinear optical processes in the slowly varying envelope approximation. However, no such approximation has been made. As shown in Appendix B, Eq. (22) together with Eq. (23) is an exact solution of Eq. (20) with \( \hat{P}^{(2)}(k_T,z) \) as the source term. The field \( \hat{E}^{p}(k_T,z) \) in the medium does not have a slowly varying amplitude since \( \frac{\partial^2 |\hat{E}^{p}(k_T,z)|}{\partial z^2} \) is not negligible in comparison with \( 2k_3 |\hat{E}^{p}(k_T,z)|/\partial z \). In fact, the slowly varying envelope approximation is equivalent to assuming for each polarization a wave propagating in one direction only.

As a check, we can use Eqs. (22) and (23) to derive the solution for the special case of optical mixing at an infinite boundary surface discussed by Bloembergen. We have \( k_y = 0, \ell \to \infty \), and \( \hat{P}^{(2)}(k_T,z) = p_o \hat{y} e^{ik_z z} \) in the medium. Equation (23) gives for the reflected output

\[
\hat{E}^{p}_r = -\frac{2\pi \omega^2}{c^2 k_0 (k_0 + k_s z)} \hat{y} p_o e^{-ik_0 z} \quad \text{for } z < 0 \quad (24a)
\]

and for the transmitted output

\[
\hat{E}^{p}_t = \frac{2\pi \omega^2}{c^2 k_0} \hat{y} p_o \left[ \frac{2k_0}{k_0 - k_s} e^{ik_z z} - \frac{1}{k_0 - k_s} e^{ik_0 z} \right] \quad \text{for } z > 0 \quad (24b)
\]

The above solution is, however, only true for the case with no reflection at the boundary, but the boundary effects can be easily incorporated by taking into account the linear reflection of \( \hat{E}^{p}_r \) at the boundary surface. The complete solution for the problem with a crystal-vacuum plane boundary is then given by
where \( k_z = [(\omega/c)^2 - k_x^2]^{1/2} \). Substitution of the expressions for \( \vec{E}_P^r \) and \( \vec{E}_P^t \) in Eq. (24) into Eq. (25) yields results identical to those derived by Bloembergen.\(^{34}\)

The above example suggests that the boundary effects can indeed be taken care of separately. In Sec. IIC, we shall use the same procedure to take into account the boundary conditions of optical mixing in a slab medium. Then, with the expression of \( \vec{P}(2)(\kappa_T, z) \) in Eq. (19), we can calculate from Eqs. (21) – (23) and the appropriate boundary conditions the Fourier component \( \vec{E}(\kappa_T, z) \) of the DFG output and hence the difference-frequency field \( \vec{E}(r) \) in space. In many cases, only one of the four waves in Eq. (23) is nearly phase-matched. When this happens, we need to retain only the phase-matched component in a good approximate calculation.

C. Boundary Effects

We have seen in Sec. IIB how we can take into account the boundary effects of a crystal-vacuum interface by simply incorporating linear reflection and transmission of the waves at the boundary into the solution. We now discuss the boundary effects of the more general case of a slab crystalline medium. We can consider \( \vec{E}_P^t \) in Eq. (22) as forward propagating waves starting from \( z = 0 \) in the medium and
subsequently undergoing multiple partial reflections at the two slab surfaces. Similarly, we consider \( E_\pm \) in Eq. (22) as backward propagating waves starting from \( z = L \) in the medium. Thus, the field outside the slab is given by the sum of \( E_+ \) and \( E_- \) weighted respectively by appropriate Fabry-Perot factors due to multiple reflections and transmissions. To find the Fabry-Perot factors, we first calculate the transmission and reflection matrices for ordinary and extraordinary waves at a single crystal-vacuum boundary surface, and then find the overall transmission and reflection matrices of the slab for the two waves by summing over multiple transmissions and reflections at the slab surfaces.

Consider first the case defined in Fig. 1a. The incident monochromatic plane wave \( \hat{E}_{i+} (k_T) = E_{i+}^\| \hat{\mathbf{n}} + E_{i+}^\perp \hat{\mathbf{e}} \) and the reflected plane wave \( \hat{E}_{r-} (k_T) = E_{r-}^\| \hat{\mathbf{n}} + E_{r-}^\perp \hat{\mathbf{e}} \) are related to the refracted ordinary and extraordinary waves \( \hat{E}_{o+} (k_T) \) and \( \hat{E}_{e+} (k_T) \) respectively, by the matrix relations 35

\[
\begin{pmatrix}
E_{i+}^\| \\
E_{i+}^\perp
\end{pmatrix} = A_+ \begin{pmatrix}
E_{o+} \\
E_{e+}
\end{pmatrix}
\]

\[
\begin{pmatrix}
E_{r-}^\| \\
E_{r-}^\perp
\end{pmatrix} = B_+ \begin{pmatrix}
E_{o+} \\
E_{e+}
\end{pmatrix}
\] (26)
where

\[ A_± ≡ \left( \frac{α_o^±}{1+\frac{1}{r_0^l}}, \frac{α_e^±}{1+\frac{1}{r_e^±}} \right), \quad B_± ≡ \left( \frac{β_o^± k_o}{1+\frac{1}{r_o^l}}, \frac{β_e^± k_e}{1+\frac{1}{r_e^±}} \right) \]

\[ r_o^l ≡ (k_z - k_{0z})/(k_z + k_{0z}), \quad r_o^l ≡ (k_o^2 - k_{0z}^2)/(k_o^2 + k_{0z}^2) \]

\[ r_e^± ≡ \frac{(-k_{ez} ± k_z)}{(k_{ez} ± k_z)}, \quad r_e^± ≡ \frac{(-k e^± k_z e^± k_{ez} e^±)}{(k e^± k_z e^± k_{ez} e^±)} \]

\[ α_o^± ≡ \hat{o}_± \cdot (\hat{z} \times \hat{k}_T), \quad β_o^± ≡ \hat{o}_± \cdot \left[ (\hat{z} \times \hat{k}_T) \times \hat{k}_o^± \right] \]

\[ α_e^± ≡ \hat{e}_± \cdot (\hat{z} \times \hat{k}_T), \quad β_e^± ≡ \hat{e}_± \cdot \left[ (\hat{z} \times \hat{k}_T) \times \hat{k}_e^± \right] \]

\[ γ_e ≡ \hat{e}_± \cdot \hat{k}_T. \]

With subindex "-" applied to the case of Fig. 1b with

\[ \left( \begin{array}{c} E_{1-}^l \\ E_{1-}^\| \end{array} \right) = A_- \left( \begin{array}{c} E_o^- \\ E_e^- \end{array} \right), \quad \left( \begin{array}{c} E_{r+}^l \\ E_{r+}^\| \end{array} \right) = B_- \left( \begin{array}{c} E_o^- \\ E_e^- \end{array} \right) \]

We next consider transmission and reflection of ordinary and extraordinary waves incident from the crystal side onto the boundary surface as described by Diagram a in Fig. 2. Clearly, Diagram a is equivalent
to the sum of Diagram b and Diagram c, and Diagrams b and c are identical to those in Fig. 1a and Fig. 1c respectively. We therefore have

\[ \hat{E}_{t-} = \hat{E}^t_{t-} + \hat{E}''_{t-} = \hat{T}_t \begin{pmatrix} E_{o-} \\ E_{e-} \end{pmatrix} \]

\[
\begin{pmatrix}
E_{o+} \\
E_{e+}
\end{pmatrix} = \hat{R}_p \begin{pmatrix} E_{o-} \\ E_{e-} \end{pmatrix}
\]

(27)

where

\[ \hat{T}_t = A_t - B_t A_t^{-1} B_t \]

\[ \hat{R}_p = -A_t^{-1} B_t \]

the subindices "+" and "-" now refer to cases where the crystalline medium occupy the left half-space and the right half-space, respectively.

We can now use the results in Eq. (27) to calculate the effect of multiple transmissions and reflections at the boundaries of a crystal slab. In particular, we are interested in finding the forward and backward propagating far-infrared waves outside the slab created by optical mixing inside the slab. As we mentioned earlier, we can imagine that optical mixing generates waves \( \hat{E}_+^p \) starting at \( z = 0 \) and \( \hat{E}_-^p \) starting at \( z = l \) and in getting out of the slab, these waves undergo multiple transmissions and reflections. Therefore, for the generated field outside the slab, we readily find for \( z = l \)
and for \( z < 0 \)

\[
\hat{E}_{T-} = \hat{T}_{-} F_{-} \left[ \begin{array}{c} E_{o-}(z) \\ E_{e-}(z) \end{array} \right] + \hat{P}_{-} R_{+} \left[ \begin{array}{c} E_{o+}(z) \\ E_{e+}(z) \end{array} \right]
\]  

(28b)

where

\[
\hat{p}_{\pm} = \begin{pmatrix} \exp(\pm ik_{oz}) \\ \exp(\mp ik_{ez}) \end{pmatrix}
\]

and

\[
\hat{F}_{\pm} = \left[ 1 - \hat{P}_{\pm} R_{\mp} \hat{P}_{\mp} \right]^{-1} = \sum_{n=0}^{\infty} \left[ \hat{P}_{\pm} R_{\mp} \hat{P}_{\mp} \right]^{n}
\]  

(29)

Because of the generalized Fabry-Perot factor \( \hat{F}_{\pm} \), the output fields \( \hat{E}_{T\pm} \) can be rapidly varying functions of \( k_{T}, \omega, \) and \( \ell \). In some cases, however, when the pump laser beams have fairly broad linewidths or the crystal slab is wedged or not sufficiently well polished, it is more appropriate to find an average Fabry-Perot factor or the average output by averaging over one Fabry-Perot period. For example, in the nearly isotropic case, we find from Eq. (28) after some manipulation,
where $\gamma$ is the attenuation constant along $\hat{z}$.

D. Far-Infrared Output Power and Its Far-Field Angular Distribution.

The total far-infrared power outputs from the slab in the forward and backward directions are

$$P_{\pm} = \frac{c}{2\pi} \int dx dy (\hat{z} \cdot \hat{n}) |\vec{E}_{T\pm} (x,y)|^2$$

evaluated at large $z$. By Parseval's Theorem, this can be written as

$$P_{\pm} = \frac{c}{2\pi} \int \frac{dk_x dk_y (\hat{z} \cdot \hat{k})}{k_x^2 + k_y^2} \frac{dk_z}{k_z} |\vec{E}_{T\pm} (\hat{k})|^2$$

where $\vec{E}_{T\pm} (\hat{k})$ is given by Eq. (28).

In most practical cases, we are also interested in the far-field angular distribution of the output power. As shown in the Appendix of Miyamoto and Wolf,\textsuperscript{36} it has the expression

$$\frac{dP_{\pm} (\theta, \phi)}{d\Omega} = \frac{c}{2\pi} \frac{\omega^2}{c^2} \cos^2 \theta |\vec{E}_{T\pm} (\hat{k} T) = \frac{\omega}{c} \sin \theta (\hat{x} \cos \phi \hat{y} \sin \phi)|^2$$
III. Results of Numerical Calculations

In this section, we shall present numerical calculations of far-infrared generation by difference-frequency mixing using the equations given in the previous section. We choose somewhat arbitrarily the following values for the characteristic parameters of the nonlinear crystal: \( n_o = 2, n(\omega) = 4, \) and \( \chi^{(2)} = 1.87 \times 10^{-6} \) esu. The two nearly overlapped pump beams, one ordinary and one extraordinary, are assumed to have the same focal spot in the crystal with both beams always along the normal to the slab. The question we propose to answer is how various quantities such as phase mismatch, focusing, beam walkoff, and absorption affect the far-infrared output at different frequencies.

A. Far-infrared Generation in the Absence of Absorption and Optical Walkoff

We assume in this case that the optical axis of the crystal is in the plane of the slab along \( \hat{x} \). The two pump beams, one ordinary and one extraordinary, propagate along the normal to the slab, \( \hat{z} \), with essentially no walkoff, the nonlinear polarization \( \vec{p}_{NL} \) is along \( \hat{y} \), and the common focal spot of the two pump beams is at the center of the slab. We also assume that the extraordinary refractive index \( n_{em} \) of the pump beam can be varied by external means such as temperature in order to adjust the amount of phase mismatch in DFG and that only the ordinary far infrared waves in the forward direction can be nearly phase matched. Since the phase mismatch is different for different Fourier components \( \vec{E}(\vec{k}_\omega) \) of the far-infrared output, we define an axial phase mismatch \( \Delta k_a = k_{1}(\omega_1) - k_{2}(\omega_2) - k_{0}(\omega) \) to describe the overall phase matching condition.
Figure 3 shows the far-field angular distribution of the far-infrared output, $dP(0)/d\Omega$ versus $\theta$, at $100 \text{ cm}^{-1}$ calculated from Eq. (33). In the calculation the slab has a thickness of 1 cm, the focal spot size is $w = 25 \mu m$, and the axial phase mismatch corresponds to $\Delta k_a = -5.1 \text{ cm}^{-1}$. Since the far-infrared output is approximately symmetric about $z$ (i.e., nearly independent of the azimuth angle $\phi = \tan^{-1}(k_y/k_x)$), Fig. 3 actually shows a distribution in the form of a hollow cone.

The radiation peaks at the angle $\theta_m = \sin^{-1}\left\{n_o^2(\omega) - \left[n_o(\omega) + \Delta k_a c/\omega\right]^2\right\}^{1/2}$ at which phase matching $\Delta k_z = k_1 - k_2 - k_{oz}(\omega) = 0$ occurs. The secondary maxima of the phase-matching curve can also be seen. They become more pronounced for shorter far-infrared wavelengths as the effect of diffraction becomes less important. From the expression of $\theta_m$, it is seen that if $\Delta k_a = 0$, then $\theta_m = 0$ and the far-infrared output appears as a narrow solid cone along the $z$ axis. If $\Delta k_a > 0$, then there is no solution for $\theta_m$ and the far-infrared output is strongly suppressed by phase mismatch; the angular distribution may show a weak central peak at $\theta = 0$ and some secondary maxima at finite $\theta$. For negative $\Delta k_a$, the phase-matched peak shifts to larger $\theta_m$ until $\theta_m = \pi/2$; then because of total reflection at the surface, the far-infrared radiation in the phase-matched direction can no longer get out of the slab and the output peak at $\theta = \pi/2$ drops quickly.

The total far-infrared power output $P$ versus $\Delta k_a$ is shown in Fig. 4 with the same set of parameters used for Fig. 3. The curve has a maximum around $\Delta k_a = -5.1 \text{ cm}^{-1}$ corresponding to the full development of the hollow phase-matched cone in Fig. 3. The steep rise of the
curve at $\Delta k_a \simeq 0 \text{ cm}^{-1}$ is due to the initial appearance of the phase-matched cone. The gradual decrease between $\Delta k_z \approx -10 \text{ cm}^{-1}$ and $-75 \text{ cm}^{-1}$ is due to the combined effects of decrease of the far-infrared transmission coefficients and decrease of the effective $p^{(2)}$ for the generation of ordinary far-infrared waves around the phase-matched direction. The steep drop after $\Delta k_a \approx -75 \text{ cm}^{-1}$ is due to total reflection of an increasing portion of those far-infrared waves generated near phase matching.

If the far-infrared wavelength $\lambda$ inside the crystal becomes much smaller than the focal spot size $w$, the variation of far-infrared output versus phase mismatch $\Delta k_a$ appears more like the usual phase-matching function $\sin^2(x)/x^2$ for the ideal plane wave case. An example is shown in Fig. 5 for the case of $\lambda = w/8$. Because of the smaller $\lambda/w$ ratio, the off-axis Fourier components of the far-infrared become relatively less important and hence the output drops more rapidly with increase of $\Delta k_a$. The curve in Fig. 5 is, however, still noticeably asymmetric and its peak occurs at $\Delta k_a = -2 \text{ cm}^{-1}$ rather than $\Delta k_a = 0$. As the ratio of $\lambda/w$ decreases further, the effect of far-infrared diffraction becomes even smaller; the phase-matching curve $P$ versus $\Delta k_a$ then develops more clearly defined secondary peaks and approaches the symmetric form $\sin^2(\Delta k_a \lambda/2)/(\Delta k_a \lambda/2)^2$.

The focusing geometry of the Gaussian pump beams is completely characterized by the focal spot size $w$. In order to see how the far-infrared output varies with focusing, we calculate the $\zeta = 0$ curve in Fig. 6 which shows the maximum of $P(\Delta k_a)$ as a function of $w$. Because
of the higher pump intensity resulting from tighter focusing, the far-infrared output increases sharply with decrease of \( w \). It, however, reaches a maximum at \( w = 13 \mu m \) as the corresponding reduction of the longitudinal focal dimension takes its toll. It is interesting to note that in the model of collimated Gaussian pump beams with a radius \( w \) and with \( w^2 |E_1 E_2| = \) constant, \( P \) versus \( w \) has no maximum. This is because when \( k_T w < 1 \) for all significant far-infrared Fourier components, \( P^{(2)}(k_T, z) \) in Eq. (19) becomes independent of \( k_T \) and \( w \).

While Figs. 3-6 are for \( w = 100 \text{ cm}^{-1} \), Figs. 7-9 show results of similar calculation for \( w = 10 \text{ cm}^{-1} \). The far-field angular distribution of the output is given in Fig. 7 for two values of the azimuth angle \( \phi = \tan^{-1}(k_y / k_x) = 0 \) and \( \pi/2 \). In this case, because \( \lambda / w = 10 \) is large, far-infrared diffraction is more important; phase matching occurs around \( \theta = \pi/4 \) and the phase-matched peak is very broad. As a result, the output asymmetry with respect to \( \phi \) shows up because at relatively large \( \theta \), the transmission coefficient for the ordinary far-infrared wave across the slab boundaries is different for different \( \phi \). For \( \phi = 0 \), the wave is linearly polarized perpendicular to the plane of incidence, while for \( \phi = \pi/2 \), the wave is linearly polarized in the plane of incidence. The latter case has a Brewster angle at \( \theta = 76^\circ \).

Figure 8 shows the total far-infrared output at \( w = 10 \text{ cm}^{-1} \) as a function of the axial phase mismatch \( \Delta k_a \). The curve again resembles the well-known phase-matching curve \((\sin^2 x) / x^2 \) for the plane wave case except that its maximum is at \( \Delta k_a = -4 \text{ cm}^{-1} \) instead of \( \Delta k_a = 0 \) and it has no well-defined nodes. However, this resemblance does not
occur because diffraction is unimportant. It occurs because, when the far-infrared wavelength is sufficiently long, then all the far-infrared Fourier components $\tilde{E}(k_T, z)$ have roughly the same $\Delta k_z \approx \Delta k_a / \lambda$; in other words, if $\Delta k_a = 0$, then all the far-infrared Fourier components are nearly phase matched. The small difference of $\Delta k_z / \lambda$ among the Fourier components, however, broadens the phase-matching peak and obscures the fine structure.

The $\zeta = 0$ curve in Fig. 9 describes the peak value of $P(\Delta k_a)$ at $\omega = 10 \text{ cm}^{-1}$ as a function of the focal spot size $w$. We notice that in the range of our calculation, this maximum output power $P_{\text{max}}(\Delta k_a)$ always increases with decrease of $w$. In this case, $k_T w$ becomes so much smaller than 1 at small $w$ that the nonlinear polarization $\tilde{P}^{(2)}(k_T)$ approaches a constant independent of $k_T$, $w$, and $z$ in spite of the factor $(1+\zeta^2)$ in the exponential function in Eq. (19).

Consequently, the $\zeta = 0$ curve of Fig. 9 flattens out at small $w$. Eventually, for even smaller $w$, we should expect the curve to go through a maximum like the $\zeta = 0$ curve in Fig. 6 for $\omega = 100 \text{ cm}^{-1}$.

B. Far-Infrared Generation with a Finite Walkoff Angle Between the Pump Beams

We now consider the effect of optical walkoff on far-infrared generation. We still assume that the pump beams propagate normal to the slab and absorption is negligible, but the orientation of the optical $\hat{c}$ axis of the crystal is now varied in the $\hat{x}-\hat{z}$ plane in order to vary the walkoff angle $\zeta$. The primary effect of optical walkoff is that it limits the effective interaction length of the beams. When $\zeta$ is much larger than the divergence angle of the pump beams, the two
pump beams overlap in the focal region only over a distance of $2w/|\zeta|$; most of the far-infrared radiation is generated from this overlapping region. As $|\zeta|$ increases, the effective interaction length decreases, and hence the phase-matching peak in the far-field angular distribution becomes weaker and broader as shown in Fig. 10 for $w = 10 \text{ cm}^{-1}$. For smaller focal spot sizes $w$, the walkoff effect is stronger. This gives rise to a lower maximum at a larger $w$ for the $\zeta \neq 0$ curves in Figs. 6 and 9.

The far-infrared output should in general consist of both ordinary and extraordinary waves. We have so far assumed that the e-wave is strongly phase-mismatched and can be neglected. This is true for $\theta_c = \cos^{-1}(\hat{\varepsilon} \cdot \hat{z})$ larger than the total reflection angle $\theta_R$. However, when $\theta$ approaches $\hat{\varepsilon}$ or $\theta_c$ approaches zero, the phase mismatch of the e-wave is greatly reduced and the e-wave output becomes non-negligible. For $\theta \approx 0$, we have the nearly degenerate case where the e-wave and the o-wave contribute almost equally to the far-infrared output.

There are two other less important effects of optical walkoff on far-infrared generation. First, the $\exp(ik_x \zeta z/2)$ term in Eq. (19) contributes to the phase matching relation which now becomes

$$\Delta k_z = k_1 - k_2 + k_x \zeta - k_z = 0.$$  

This term shifts the center of the phase-matching cone in Figs. 3, 7, and 10 from $k_x = 0$ to $k_x = \zeta (\Delta k_a + n_o \omega/c)/2$. Since for $\phi = 0$, the far-infrared transmission coefficient for o-waves at the boundary falls off monotonically with increase of $\theta$, this increases the phase-matched output for $k_x > 0$ and decreases that for $k_x < 0$. Second, as seen from Eq. (19), the maximum of $|\tilde{P}_n^{(2)}(k_T^*, z)|$ is
shifted from \( k_x = k_y = 0 \) to \( k_x = k_1 \frac{\zeta z}{(1+\zeta^2)} \) and \( k_y = 0 \); its effect on the far-field angular distribution is just the opposite of that due to the shift of the phase matching cone. Depending on the situation, one effect may dominate over the other. They are responsible for the slight asymmetry of the \( \zeta = 0 \) curves in Fig. 10. The phase-matching effect is more important for the \( \zeta = -0.01 \) case while the \( |p^{(2)}(k_T)| \) effect is more important for the \( \zeta = -0.02 \) case. For shorter crystals (\( \ell \lesssim 0.5 \) cm), the phase-matching effect is more important.

C. Effects of Linear Absorption on Far-Infrared Generation

In practice, nonlinear far-infrared generation in crystals is always limited by far-infrared absorption. This is the main reason why far-infrared DFG in solids has in most cases been restricted to the range between 1 and 200 cm\(^{-1}\). Roughly speaking, with an absorption coefficient \( \gamma \), the effective length of the crystal for DFG cannot be much more than \( 2/\gamma \).

Figure 11 shows how the far-infrared output from a 1-cm slab decreases as a function of the far-infrared absorption coefficient \( \gamma \) for \( \omega = 10 \) and 100 cm\(^{-1}\). In the calculation, the focal spot size was chosen as \( w = 25 \) \( \mu \text{m} \) and the location of the focal spot was at the center of the slab for \( \gamma = 0 \), while for increasing \( \gamma \) it moves towards the end surface of the slab. As we mentioned earlier in Sec. IIIA, for \( \omega = 10 \) cm\(^{-1}\), all the significant far-infrared Fourier components are nearly phase-matched (\( \Delta k_z \ell < \pi \)). Therefore, the curve for \( \omega = 10 \) cm\(^{-1}\) in Fig. 11 agrees fairly well with that described by \( [1-\exp(-\gamma\ell/2)]^2/(\gamma\ell/2)^2 \) for the phase-matched plane wave case. For
ω = 100 cm⁻¹, since not all the significant far-infrared Fourier components can be nearly phase-matched, the reduction of far-infrared output with increasing absorption is slower and cannot be approximated by the phase matched plane-wave form at small γ.

In some respects, the effects of γ for γl ≳ 2 can be simulated by an absorptionless crystal with a length 2γ⁻¹. An increase of γ increases the phase-matching angle and broadens the phase-matched peak in the angular distribution of the far-infrared output. It also makes (Δkₐ)opt, the optimum axial phase mismatch for maximum total far-infrared output, more negative. This latter effect is quite pronounced for ω = 100 cm⁻¹ as shown in Fig. 12.

IV. Comparison with Other Models and with Calculations of Second Harmonic Generation

We now compare the results of our detailed calculations with those obtained from two simple models for the case where the optical walk-off effect is negligible. One is the Gaussian distribution (GD) model in which we assume a Gaussian profile for the nonlinear polarization at the difference frequency.

\[ \hat{e}^{(2)}(r,t) = (\chi^{(2)}, \hat{e}_1, \hat{e}_2) \exp[-2(\chi^2 + y^2)/\omega^2 + i(k_1 - k_2)z - i\omega t] \]  

in the crystal slab where the pump fields are given by

\[ \hat{e}_j(r,t) = \hat{x}_j \exp[-(\chi^2 + y^2)/\omega^2 + ik_jz - i\omega_j t], \quad j = 1,2. \]
This is an extension of an earlier model used by Zernike and Berman\textsuperscript{27} and Faries et al.\textsuperscript{26} which assumes a uniform amplitude for $\hat{P}^+(2)(r,t)$ throughout a cylinder with a finite radius. The other simple model is the usual plane wave model in which we assume that the geometric ray approximation is valid and that each beam can be described by a cylindrical pencil of rays with a single wave vector.

From the GD model, we obtain for the lossless case a total output power at $\omega$ of

$$
p_{GD}(\omega) = \frac{2}{c^2} \frac{3}{4} \chi^{(2)} \frac{\hat{P}^+(2)}{2} \frac{\hat{2}^2}{1} \frac{g_2}{2} \\
\times \int_{0}^{\omega/c} \text{d}k_{\text{TR}}(k_{w}/k_{oz}) \left( T(k_{TR}) \right)_{\phi} C(\Delta k_{z}) e^{-\omega^2 k_{\text{TR}}^2 / 4}
$$

where $\left( T(k_{TR}) \right)_{\phi}$ is the far-infrared transmission factor averaged over the azimuthal angle $\phi$ with multiple reflections at the slab boundaries taken into account, and $C(\Delta k_{z})$ describes the effect of phase mismatch. They are given by

$$
\left( T(k_{TR}) \right)_{\phi} = \frac{1}{2(k + k_{oz})} \left[ \frac{k_{oz}^2 (k_{oz} + n_{o} k_{z})^2}{k_{oz} k_{oz}^2 + n_{o}^2 k_{z}^2} + \frac{(k_{oz} + k_{z})^2}{k_{oz} k_{oz}^2 + k_{z}^2} \right]
$$

$$
C(\Delta k_{z}) = \sin^2 \left( \frac{\Delta k_{z} \ell / 2}{\Delta k_{z} \ell / 2} \right) \left( \frac{\Delta k_{z} \ell / 2}{\Delta k_{z} \ell / 2} \right)
$$

with $\Delta k_{z} = n_{o} \omega / c + \Delta k_{a} - k_{oz}$ and $\Delta k_{a} = k_{1} - k_{2} - n_{o} \omega / c$. 

\textsuperscript{27} Zernike and Berman.

\textsuperscript{26} Faries et al.
The output powers at 100 cm\(^{-1}\) and 10 cm\(^{-1}\) calculated from Eq. (35) as a function of \(w\) are shown in Figs. 13 and 14 respectively in comparison with the results of Eq. (32) from our more exact calculations. At 100 cm\(^{-1}\), the only perceptible difference between the two curves occurs at small beam sizes and amounts to 6\% at \(w = 13 \, \mu\text{m}\). At 10 cm\(^{-1}\), the two curves are virtually indistinguishable. Thus, the GD model appears to be a very satisfactory approximation.

The output power from the plane wave model without boundary conditions is given by

\[
P_{\text{PW}}^{\parallel}(\omega) = \frac{\pi^2 \omega^2 w^2}{2 \, \text{cm} \, \text{n}} \left| \chi^{(2)} : \mathcal{E}_{1}^{\parallel} \mathcal{E}_{1}^{\parallel} \right|^2 \mathcal{E}^{2} (T(0)) \phi \tag{37}
\]

The result calculated from Eq. (37) is also shown in Fig. 13. It is 20\% higher than the correct value at \(w = 0.02 \, \text{cm}\). The deviation becomes much worse at smaller \(w\) and diverges as \(w\) approaches zero. This shows that the plane wave model gives unacceptable results at small \(w\) because of its diffractionless approximation. With diffraction, the total far-infrared output power is decreased by total reflection of those Fourier components with large \(k_{z}\) and by phase mismatch (described by \(G(\Delta k_{z})\) in Eq. (35)) for other Fourier components.

The plane wave calculation is, however, simple and does not require numerical integration. It is therefore preferred when one wants to crudely estimate the output power. We can make the estimate more exact by multiplying the calculated result by a correction factor. Comparison of Eq. (35) and (37) shows that this correction factor is given by
\[
F = \frac{P^CD}{P^PW} = \frac{k_0 w^2}{2(T(0))} \int_0^{\omega/c} \frac{\Delta k_z}{k_{oz}} \langle T(k_T) \rangle k_0 \exp\left(-\frac{2w^2k_T^2}{4}\right).
\]

We approximate \(\langle T(k_T) \rangle k_0 \exp\left(-\frac{2w^2k_T^2}{4}\right)\) in the integral by \(\langle T(0) \rangle k_0 \exp\left(-\frac{2w^2k_M^2}{4}\right)\) for \(k_T < k_M\) and by 0 for \(k_T > k_M\), where \(k_M = \sqrt{2\Delta k_c (n_0 \omega/c - \Delta k_c/2)}\) with \(\Delta k_c\) being the smaller of the two quantities \(2\pi/\lambda\) and \((n_0 - \sqrt{n_0^2 - 1})\omega/c\).

Physically, at \(k_T = k_M\), dP/d\omega either has dropped to half of its peak value or has been cutoff by total reflection. The correction factor then becomes

\[
F = 1 - \exp\left(-\frac{2w^2k_M^2}{4}\right).
\]

The output power calculated from \(FP^PW\) using Eqs. (37) and (39) is within 20% of the correct value.

We now discuss similarities and differences between difference-frequency generation (DFG) and second-harmonic generation (SHG). In both cases, each pump field \(\vec{E}_i\) with finite beam radius has a distribution of Fourier components with wave vectors spreading effectively over an angle \(2\delta_i\). The output of DFG or SHG from a nonlinear slab is significant only when part of these significant Fourier components within the angular spread \(2\delta_i\) can satisfy the axial phase matching condition \(\Delta k_z = 0\). As shown in Fig. 15, this happens for SHG only if \(\Delta k_S^a \equiv 2k(\omega_1) - k(2\omega_1) > 0\) and \(\Delta k_S^g \equiv 2k(\omega_1)(1-\cos\delta_1) > \Delta k_S^a\), and for DFG only if \(\Delta k_D^a \equiv k_1 - k_2 - k(\omega) < 0\) and \(\Delta k_D^a \equiv k(\omega)(1-\cos\delta) > -\Delta k_D^a\), where \(2\delta\) is the angular spread of the significant far-infrared Fourier components which can get out of the crystal slab. We emphasize that for an efficient nonlinear interaction we must have \(\Delta k_S^a < 0\) for SHG and \(\Delta k_D^a > 0\) for DFG.
The quantity $\Delta k_a^S$ in SHG or $\Delta k_a^D$ in DFG governs the qualitative behavior of the phase-matching curve $P$ versus $\Delta k_a$. As we mentioned before, the output is most efficient when $\Delta k_a^S$ (or $-\Delta k_a^D$) falls in the range between $0$ and $\Delta k_a^S$ (or $\Delta k_a^D$). Therefore, if $\Delta k_a^S$ (or $\Delta k_a^D$) $\gg 2\pi$, then the phase-matching curve has a broad peak; it rises sharply to the peak around $\Delta k_a^S$ (or $\Delta k_a^D$) = $0$, then slopes downward as $\Delta k_a^S$ (or $-\Delta k_a^D$) increases from $0$ to $\Delta k_a^S$ (or $\Delta k_a^D$), and finally in the case of DFG falls rapidly at a certain $\Delta k_a^D$ value because of the cutoff due to total reflection at the boundaries. Examples are shown in Fig. 4 for DFG with $\Delta k_a^D = 80$ and in Fig. 16 for SHG with $\Delta k_a^S = 100$. Such a phase-matching curve is characteristic of SHG with strong focusing of the pump beam. In DFG, it occurs when the pump beams are more weakly focused because of large far-infrared diffraction. When $\Delta k_a^S$ (or $\Delta k_a^D$) $\leq 2\pi$ the range of $\Delta k_a$ (or $\Delta k_a^D$) for efficient output is much narrower, and the phase-matching curve now shows a central peak and secondary maxima and minima, resembling the well-known plane-wave phase-matching curve described by $\sin^2(\Delta k_a^S/2)/(\Delta k_a^S/2)^2$. Examples are shown in Fig. 5 for DFG at 100 cm$^{-1}$ with $\Delta k_a^D = 4$, in Fig. 9 for DFG at 10 cm$^{-1}$ with $\Delta k_a^D = 8$, and in Fig. 17 for SHG with $\Delta k_a^S = 5.68$. All these curves are, however, slightly asymmetric with a small shoulder on one side. This is because for $\Delta k_a^S < 0$ (or $\Delta k_a^D > 0$), the phase-matching condition $\Delta k_z = 0$ is not satisfied for any of the Fourier components.

There are several minor differences between the SHG and DFG phase matching functions. For $\Delta k_a^S$ (or $\Delta k_a^D$) $\gg 2\pi$, the phase-matching curve
for DFG, as shown in Fig. 4, has a sharp drop around the value of $\Delta k_a^D$ where significant Fourier components of the far-infrared output begin to be totally reflected at the boundaries. In SHG, however, total reflection is never important and therefore as shown in Fig. 16, no sudden drop of the output power occurs as $\Delta k_a^D$ increases. Because of the weaker diffraction effect, the phase-matching curve for SHG has, in general, more pronounced fine structure than that for DFG.

V. Conclusion

We have developed here the theory of far-infrared generation by optical mixing in a nonlinear medium, using an extension of a formalism developed earlier for second-harmonic generation by focused laser beams. The theory takes into account the effects of focusing, diffraction, and double refraction of the pumped beams and the effects of diffraction, absorption and reflections at the boundaries of the far-infrared output beam. Numerical calculations showing these effects are presented. Both the total power output and its angular distribution are calculated.

We have found that focusing of the pump beams can greatly enhance the far-infrared output. In a crystal of 1 cm long, the optimum focal spot radius is roughly equal to or smaller than the far-infrared wavelength for output frequencies less than $100 \text{ cm}^{-1}$. The walkoff effect of the pump beams in birefringent crystals does not reduce the output by more than a factor of 2. Far-infrared absorption and boundary reflections are however extremely important. The former is often the factor which limits the output power.
We show that the usual plane-wave model which neglects the effects of far-infrared diffraction and boundary reflections does not give a correct description of the far-infrared output, especially for tightly focused pump beams. A simple model treating the non-linear polarization as a constant 1/e radius Gaussian distribution of radiating dipoles is, however, a good approximation to the real picture. We also compare our results with those of second-harmonic generation and notice a great deal of similarities. Most of the differences can be ascribed to the boundary effects including total reflection which are more important in the case of far-infrared generation.
REFERENCES

1. For a recent review of nonlinear optical processes for generating far-infrared radiation which covers both applicable DFG experiments and basic theory see Y. R. Shen, Prog. Quantum. Electron. 4, 207 (1976).


35. M. Berek, Z. Kristallog. 76, 396 (1931). The fields in this work are magnetic fields (in-spite-of notation which suggests they are electric fields) because the Brewster's angle occurs for the "senkrecht" or perpendicular polarization. After these equations are rewritten in-terms-of electric fields, the "one-ray" coefficients are the elements of our A and B matrices. For an English translation with some typographical errors see G. N. Ramachandran and S. Ramasesham, "Crystal Optics" in Handbuch der Physik, S. Flugge, ed., Vol. XXV/1 (Springer-Verlag, Berlin, 1961) pp. 117-119.

36. K. Miyamoto and E. Wolf, J. Opt. Soc. Am. 52, 615 (1962). Note that their definition of the Fourier transform is smaller than our definition by a factor of $1/2\pi$. 
37. For tightly focused \( \exp \left[ -\omega^2 k_T^2 / 4 \right] \approx 1 \) when \( k_T < \frac{\omega}{c} \), and this factor can be pulled outside the integral in Eq. (35) along with \( \langle T \left( k_T \right) \rangle \). Then, the remaining integral is \( \int_0^{\omega/c} k_T dk_T C (\Delta k_T)/k_{oz} \) which has the value \( \approx \pi/\lambda \) for thick crystals or high far-infrared frequencies and the value \( \approx (\omega/2c) \left[ n(\omega) - \sqrt{n^2(\omega) - 1} \right] \) for thin crystals or low far-infrared frequencies.
CHAPTER III. FAR-INFRARED GENERATION BY PICOSECOND PULSES IN ELECTRO-OPTIC MATERIALS

The development of high power mode-locked lasers with pulse widths in the picosecond range has made optical rectification a feasible method of generating broadband radiation of high peak power (~1 kW), as supported by the recent experimental results. Theoretically, Gustafson et al. have calculated the rectified field for an infinite plane wave in the limit that the optical and far infrared phase velocities differ negligibly. They have also neglected reflection and refraction at the crystal boundaries. This letter reports a more realistic calculation which includes the various effects due to a finite beam cross-section, crystal boundaries, and the significantly different optical and far-infrared phase velocities.

Consider a short laser pulse incident normally on a thin slab of electro-optic material. The slab has a transverse dimension much larger than the beam diameter, and we can assume that the laser pulse propagating in the slab in a single transverse mode is given by

\[ E_{\xi} = \sum_j E_{\xi,j}(r,t) \]

with

\[ E_{\xi,j}(r,t) = \xi_{\xi,j} \left\{ \frac{1}{(1+i\xi_j)} \exp \left[ -\frac{x^2+y^2}{w_0^2(1+i\xi_j)} - \frac{(n_j z/c - t)^2}{\sigma^2} \right] + \right. \]

\[ + i \omega \left( n_j z/c - t \right) + i\phi \right\} \] (40)

where \( w_0 \) is the beam radius in the focal plane, \( \sigma \) is the pulse width, \( \xi_j = (L + z/n_j)/(\omega w_0^2/2c) \), \( L \) is the distance between the focal point
and the front surface of the slab, and $z$ is the distance away from the front surface into the slab. The subindex, $j$, indicates the polarization state of the laser field. The other quantities have their usual physical meaning.

The laser pulse induces in the slab a nonlinear polarization at difference frequencies of the form

$$ P_{NL}(x,t) = \chi_{NL}^{*} \cdot E_{\nu}E_{\phi}^{*} $$

(41)

if we neglect the dispersion of the nonlinear susceptibility $\chi_{NL}$. The far infrared radiation field $E(x,t)$ generated in the slab can then be obtained by solving the wave equation

$$ \left[ \nabla \times (\nabla \times) + (\varepsilon_{0}/c^{2}) \frac{\partial^{2}}{\partial t^{2}} \right] E(x,t) = - \frac{4\pi}{c^{2}} \frac{\partial^{2}}{\partial t^{2}} P_{NL}(x,t) $$

(42)

with the proper boundary conditions. Here, we have also neglected the dispersion of $\varepsilon_{0}$.

To solve Eq. (42), we use essentially the scheme of Bjorkholm. From the Fourier transform of $E(x,t)$ and $P_{NL}(x,t)$ on $x$, $y$ and $t$, we obtain the Fourier components $E(k_{T},\omega,z)$ and $P_{NL}(k_{T},\omega,z)$ respectively. We then use the Green's function method to find $E(k_{T},\omega,z)$. Although far-infrared radiation is generated in the slab in all directions, only the part which propagates in forward and backward directions with nearly normal incidence on the plane surfaces of the slab can get out of the slab because of the large refractive index of a crystal in the far infrared. If we are interested only in that part of the far-infrared
radiation, then we can use the Green's function for normal incidence as an approximation in finding \( E(k_T, \omega, z) \). Multiple reflections at the plane boundaries of the slab are clearly important, so that the solution should be proportional to a Fabry-Perot factor. For far-infrared field in the \( i \) polarization state, we find at the back surface of the slab, \( z = \xi \),

\[
E_i(k_T, \omega, \xi) = F_i \left[ E_i^{(S)}(k_T, \omega, z=\xi) - R_i E_i^{(S)}(k_T, \omega, z=0) \exp \left( i\omega n_i^{(w)} \xi/c \right) \right] \tag{43}
\]

with

\[
E_i^{(S)} = (2\pi \omega /c n_i^{(w)}) \int_0^\xi dz' F_i^{NL}(k_T, \omega, z') \exp \left( i\omega n_i^{(w)} |z-z'|/c \right)
\]

where \( F_i \approx (1-R_i)/(1-R_i^2 \exp(i2\omega n_i^{(w)} \xi/c)) \) is the Fabry-Perot factor, and \( R_i = (1-n_i^{(w)})/(1+n_i^{(w)}) \) is the reflection coefficient for the \( i \) polarization state.

Experimentally, the far infrared output from the slab is collected by a tapered light pipe leading to a solid-state detector. Since wave propagation in the light pipe has a cutoff angle \( \phi_M \), the total far infrared power seen by the detector is given by

\[
P_i(\omega) = (c/2\pi) \int_0^{k_T=|\omega/c| \sin \phi_M} d^2 k_T \left| E_i(k_T, \omega, \xi) \right|^2 \cos \phi \tag{44}
\]

where \( \phi = \sin^{-1}(k_T c/\omega) \leq \phi_M \). To calculate \( P_i(\omega) \) from Eq. (44) with the help of Eqs. (40), (41), and (43), we use the following approximations.

We assume that the cross-section of the laser beam remains unchanged
in traversing the thin slab. If both ordinary and extraordinary laser beams are present, then we also assume that walkoff of the two beams in the slab is negligible. Both assumptions are clearly good approximations when the slab is not unusually thick (< a few mm.). Since \( \phi_m \) is often small, we also approximate \( \cos \phi \) by 1 in Eq. (44). Then, if \( \chi_{ijk}^{NL} \) is the only dominating nonlinear susceptibility in difference-frequency generation, we find, for a slab of thickness \( \ell \),

\[
P_i(\omega) = I_i |F_i|^2 M_i D \tag{45}
\]

where

\[
I_i = (\pi/2c)(\omega_0/n_i(\omega))^2 |\chi_{ijk}^{NL} \xi_j \xi_k / (1+\xi_i^2)|^2 \ell^2
\]

\[
\Lambda = \pi \omega_0^2 (1+\xi_i^2)/4, \quad \xi = \xi_j (z=0)
\]

\[
M_i = |(M^-_{ijk} + M^-_{ikk}) - R_i (M^+_{ijk} + M^+_{ikk})|^2
\]

\[
M^\pm_{ijk} = [1 - \exp (i\Delta k^\pm_{jk} \ell)] / i\Delta k^\pm_{jk} \ell
\]

\[
\Delta k^\pm_{jk} = (1/c)[(\omega_0 + \omega/2) n_j^{(\omega_0)} - (\omega_0 - \omega/2) n_k^{(\omega_0)}] \pm \omega n_i^{(\omega)}
\]

\[
D = 1 - \exp \left[ -(\omega^2/4c^2) \omega_0^2 (1+\xi_i^2) \sin^2 \phi_m \right]
\]

\[
S = \exp \left( -\omega^2 \sigma^2 / 4 \right)
\]
The various quantities in the above equation have the following physical meanings. $A$ is the effective cross-section of the beam at the slab. $N_{jk}^{\pm}$ takes care of the phase mismatch in the difference-frequency generation process, with $\Delta k_{jk}^-$ and $\Delta k_{jk}^+$ being the average momentum mismatches for far-infrared waves propagating in the forward and backward directions respectively. $D$ accounts for the diffraction effect due to the finite beam cross-section. $S$ is the spectral content of the picosecond laser pulse. Finally, $I_A$ gives the far-infrared power spectrum if all the other factors in Eq. (45) are unity.

We now use Eq. (45) to calculate the spectra of the far-infrared output for two cases. In the first case, a 0.1 cm LiNbO$_3$ slab is oriented with the $c$-axis parallel to the plane surfaces of the slab. A 2-ps laser pulse at 1.06 $\mu$m, polarized along the $c$-axis, is normally incident on the slab, so that $\chi_{33}^{NL}$ is the only nonlinear susceptibility responsible for the difference-frequency generation. With $\hat{j} = \hat{k} = \hat{i}$ along $\hat{c}$ and $n_1^o \neq n_1(\omega)$ in Eq. (45), phase matching occurs only at $\omega = 0$. The calculated spectrum is shown in Fig. 18. The dashed curve gives the spectrum without the Fabry-Perot boundary condition. The peaks at 5, 8.4, and 11.8 cm$^{-1}$ are the secondary peaks of the phase-matching curve, which would have the major phase-matching peak at $\omega = 0$ if it were not for the low-frequency cutoff. This low-frequency cutoff is mainly due to the $\omega^2$-dependent radiation effects, and gives rise to the first peak at 2 cm$^{-1}$. The diffraction effect ($D$) only makes the cutoff even sharper, but does not affect the spectrum significantly beyond the first peak. On the high-frequency side, the spectrum is
limited by the spectral content $S$ of the input pulse. With the Fabry-Perot boundary condition included, the spectrum is then modified by the interference pattern, as shown by the solid curve with spikes in Fig. 18. For an input pulse of $1 \text{ GWatt}$ peak power, the total far-infrared output energy is about $0.1 \text{ erg}$. Our results here agree with those of Gustafson et al. in the limit $n_{1}^{(ω)} = n_{1}^{(0)}$ and when diffraction and boundary conditions are neglected.

In the second case, the LiNbO$_3$ slab is oriented with the c-axis tilted at $16.8°$ away from the normal of the slab and the a-axis is in the plane defined by the c-axis and the normal. The normally incident laser pulse is linearly polarized at $45°$ with respect to the plane such that only $\chi_{24}^{NL}$ is responsible for the difference-frequency signal with polarization perpendicular to the plane. We then find from Eq. (45) that the phase-matching conditions $\Delta k_{j}^{−} = 0$ and $\Delta k_{j}^{+} = 0$, for far infrared generation in the forward and the backward directions, respectively, can be satisfied at $ω = 15$ and $7.5 \text{ cm}^{-1}$, respectively. The far-infrared spectrum is then essentially the superposition of the two phase-matching curves modified by $ω^2 S(ω)$ and the boundary conditions. If the boundary conditions are neglected ($R_{i} = 0$), then only the far-infrared generated in the forward direction contributes to the spectrum as represented by the dashed curve in Fig. 19. With the boundary conditions, $R_{i} \neq 0$, the far-infrared generated in the backward direction now appears in the output. Its spectrum dominates over that of the far-infrared generated in the forward direction because of the high-frequency cutoff due to $S(ω)$. The total spectrum
is given by the solid curve in Fig. 19, where the spikes are again the result of Fabry-Perot interference. Diffraction has little effect in this case. For a laser pulse with a 1 - GWatt peak power, the total far-infrared energy generated here is 0.0064 erg. Both cases discussed above have been investigated experimentally. Preliminary results show good agreement with our theoretical calculations.¹

The time dependence of the far-infrared pulse must also be understood, especially the features responsible for the oscillation periods in the spectra of Figs. 18 and 19. These oscillation periods are due to the following three characteristic times: the far-infrared round-trip time, the sum of the optical and far-infrared transit times, and the difference of the optical and far-infrared transit times.

The far-infrared round-trip time, \( \tau_{rt} = 2n_1(\omega)l/c \), is responsible for the prominent Fabry-Perot oscillation period in the solid curves of Figs. 18 and 19. The sum and difference of the far-infrared and optical transit times, \( \tau_\pm = (n_1(\omega) \pm n_j)l/c \), are responsible for the node spacing of the backward and forward phase-matching functions, respectively. Since the characteristic times are independent of the temporal pulse shape, we choose a square pulse to illustrate the significance of \( \tau_\pm \). We shall also ignore diffraction in the following discussion.

Ignoring diffraction reduces Eq. (42) to a pair of scalar one-dimensional wave equations, one for the far-infrared o-ray and another for the e-ray:
where $P_i = \hat{i} \cdot \mathbf{p}_i^{NL}(z,t)/[1-(\hat{i} \cdot \hat{z})^2]$, $\hat{i}$ is the direction of the electric field, and $n_i$ is the far-infrared refractive index for the o- or e-ray.

When the nonlinear slab is imbedded in a medium with an identical linear dielectric tensor (reflectionless boundary conditions), the solution of Eq. (46) can be written down immediately from the standard Green's function for an outgoing-wave:

$$E_i(z,t) = -\int_0^L dz' \int_{-\infty}^{\infty} dt' H(t-t') \frac{|z-z'|}{c} n_i \frac{2\pi}{n_i c} \frac{\partial^2}{\partial t'^2} P_i(z',t')$$

(47b)

$$E_i(z,t) = -\int_0^L dz' \frac{2\pi}{n_i c} \frac{\partial}{\partial t'} P_i(z',t - \frac{|z-z'|}{c} n_i)$$

(47b)

where $H(u)$ is the unit step function [$H(u) \equiv 0$ for $u < 0$, and $H(u) \equiv 1$ for $u \geq 0$].

The case in which an optical o- or e-ray beats with itself is most easily understood by following a short pulse through the nonlinear slab. First, between its entry at $z = 0$ and its arrival at $z = L$ the optical pulse creates the same transmitted and reflected far-infrared pulses as it would if the nonlinear medium filled the entire half-space to the right of the $z = 0$ interface. Since $P_i$ is proportional to the optical intensity and dispersion is ignored, the solution of Eq. (46) for a single interface at $z = 0$ can be written down immediately from the solution for a monochromatic plane wave. Its transmitted
inhomogeneous \[ E^i_1(z,t) \], transmitted homogeneous, and reflected homogeneous far-infrared pulses are given by:

\[
E^i_1(z,t) = -\frac{4\pi}{n_1^2-n_j^2} P_i(0,t-zn_j/c) \quad \text{for } z > 0, \tag{48a}
\]

\[
E^r_1(z,t) = \frac{n_i-n_1}{2n_1} E^p_1(0,t+zn_i/c) \quad \text{for } z < 0, \tag{48b}
\]

\[
E^t_1(z,t) = -\frac{n_i+n_1}{2n_1} E^p_1(0,t-zn_i/c) \quad \text{for } z > 0. \tag{48c}
\]

\([E^p, E^r, \text{and } E^t \text{ are zero in the half-spaces not described in Eq. (48).}\]

At \( z = 0 \) all three of these pulses have the time-dependence of the optical intensity, except for sign reversals. Next, when the inhomogeneous pulse arrives at the \( z = \lambda \) interface, it generates another reflected and another transmitted far-infrared pulse. At \( z = \lambda \) both of these pulses and the optical intensity have identical time-dependence; also, the reflected and transmitted pulses have opposite polarity from the corresponding pulse generated at \( z = 0 \) because the inhomogeneous pulse propagates toward the \( z = \lambda \) interface, but away from the \( z = 0 \) interface. Finally, the homogeneous transmitted pulse from \( z = 0 \) and the homogeneous reflected pulse from \( z = \lambda \) arrive at and pass through the opposite surface. Since each of these pulses automatically satisfies the (linearly) reflectionless boundary conditions at the opposite surface, the transmitted (reflected) far-infrared wave is the sum of the transmitted (reflected) homogeneous pulses from \( z = 0 \) and \( z = \lambda \).
Figure 20 illustrates this case for a 2 psec square input pulse and the crystal parameters of the calculation shown in Fig. 18. The two opposite polarity single-surface pulses discussed above are clearly present in both the transmitted (lower curve) and reflected (upper curve) far-infrared fields. The first pulse of the transmitted electric field coincides with the optical pulse at \( z = \ell \) and the second follows it after a delay of the optical minus the far-infrared transit time. This delay of 9.5 psec causes the prominent \( 3.51 \text{ cm}^{-1} \) oscillation in both curves of Fig. 18. The reflected electric field at \( z = 0 \) (upper curve) is similar. The first pulse is the reflection off the input \( (z = 0) \) face of the slab while the second is the reflection off the exit \( (z = \ell) \) face. The time lag between the two pulses, \( \tau_+ = 24.167 \text{ psec} \), generates a \( 1.38 \text{ cm}^{-1} \) oscillation period in the solid curve in Fig. 18 which, unfortunately, is obscured by the \( 0.99 \text{ cm}^{-1} \) Fabry-Perot oscillation period. However, the \( 1.38 \text{ cm}^{-1} \) period is clearly present in the spectrum of the sum of the forward wave and the first reflection of the backward wave that, for the 2 psec square pulse, is shown in Fig. 21.

Figure 22 illustrates the time-dependence of the electric field generated by the beating of the \( o \)-ray and \( e \)-ray components of a single 2 psec square optical pulse for reflectionless boundary conditions. The crystal parameters are those used for the calculation shown in Fig. 19. As in Fig. 20, there are two regions of approximately 2 psec duration separated by \( \tau_+ \) in the backward wave at \( z = 0 \) (upper curve) or by \( \tau_- \) in the forward wave at \( z = \ell \) (lower curve) which are due to the single surface nonlinear reflections or transmissions. The signal
between the two surface pulses is due to the interference of the
radiation generated by the leading and trailing edges of the pulse;
its amplitude depends critically on the pulse length and is largest
when \( \omega_{jk} = (2m+1)\pi m = 0,1, \ldots \) (see Appendix D). Both during and
between the surface pulses, the time dependence is nearly sinusoidal
with a frequency that is given approximately by the corresponding
phase-matching condition \( \Delta k^+ = 0 \) or \( \Delta k^- = 0 \).

The nearly sinusoidal time dependence is easily explained: Due
to the different \( o \)- and \( e \)-ray phase velocities, the square pulse creates
a standing wave modulation, \( \cos[(n_j-n_k)\omega_0 z/c] \), of the nonlinear
polarization (plus a propagating second harmonic modulation that
doesn't concern us here) within the moving window determined by its
duration. If the input beam were continuous wave, this polarization
would have no time dependence and could not radiate; however, the
motion of the pulse edges creates a time-dependent source. (In the
one-dimensional plane wave case, only these edges radiate.) If we
consider just the effect of the leading edge of the pulse on the far-
infrared wave at \( z = \lambda \), the radiation arriving at time \( t \) was generated
at the retarded time \( t' = t-(\lambda-z') n_j/c \) at the point in space \( z' = ct'/n_j \)
which the leading edge then occupied; thus, the relationship between
the time of observation at \( z = \lambda \) and the point of emission is
\[ t - \lambda n_j/c = z'(n_j-n_1)/c, \]
and the contribution of the leading edge is
sinusoidal at the difference frequency \( \omega = \omega_0 (n_j-n_k)/(n_j-n_1) \). This is
approximately the frequency at which \( \Delta k^- = 0 \) when \( n_j-n_1 \gg n_j-n_k \).
Between the two surface pulses the velocity difference between the
leading \((c/n_j)\) and trailing \((c/n_k)\) edges causes a deviation from a truly sinusoidal field that is negligible unless the optical birefringence is large.

We have neglected dispersion and absorption in the above discussion. They can, however, be easily incorporated in the computer spectrum calculations. The effects vary from crystal to crystal. In \(\text{LiNbO}_3\), the absorption coefficient, \(\alpha\), in the far-infrared is roughly proportional to \(\omega^2\). \((\alpha \sim 18\ \text{cm}^{-1} \text{ at } 30\ \text{cm}^{-1})\)\(^\text{10}\). The decrease of the far-infrared power due to absorption is less than 20\% below 10 cm\(^{-1}\). We have also neglected the effect of possible frequency chirping of a mode-locked pulse.\(^\text{11}\) This is not important here since, in the product \(E_{\omega j}^* E_{\omega k}\), any phase modulation in \(E_{\omega j}\) is almost completely cancelled out by the same phase modulation in \(E_{\omega k}^*\). Finally, for a train of \(N\) identical mode-locked pulses with a time interval \(\tau\) between pulses, the far-infrared spectrum of Eq. (45) should be modified by the factor

\[ |\frac{1-\exp(iN\omega\tau)}{1-\exp(i\omega\tau)}|^2.\]

The total far-infrared energy is increased by a factor of \(N\).
REFERENCES


5. For extraordinary far-infrared ray, the equation should be modified to take into account the fact that $\mathbf{E}$ is not perpendicular to $\mathbf{k}$. See J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).


7. For $j = k$, we should replace $I_j$ by $I_j/4$ if we use the conventional definition for $\chi^{NL}_{ijkl}$.

8. See, for example, P. M. Morse and H. Feshbach, Methods of Theoretical Physics, (McGraw-Hill, New York, 1953), p. 843.

9. See, for example, N. Bloembergen, Nonlinear Optics, (Benjamin, New York, 1965), pp. 74-81.

CHAPTER IV. PHASE MATCHED FAR-INFRARED GENERATION BY DIFFERENCE-FREQUENCY MIXING OF TWO DYE LASER BEAMS

I. Introduction

The use of non-linear difference-frequency mixing of optical or infrared lasers to generate tuneable far-infrared radiation has been experimentally demonstrated many times.\(^1\)\(^-\)\(^10\) Various combinations of lasers and non-linear crystals have been used: temperature-tuned ruby lasers with LiNbO\(_3\);\(^1\)\(^,\)\(^3\) grating tuned CO\(_2\) lasers with InSb;\(^2\) ZnGeP\(_2\);\(^4\) and GaAs;\(^7\)\(^-\)\(^9\) a spin-flip Raman laser and a CO\(_2\) laser with InSb;\(^6\) and grating tuned ruby pumped dye lasers with LiNbO\(_3\), ZnO, and CdS\(^10\) and with reduced (black) LiNbO\(_3\).\(^5\) Only the dye laser system has demonstrated output that can be continuously tuned over the entire 20 to 190 cm\(^{-1}\) frequency range with a single laser system.\(^10\) Although the ruby pumped dye laser systems have a low repetition rate, the results obtained with them suggest that a suitably chosen flashlamp pumped dye laser system could be used to generate a greater than 1 Hz repetition rate, continuously tunable, far-infrared source using a LiNbO\(_3\) crystal; this source would operate from 20 to 160 cm\(^{-1}\) with a peak power of at least a few milli-watts.

This chapter describes the difference frequency generation of continuously tunable far-infrared radiation in the 20 to 190 cm\(^{-1}\) frequency range using two distinct ruby-pumped dye laser systems. In the experiments described here, we observed far-infrared radiation in the 20 to 160 cm\(^{-1}\) range with various phase-matching schemes in LiNbO\(_3\): forward collinear (FCPM), backward collinear (BCPM), and noncollinear
We have also investigated FCPM in ZnO, ZnS, CdS, and CdSe at selected frequencies as high as 190 cm\(^{-1}\). The observed far-infrared power is summarized in Table 1.

II. Synopsis of Plane-wave Theory

A. Plane-wave Theory for Collinear Phase Matching

Because our optical focal spot diameter of 3 mm was much larger than any of the far-infrared wavelengths that we investigated in LiNbO\(_3\), the plane wave theory of difference frequency generation (see Eq. (37) of Chapter II) adequately describes our experimental results above 40 cm\(^{-1}\). When, as in our experiments, the optical absorption is negligible and only \(\chi^{(2)}_{24}\) contributes to the difference-frequency signal, the far-infrared power can be approximated by

\[
P(\omega, \theta) = \frac{1}{4 \pi \omega c} \int_{-2\pi \omega c}^{2\pi \omega c} d\varepsilon \frac{n\omega^2}{4} I(\omega+\varepsilon, \theta)
\]

where

\[
I(\omega+\varepsilon, \theta) = \sin^2 \theta \left\{ \frac{8\pi^3 T_1 T_2 (\omega+\varepsilon)^2}{c n_1 n_2 [n^2+c\varepsilon^2/4(\omega+\varepsilon)^2]} |\chi^{(2)}_{24}|^2 I_1 I_2 \right\}
\]

\[
\times \left[ 1 - \exp(-\alpha^2/2) \exp\left\{ i[\Delta k(\omega, \theta) - \text{Be/c}] \right\} \right]^2 / [\Delta k(\omega, \theta) - \text{Be/c}]^2 + \alpha^2/4
\]

is the intensity generated by two monochromatic lasers at their difference frequency \(\omega+\varepsilon\); \(\alpha\) is the far-infrared absorption coefficient at \(\omega\); \(\Delta k(\omega, \theta)\) is the phase mismatch for the nominal laser frequency.
and the difference frequency \( \omega \); \( B \) is given by either \( B^+ = n + n_1 \) for FCPM or by \( B^- = n - n_1 \) for BCPM; 2\( \sigma \) is the effective bandwidth of our dye lasers; \( I_1 \) and \( I_2 \) are the pump laser intensities; \( T, T_1, \) and \( T_2 \) and \( n, n_1, \) and \( n_2 \) are the far-infrared, high-frequency optical, and low-frequency optical transmission coefficients and refractive indices, respectively; and \( \theta \) is the angle between the optical beams and the c-axis of the crystal.

B. Backwards Collinear Phase-matching

In crystals that have an optical birefringence sufficient to collinearly phase-match the difference frequency generation of a forward propagating far-infrared wave, there is also a range of frequencies in which the generation of a backward propagating or reflected difference frequency wave can be phasematched. This occurs because in the frequency range below the lowest infrared active TO phonon, the far-infrared refractive index is typically a monotonically increasing function of frequency and is usually greater than the optical refractive indices. This reflected difference frequency wave is similar to the reflected second harmonic wave generated in III-V semiconductors\(^{11} \) however, since it is phase-matchable, the coherence length can be a substantial fraction of the crystal thickness rather than just half a second harmonic wavelength.

If the forward, \( P_F \), and the backward, \( P_B \), collinearly phase matched powers are generated by identical pump laser beams in crystals that are identical except for their phase-matching angles, then according to Eq. (49) their ratio is the product of two factors:
1) the ratio of \( \sin^2(\theta) \) at their respective phase matching angles, and

2) the ratio of the mean values of their phase-matching functions averaged over the far-infrared linewidth. The ratio of the backward to the forward collinearly phasematched \( \sin^2 \theta \) factors is \( (n+n_1)/(n-n_1) \) when \( \omega_1 \gg \omega \) and when dispersion at the pump laser frequencies can be neglected. When the far-infrared generation process is phase matched at line center (\( \varepsilon=0 \) in Eq. (49)), \( P_B/P_F \), including the above ratio of the \( \sin^2(\theta) \) factors, is given by:

\[
P_B/P_F \equiv f[(n+n_1)\sigma]/f[(n-n_1)\sigma]
\]  \hspace{1cm} (50)

where

\[
f(u) \equiv \int_{-2\pi u}^{2\pi u} \frac{dv}{4v^2+\alpha^2} |1-\exp[-(2iv+\alpha)u/2]|^2.
\]

Thus, BCPM is more efficient than FCPM for type II collinear generation of a far-infrared o-ray in LiNbO₃; in the limit of a large absorption coefficient, \( \alpha \), the power ratio is given by \( P_B/P_F = (n+n_1)/(n-n_1) \) which can be as large as a factor of two in LiNbO₃.

C. Noncollinear Phase Matching

The most general phasematching configuration is non-collinear phasematching; the forward and backward collinear phasematching configurations are, in fact, limiting cases of non-collinear phasematching. The general phasematched three wave interaction is described by the wavevector triangle shown in Fig. 23a. For our difference-frequency case the index 1 and 2 waves are optical beams and the index 3 wave is the far-infrared beam. The special case \( \phi = \psi = 0 \) is forward collinear
phasematching with $\Delta k_{fc} = k_1 - k_2 - k_3 = 0$ while $\phi = 0$, $\psi = \pi$ is backward collinear phasematching with $\Delta k_{bc} = k_1 - k_2 + k_3 = 0$. By an elementary application of the law of cosines and half-angle formulas, we obtain for the angle between the two optical beams:

$$\sin^2\left(\frac{\phi}{2}\right) = -\frac{(k_1 - k_2)^2 - k_3^2}{4k_1 k_2}.$$  \hspace{1cm} (51)

and for the angle between the far-infrared beam and the high-frequency optical beam:

$$\sin^2\left(\frac{\psi}{2}\right) = \frac{1}{2} \left\{ 1 - \frac{n_2}{n_3} - \frac{(n_1 - n_2)\omega_1}{n_3\omega_3} \right\} \left\{ \frac{n_1 + n_2}{2n_1} - \frac{(n_3 + n_2)\omega_3}{2n_1\omega_1} \right\}.$$  \hspace{1cm} (52)

For far-infrared generation with optical or near-infrared lasers, the angle $\phi$ is small because $k_3$ is much smaller than either $k_1$ or $k_2$. However, the angle $\psi$ is often large.

Far-infrared generation with non-collinear phase matching is more sensitive to the divergence of the pump lasers than far-infrared generation with collinear phasematching whenever the angle, $\phi$, between the pump beams is more than about twice their divergence angles. The far-infrared radiation is generated by the interaction of the various plane-wave components of the pump laser beams. Thus, for phase matched mixing of divergent pump laser beams, Fig. 23a illustrates the phase matched mixing of the axial plane wave components that are at the center of the far-field diffraction patterns of the pump laser beams, and Fig. 23b illustrates the mixing of this component of one
pump beam with a plane wave component at the edge of the main diffraction lobe of the second. The value, $\Delta k'$, of the phase mismatch for the case when $\Delta \vec{k}$ and $\vec{k}_3$ in Fig. 23b are collinear is a measure of the effect of the pump laser divergence angle $\delta$; its magnitude is the value of $|\Delta \vec{k}|$ for the most nearly phase-matched plane-wave component of the far-infrared radiation in Fig. 23b, and it is given by:

$$\Delta k = \frac{u}{k_3 + \sqrt{(k_3^2 + u)}} \sim k_3 \delta \cdot \sin \psi$$

(53)

where $u = 4k_1k_2 \sin(\delta/2) \sin(\phi + \delta/2)$ and the approximation in Eq. (53) is valid when $|\Delta k| \ll k_3$ and $\delta \ll 1$. In the collinear phasematching cases ($\phi = 0$), the range of the phase mismatch is of the order of $\delta^2$ and thus a few milliradians of beam divergence can often be ignored. In the non-collinear case ($\phi > 2\delta$), the phase mismatch becomes significant more quickly because it grows linearly with $\delta$.

### III. Experimental Equipment and Techniques

#### A. Laser Systems

For many of the collinear and all of the noncollinear phase-matching experiments, a conceptually simple system of two separate dye lasers pumped by a single Q-switched ruby laser was used. The active medium was a solution of 3,3'-diethylthiatricarbocyanine iodide (DTTC iodide) in dimethyl sulfoxide (DMSO) contained in a 1 cm Beckman spectrophotometer cell. Each laser was arranged in Bradley's nearly longitudinally pumped configuration with a 312 mm$^{-1}$ blazed echelle
grating used in a sixth-order Littrow or back reflection geometry and a 70% reflectivity dielectric mirror to complete the laser cavity. The .5 J, 30 ns ruby pump pulse used in each dye laser generated about 24 mJ of dye laser energy that was continuously tunable from about 810 nm to greater than 840 nm.

With this pump pulse, the dye laser output was insensitive to factor-of-two changes in dye concentration from our operating concentration (20% low-level light transmission at 600 nm). Although the low-level light transmission at 6943 Å was less than 1%, the ruby laser saturated the dye absorption band since its energy burned black masking tape after passing through the dye cell. The only precautions taken with the dye solution were to cap the dye cell to keep moisture away from the hygroscopic DMSO solvent and to operate the laser in a darkened room to retard the degradation of the DTTC iodide by ambient light. To obtain a reasonable power output, the incident ruby light was plane polarized parallel to the grating rulings to match the maximum dye laser gain to the maximum reflectivity of our gratings.

Figure 24 describes a more novel dye laser that was used for some collinear phasematching experiments. A 1 J, 30 ns ruby pump pulse provided a large gain for both the orthogonal linear polarizations selected by the intracavity Glan-Thompson prism. This prism makes possible the independent tuning and selection of a frequency for each polarization with two echelle gratings. To obtain good temporal overlap of and high power for the two frequency components, the net gains of the two arms of this dye laser were approximately equalized by
polarizing the ruby pump pulse either circularly or linearly at 45° to
the transmitted polarization of the Glan-Thompson prism. The glass
microscope intracavity beam splitter provided fine tuning of the relative
gains of the two polarizations. Under our operating conditions, the
output polarizations for each beam had no more than 10% of their energy
in the undesired orthogonal linear polarization.

An amplified Q-switched ruby laser pulse pumped the dye lasers.
The oscillator consisted of a right angle prism Q-switch rotated at
400 sec⁻¹ with a hysteresis-synchronous motor; a 4 inch long, 5/16 inch
diameter ruby rod pumped by two water cooled linear flashlamps in a
double ellipse cavity; and a glass resonant reflector with 25%
reflectivity. The amplifier was a 4 inch long, 3/8 inch diameter
ruby rod pumped by the same configuration of flashlamps as the oscillator
rod. Both ruby rods were water cooled from a common tank to equalize
their operating temperatures and to decrease the necessary delay between
successive shots. The oscillator and the amplifier rods were pumped
simultaneously to simplify the trigger-circuit electronics. The
oscillator was Q-switched about 400 μs after the peak of the flashlamp
pulse which was about 800 μs long.

Since misaligning the amplifier rod increased the laser beam
divergence and made its near field intensity asymmetric and non-uniform,
the amplifier rod faces were aligned parallel to the oscillator resonant
reflector and ruby rod faces; then, the oscillator and amplifier, which
were less than four feet apart, operated as a coupled system. With
new flashlamps, the oscillator and amplifier flashlamps discharged
550 J from a 360 μfd capacitor bank and 750 J from a 395 μfd capacitor bank, respectively. Amplifier flashlamp discharges of up to 1500 J were possible and up to 1100 J discharges were employed regularly. The oscillator flashlamp discharge energy was set to just below the threshold for multiple-pulses which was about 200 J above the threshold for lasing. Under these operating conditions, the ruby laser system generated about 1 J in a single 30 ns pulse.

B. Far-infrared Optics and Detection

To measure the far-infrared difference frequency signals generated in our experiments, we used an InSb Putley detector between 20 and 95 cm\(^{-1}\) and a Ge:Ga photoconductive detector between 95 and 180 cm\(^{-1}\). The InSb detector was operated at 1.4°K in a 14.5 kG magnetic field. Both the low temperature and the magnetic field increased the responsivity and the signal-to-noise ratio of the detector-amplifier system by increasing the resistivity of the InSb sample. The magnetic field was also responsible for the high frequency end of this detector's response by tuning the InSb cyclotron resonance peak\(^{19}\) to approximately 95 cm\(^{-1}\). The rotary vacuum pump used to maintain the He temperature at 1.4°K was attached to the He dewar through a large reservoir to prevent modulation of the InSb detector's responsivity at the rotation speed of the pump. The Ge:Ga detector was operated at 4.2°K.

In our forward collinear and non-collinear phasematching experiments, the far-infrared radiation was collected by a 1 cm diameter evacuated light pipe beginning approximately .5 cm from the non-linear crystal. The radiation propagated along a straight section of light pipe,
reflected off the flat brass mirror of a 90° bend, and then propagated down another straight section of light pipe to the detector; the Ge:Ga detector had a condensing cone at the bottom of the light pipe to focus the radiation onto the detector surface. Three .25 mm thick sheets of black polyethylene were used to filter out unwanted radiation including that of the dye lasers; one of these sheets also served as the outermost vacuum seal for the light pipe. No signal was observed when the non-linear crystal was removed and the dye lasers were fired directly into the light pipe.

Both detector systems including their polyethylene filters were calibrated against the flat response of a Golay cell with a diamond window by conventional Fourier transform spectroscopy. A separate interferogram was taken with each of the 3 detector systems; the same mercury arc source, Michelson interferometer, and light pipe system were used for each interferogram. Each interferogram was digitally Fourier transformed and the frequency dependence of the responsivities of the Ge:Ga and InSb detectors were determined as the point-by-point quotient of that detector's spectrum to the spectrum measured by the Golay cell. Differences in the product, \( A\Omega \), of the area and collection solid angle of the detectors were assumed not to effect their relative frequency response.

K. H. Yang determined the absolute responsivity of the Ge:Ga detector at 125 cm\(^{-1}\). He measured the response of both the Ge:Ga detector and the Golay cell to the mercury arc source through the Michelson interferometer at zero path difference; the ratio of the
Ge:Ga to the Golay cell signal was $4.8 \times 10^{-3}$. From the responsivity curve of the Ge:Ga detector, the amount of $125 \text{ cm}^{-1}$ radiation that would generate the same response was calculated to be $2/3$ of the radiation incident upon this detector from the mercury arc lamp. By correcting for this and the 3 times larger $A_\Omega$ value of the Ge:Ga detector, we concluded that the ratio of the responsivity of the Ge:Ga detector at $125 \text{ cm}^{-1}$ to that of the Golay cell was $2.4 \times 10^{-3}$. Since (according to its manual) the responsivity of the Golay cell is $8.3 \times 10^5 \text{ V/Watt}$, the responsivity of the Ge:Ga detector is $2 \times 10^3 \text{ V/Watt}$.

K. H. Yang also determined the noise equivalent power (NEP) of the Ge:Ga detector. He measured the electrical noise voltage of this detector at the $1.35 \text{ V}$ bias voltage used in our experiments with a lock-in amplifier at several chopping frequencies at or above $280 \text{ Hz}$. Under his measuring conditions the contribution of the preamplifier of the lock-in amplifier to the observed noise signal was negligible. The $1.2 \times 10^{-7} \text{ V/Hz}$ noise thus measured gives a NEP of $6 \times 10^{-11} \text{ W/Hz}$ or $4.24 \times 10^{-8}$ Watts within the $500 \text{ kHz}$ bandwidth of the detection system. The noise level of the operational amplifier with the detector replaced by its equivalent room-temperature resistor was $.3 \text{ mV}$ peak-to-peak compared to the $2 \text{ mV}$ noise level of the combined detector-amplifier system. Thus, the detector-amplifier system was detector noise limited and the observed signal-to-noise ratio is relative to this detector NEP.

Based on the above NEP value, the observed signal-to-noise ratio of 66 (relative to the $.7 \text{ mV}$ rms noise) implies that about $4.9 \text{ mW}$ of far-infrared power was generated at $125 \text{ cm}^{-1}$ in our FCPM experiment in
LiNbO$_3$. To arrive at this number three corrections had to be made: First, the .75 mm black polyethylene filter used in the experiment, but not in the detector calibration, attenuates the $125\ cm^{-1}$ radiation by a factor of 8.65. Second, a factor of 3 attenuation due to the light pipe was experimentally measured. Finally, since the 30 ns far-infrared pulse generates an impulse response in the detector, the detector averages the pulse energy over its $2\ \mu s$ response time to record a signal corresponding to $30/2000$ or $1/67$ times the peak far-infrared power.

C. Sum-Frequency Normalization

Because of the low repetition rate of a ruby laser system, we resorted to sum-frequency normalization to reduce the shot-to-shot fluctuations in our measurements. In the collinear experiments, a fraction of the combined two-frequency laser beam was split off with a glass microscope slide and directed onto a polished (110) surface of a GaAs or InAs crystal. Since both SFG and DFG are second order processes, the effects of power fluctuations and temporal overlap fluctuations are eliminated by this scheme. However, even with identical field distributions, the fluctuations in spatial overlap are not completely eliminated by this scheme as the long wavelength of the difference-frequency signal causes the fields from neighboring hot-spots to interfere due to the large diffraction angle from each hot-spot as was discussed in chapter II. In the collinear experiments using two separate dye lasers, we located the sum and difference frequency crystals equal distances from the sum-frequency beam-splitter.
Since the latter followed all the beam combining and focusing optics in our system, each of the two fields at one crystal was proportional (as nearly as possible) to the corresponding field at the other crystal. Thus, for this group of experiments, spatial overlap fluctuations will be reduced for hot-spots that are large compared to the far-infrared wavelength.

Discrimination of sum-frequency signals against second harmonic signals was achieved with Armstrong's scheme\(^2\) by making use of the \(\bar{4}3m\) crystal symmetry in which \(\chi_{ijk}\) vanishes if \(i,j,k\) is not a permutation of \(1,2,3\). Linearly polarizing one laser parallel to the \([001]\) direction eliminated its second harmonic signal. The second laser was polarized perpendicular to the \([001]\) direction; thus, its second harmonic was plane-polarized along the \([001]\) direction and was eliminated by a linear polarizer between the crystal and the photomultiplier. The \([001]\) direction was made normal to the plane of incidence of the two laser beams to ensure that the above polarization directions were maintained inside as well as outside the crystal.

Since we measured the frequency dependence of the far-infrared power, we needed a sum-frequency normalization that did not vary with the difference frequency, \(\omega\), except through the product of the pump laser intensities. The novel use of SFG by reflection from a highly absorbing crystal eliminated one source of variation with \(\omega\); the highly damped solution has no Maker fringes to make the sum-frequency signal a rapidly varying function of the input laser frequencies as the large phase mismatch of the reflection geometry is nearly constant.
over the fractionally small tuning ranges used in far-infrared DFG experiments. Phase matching could also be used to eliminate the effect of Maker fringes, but it is impractical for our experiments because it requires a tedious adjustment of the orientation of the sum-frequency crystal each time the frequency of either dye laser is changed. Other experimenters have used sum-frequency generation by transmission through a transparent crystal with a ground exit surface \(^{24,25}\) for this purpose; we could have used this technique with a crystal like KDP, but the collection of the output sum-frequency signal would have been complicated by its poor colimation. Fortunately, the much larger sum-frequency susceptibilities of GaAs and InAs compared to those of readily available transparent crystals compensates for the much shorter coherence length of the reflected wave geometry. Because the 12,000 to 12,200 cm\(^{-1}\) frequency range that our dye lasers were tuned through in these experiments is small compared to the separation between the dye laser frequencies and band gap frequencies of GaAs (\(\sim 10900\) cm\(^{-1}\)) and InAs (\(\sim 2130\) cm\(^{-1}\)), the sum-frequency susceptibility of our normalization crystal was nearly independent of the difference frequency \(\omega\); together with the lack of Maker fringes, this made the sum-frequency signal a good normalization against the frequency dependence of the dye laser power.
IV. Forward Collinear Phase-matching Experiments

A. Experimental Measurements and Data Analysis for LiNbO$_3$

Since the experimental data available in the literature on the o-ray absorption coefficient of LiNbO$_3$ between 70 and 1000 cm$^{-1}$ is based on a Kramers-Kronig analysis of far-infrared reflectivity data$^{26}$ rather than a direct measurement and since sample-to-sample differences are possible due to impurities or slightly non-stoichiometric composition, we determined it at each frequency by measuring the far-infrared power as a function of wavevector mismatch. DeMartini has used this technique, known as momentum spectroscopy,$^{24,27}$ to measure the far-infrared absorption coefficient and the difference-frequency susceptibility of GaP.$^{24,25}$ However, we have used the birefringence of LiNbO$_3$ to vary the momentum or phase mismatch while De Martini used the noncollinearity of the pump beams and the various plane wave components of his far-infrared radiation in the optically isotropic GaP crystal.

We used seven LiNbO$_3$ slabs cut from a single Crystal Technology, Inc. boule with angles between the c-axis and the slab normal of 15°, 25°, 35°, 45°, 55°, 65°, and 90° (the a-axis, c-axis, and slab normal were coplanar). This permitted the frequency range 20 to 127 cm$^{-1}$ to be spanned without the complications of angles of incidence over 20°. The surfaces were ground flat on a series of SiC abrasive papers and optically polished with 1 μ diamond abrasive. For our experimental measurements these LiNbO$_3$ slabs were mounted on the axis of a rotating platform with their c-axes, their slab normals, and the laser beam
direction all perpendicular to the axis of rotation. At each frequency, $\omega$, above 40 cm$^{-1}$ we measured the power as a function of the orientation of the crystal, $P(\omega, \theta)$, over a wide enough range of $\theta$ to determine its angular width and thus its absorption coefficient, $\alpha(\omega)$.

To obtain the far-infrared absorption coefficient $\alpha(\omega)$ from a set of experimental measurements, \(\{(P_i, \theta_i) / i=1,2,\ldots,m\}\), we used a nonlinear least-squares fit. To make the computation tractable on a small computer, we neglected the $\varepsilon$-dependence of the term of Eq. (49b) that is enclosed in braces and eliminated the terms that contain $\exp(-\alpha \varepsilon/2)$; since $\alpha \varepsilon \gg 2$ at the frequencies at which we determined $\alpha$, the latter approximation is valid in our experiments. We also replaced $\Delta k(\omega, \theta)$ with $2\pi[\Delta_0 + \Delta(\theta)]$ where $2\pi\Delta(\theta)$ is the phase mismatch at the difference frequency $\omega$ and at the nominal laser frequency, $\theta$ is the angle between the laser beam axis and the $c$-axis, and $\Delta_0$ accounts for the experimental uncertainties in the values of the nominal laser frequency and the angles $\theta_i$. With these assumptions $P(\omega, \theta)$ from Eq. (49) is given by

$$P(\omega, \theta, \alpha) = \frac{A \sin^2 \theta}{2\sigma \sqrt{\Gamma}} \left\{ \tan^{-1} \left( \frac{\Delta_0 + \Delta(\theta) + \beta \sigma}{\sqrt{\Gamma}} \right) - \tan^{-1} \left( \frac{\Delta_0 + \Delta(\theta) - \beta \sigma}{\sqrt{\Gamma}} \right) \right\}$$

(54)

with $\Gamma = \alpha^2/16\pi^2$ and $\alpha \equiv (A, \Delta_0, \Gamma)$. We minimized the function

$$\chi^2(\Lambda, \Delta_0, \Gamma) = \sum_{i=1}^{m} \frac{(P_i - P(\omega, 0_i))^2}{(m-3)}$$

with respect to $\Lambda$, $\Delta_0$, and $\Gamma$ by the minimization process discussed in Appendix E.

As we can see from Eqs. (49) or (54), determination of $\alpha$ from our data requires that the effective bandwidth, $2\sigma$, of our dye lasers be measured. From Eq. (54) we can also see that, for a long crystal
(\alpha l \gg 2), \sigma is determined by measuring \( P(\omega, \theta) \) at a frequency where \( \alpha \) (and hence \( \Gamma \)) is known. For any value of \( \alpha \), the minimum value of \( \sigma \) that can be determined from the angular width of \( P(\omega, \theta) \) is proportional to \( \alpha / 4\pi B \) since Eq. (54) is nearly independent of \( \sigma \) when both \( \tan^{-1} \) functions can be approximated by their arguments. For a crystal less than two absorption lengths thick, the finite thickness makes the width of \( P(\omega, \theta) \) even less sensitive to \( \sigma \) as can be seen from Fig. 25.

To measure the effective bandwidth of our dye lasers, we used a 1.61 mm thick LiNbO\textsubscript{3} sample that was cut from a Hansen Microwave Lab. boule with its slab normal at 16° to the c-axis. At a far-infrared frequency of 21 cm\(^{-1}\), effective dye laser bandwidths smaller than about .8 cm\(^{-1}\) could not be measured even with a crystal many absorption lengths thick. However, since our sample was less than two absorption lengths thick (\( \alpha = 9.5 \) cm\(^{-1}\)), the smallest measurable bandwidth was about 1.5 cm\(^{-1}\) as can be seen from Fig. 25; this sensitivity to relatively narrow bandwidths is due primarily to the large difference (4.4) between the optical and far-infrared \( o \)-ray refractive indices. The results of our experimental measurement are shown in Fig. 26; they correspond to a 3 cm\(^{-1}\) bandwidth for our far-infrared radiation.

B. Results of Forward Collinear Phase-matching Experiments

The results of our measurements of the \( o \)-ray absorption coefficient, \( \alpha_o \), in LiNbO\textsubscript{3} are summarized in Fig. 27. The solid curve is a composite of Bosomworth's\textsuperscript{28} far-infrared transmission measurements for \( \omega \leq 70 \) cm\(^{-1}\) and Barker and Laudon's\textsuperscript{26} Kramers-Kroenig analysis of their far-infrared reflectivity data for \( \omega > 70 \) cm\(^{-1}\). Our results agree satisfactorily
with those of Barker and Loudon between 80 and 110 cm\(^{-1}\). Between 40 and 70 cm\(^{-1}\) our values of \(\alpha_0\) are significantly larger than the values of Bosomworth. The difference is due to a weak, broad absorption peak centered near 65 cm\(^{-1}\) that may be due to two phonon absorption. \footnote{29} Bosomworth's data also suggests the presence of a peak near 65 cm\(^{-1}\), but his peak is much weaker than ours; perhaps this is just sample variation between his slightly greenish LiNbO\(_3\) crystal and our colorless sample.

Figure 28 contains a comparison of our FCPM experimental results for LiNbO\(_3\) (the circles) and the plane wave theory of Eq. (49) with the absolute power at 110 cm\(^{-1}\) treated as an adjustable parameter. Above 40 cm\(^{-1}\) this theory \footnote{30} reproduces our experimental results satisfactorily given the multi-mode nature of our dye laser beams. However, the theoretical absolute power at 125 cm\(^{-1}\) is 22.5 mW, a factor of 4.6 larger than our measured power. Below 40 cm\(^{-1}\) the observed power falls off far more rapidly with decreasing frequency than is predicted by Eq. (49).

The calculations shown in Figs. 29a and 29b strongly suggest that this reduction in the DFG efficiency below 40 cm\(^{-1}\) is due to the multi-mode nature of our laser beams and the inherent misalignment of their hot-spots. These calculations used the more complete theory of Chapter II for single mode dye laser beams; for Fig. 29a the two dye laser beams were coaxial (\(a=0\)), but for Fig. 29b their axes were 0.5 mm apart. The other parameters roughly approximate our laser beams and our LiNbO\(_3\) crystal: 300 kW in each dye laser beam, \(\lambda_1 = 833\) nm, \(w = .08\) mm, \(z_0 = 50\) cm (2 mr divergence half-angle at half intensity and 3.3 mm
1/e² diameter at the crystal), \( l = 0.65 \text{ mm}, \, \chi^{(2)} = 3.18 \times 10^{-6} \sin \theta_M \) where \( \theta_M \) is the phase-matching angle, \( n_0 = 2.257, \, n_0 - n_{em} = 0.08192 \), a cubic spline fit to the absorption coefficients in Table II, a 30° detector collection half-angle, and an axially phase-matched DFG interaction. The far-infrared radiation generated below 20 cm⁻¹ is reduced by more than a factor of 10 by the 0.5 mm separation of the laser beam axes.

At a sufficient distance, \( z_o \), from the focal plane, optical pump beams separated by many focal plane radii diffract into each other and form a Gaussian nonlinear polarization centered at the midpoint between the two beam axes. Since geometric optics adequately describes the pump laser beams at such a distance from the focal plane, at the midpoint between the axes the Pointing vectors of the two laser beams, which give the direction of the local wavevectors, lie in the plane of the laser beam axes at equal, but opposite, angles \( \pm a/2z_o \) to them. Thus, inside the crystal the difference-frequency radiation is generated preferentially about a direction at an angle \( \sin^{-1}(k_1a/k_3z_o) \) to the laser beam axes where \( k_1 \) and \( k_3 \) are the optical and far-infrared wavevectors in the crystal, respectively. For the calculation of Fig. 29b this angle passes through the total reflection angle at 24 cm⁻¹; thus, only above the far-infrared frequency \( \omega = ck_1a/z_o \), is the far-infrared power improved significantly by placing the crystal for enough away from the focal plane that the hot-spots diffract into one another (as in our experiment).

We have also extracted a rough measurement of the frequency dependence of \( \chi_{24}^{(2)} \) from our data. The results are shown in Fig. 30 together with a multiple simple-harmonic-oscillator calculation \( \chi_{24}^{(2)} \) of \( \chi_{24}^{(2)} \) in
which we used the oscillator strengths, TO phonon frequencies, and line-widths of Barker and Loudon\textsuperscript{26} and the Raman cross sections of Kaminow and Johnston.\textsuperscript{33} The agreement is satisfactory. Furthermore, the monotonically increasing $|\chi_\text{24}^{(2)}|^2$ without a broad dip between 80 and 90 cm\textsuperscript{-1} indicates that the 152 cm\textsuperscript{-1} E-symmetry mode contribution to the low frequency (say 1 to 10 cm\textsuperscript{-1}) $\chi_\text{24}^{(2)}$ has the same sign as the sum of the contributions from the electronic and the remaining E-symmetry vibrational modes.

Table I summarizes the far-infrared power generated in 1 mm thick samples of ZnO, ZnS, CdS and CdSe at their maximum phase-matchable frequencies (all samples had their c-axes parallel to the sample surfaces). The ZnO and CdS samples were pre-cut and rough-ground by the manufacturers; the two CdSe slabs were cleaved from bulk samples and rough-ground on successively finer grades of SiC paper. The final optical polishing of all these relatively soft samples was accomplished by hand lapping in a water slurry of 1 μ Al₂O₃ abrasive. The ZnS sample was a long 1 mm wide strip that was optically polished by the manufacturer; this sample was not wide enough to accept all of the dye laser beam output, so the number reported in Table I has been corrected to estimate the output attainable with a crystal wider than the laser beams.
V. Backward Collinear Phase-matching Experiments in LiNbO$_3$

A. Modifications of FCPM Experimental Apparatus for BCPM

We have observed the phasematched difference frequency generation of a reflected far-infrared wave in LiNbO$_3$. In spite of the 54% far-infrared o-ray reflectivity, above 40 cm$^{-1}$ the laser input surface cannot be used as a mirror to reflect the far-infrared wave into a detector behind the crystal because the absorption coefficient is too large. To collect the radiation from the laser input side of the crystal, we added a short piece of brass light pipe and a right angle bend to the far-infrared collection optics used for the forward collinear phasematching experiments. The dye laser beams passed through a 1/8 in diameter hole milled through the flat brass reflector of the right angle bend; the axis of the hole was parallel to the laser beams and at a 45° angle to the normal of the brass plate. This collection technique added about 5 cm of air to the far-infrared propagation path.

B. Results of BCPM Experiment

As is shown in Fig. 28, our BCPM data do not agree with the simple theory of Eq. (49) since the predicted dip near 65 cm$^{-1}$ in Fig. 28 was not observed in our experiment. We believe that this difference is due to the frequency dependent losses through the 1/8 in diameter hole in the brass light pipe. As the far-infrared frequency increases, the far-infrared beam becomes more collimated around its nominal reflected direction; and when phase matching occurs near normal incidence, more far-infrared energy can escape through the hole at high difference
frequencies that at low ones. The added 5 cm of air in the propagation path will also modify the frequency dependence of the far-infrared signal due to water vapor absorption. The net result is extremely difficult to calculate and we will not attempt to do so.

VI. Noncollinear Phase-matching Experiments in LiNbO₃

A. Experimental Methods

The experimental set-up is shown in Fig. 31. Two separate dye lasers were used; they were synchronized by pumping them with a single ruby laser pulse. The plane of incidence of the dye lasers on the LiNbO₃ sample was horizontal for experimental convenience. To maximize the far-infrared radiation generated, the non-linear polarization vector and far-infrared wavevector were made orthogonal by using vertically polarized dye lasers. The angle between the two dye laser beams was adjusted by rotating the right angle prism PM₁ and the intersection of the two beams was positioned at the input surface of the LiNbO₃ crystal by moving this prism on a translation stage. The two dye laser beams were separately focused a few cm behind the LiNbO₃ crystal. Sum frequency normalization was accomplished as shown in Fig. 31. Since the dye laser field configurations at the sum and difference frequency crystals were not similar, the sum frequency signal compensates for the power and temporal overlap fluctuations of the dye lasers but cannot be guaranteed to compensate for spatial overlap fluctuations even at high difference frequencies.

Figure 32 is a top view of the single 4 mm cube LiNbO₃ crystal used in our NCPM experiments. (LiNbO₃ was chosen for this experiment
because of its large electro-optic coefficient and its availability.)

In the shaded region, the nearly parallel dye laser beams create a
difference-frequency polarization that radiates along the direction of
the difference of the two optical wavevectors. To permit the far-
infrared radiation to leave the crystal with an acceptable reflection
loss, we cut off the corner of our LiNbO$_3$ cube at an angle corresponding
to $\psi_0 = 68^\circ$ so that the far-infrared radiation would be within 4° of
normal incidence inside the crystal over the frequency range 1 to
150 cm$^{-1}$; at 160 cm$^{-1}$ the angle of incidence was approximately 5°.

We chose our experimental geometry with all three electric fields
polarized along the c-axis of our LiNbO$_3$ sample for three reasons:
First, $\chi_{33}$ and $\chi_{24}$ are an order of magnitude larger than the other
non-zero difference-frequency susceptibility tensor elements. Second,
$\chi_{24}$ yields a far-infrared o-ray while $\chi_{33}$ yields an e-ray. The latter
has a smaller absorption coefficient than the former; thus, the $\chi_{33}$
configuration is a more efficient far-infrared source. Finally, as can
be seen from Eq. (52) when $n_1 = n_2$ and $n_3$ is approximately constant,
as in the $\chi_{33}$ configuration, the angle $\psi$ between the far-infrared and
optical wavevectors is nearly independent of $\omega_3$, and a single sample of
LiNbO$_3$ with its corner cut off at a nominal value of $\psi_0$ can be used at
all frequencies of interest. If $\psi$ varied significantly with $\omega_3$,
multiple samples would have to be used to avoid large losses due to
total internal reflection.
B. Results of Noncollinear Phase-matching Experiments

To analyze our results, we have made use of the simple approximation of a plane wave far-infrared field that propagates perpendicular to the cut off surface of the cube. Since the 4 mrad divergence of the pump laser beams creates a 140 cm\(^{-1}\) range of \(\Delta k\) values, we averaged the plane wave power over this range using a uniform weight at each \(\Delta k\). We actually used the plane wave formula for collinear phase matching with an effective length, \(l_{\text{eff}}\), that is roughly the shortest distance along the far-infrared beam between points at which the phases of the nonlinear polarization are uncorrelated. A value of \(l_{\text{eff}} = 0.05\) cm gave a good fit to our data at 40 and 55 cm\(^{-1}\); at and above 75 cm\(^{-1}\) the calculated values are independent of \(l_{\text{eff}}\) due to the large absorption coefficient. This simple analysis fits our data amazingly well as can be seen in Fig. 28. The justification for treating the pumped region as a thin slab is that the coherence length of \(\sim 0.05\) cm is much shorter than our beam diameter of \(\sim 0.3\) cm so the exact shape of the non-linear polarization region becomes unimportant as the slab like first few coherence lengths dominate. For the case of coherent pump lasers, a more general plane-wave analysis that considers the shape of the pumped region has been given by Lax and Aggarwal\(^7\) in their work using a non-collinear phase matching geometry.

VII. Discussion of Results and Conclusions

A. Comparison with Raman Scattering Results

Our experimentally derived ratios of the nonlinear difference frequency generation susceptibilities of LiNbO\(_3\), ZnO, and CdS at their
respective maximum phase matching frequencies are within 20% of independently computed values. The ratios obtained from our far-infrared power measurements are \( d_{24}^{(\text{LiNbO}_3)} : d_{15}^{(\text{ZnO})} : d_{15}^{(\text{CdS})} = 1 : 9.1 \times 10^{-3} : 5.2 \times 10^{-2} \). Based on reported values of optical refractive indices,\(^{34-36}\) far-infrared reflectivities,\(^{26,37,38}\) Raman scattering cross sections\(^{33,39,40}\) of all three crystals and based on the clamped linear electro-optic coefficient \( r_{42}^{(\text{LiNbO}_3)} \),\(^{41}\) the computed ratios are \( 1 : 1.0 \times 10^{-2} : 6.3 \times 10^{-2} \). Such close agreement is remarkable for far-infrared difference frequency generation experiments.

Figure 33 displays the dispersion of the ionic, \( r_q \), and electronic, \( r_e \), contributions to \( r_{51} \) for \( \text{CdS} \) at 80\(^{\circ}\)K based on the Raman scattering data of Ralston et al.\(^{40}\) and on Loudon's theory.\(^{42}\) Because the Raman scattering cross sections are proportional to the square of matrix elements while \( r_e \) and \( r_q \) are linear functions of the same matrix elements, there are four simultaneous solutions for \( r_e \) and \( r_q \) in-terms-of the LO and TO phonon Raman scattering cross sections; but one pair of these solutions differs from the other pair by only a sign. The magnitude of \( r_q \) is determined by the TO phonon Raman cross section; however, there are still two possible magnitudes of \( r_e \) which are shown in Fig. 33 for \( \text{CdS} \). Our far-infrared measurements require that \( r_e \) is given by the upper curve so that, for example, \( r_{51} = r_e + r_q = 8.7 \times 10^{-8} \) esu at 1.064 \( \mu \). For \( \text{ZnO} \) at 5145Å, \( r_q \) is \( 2.04 \times 10^{-8} \) esu and \( r_e \) is \( 0.546 \times 10^{-8} \) (or \( 3.35 \times 10^{-8} \)) esu based on the Raman data of Arguello et al.\(^{39}\) and the refractive indices of Bond.\(^{35}\) At 6471Å, \( r_q \) is \( 1.69 \times 10^{-8} \) esu and \( r_e \) is \( 0.97 \times 10^{-8} \) (or \( 2.27 \times 10^{-8} \)) esu based on the Raman data of Callender et al.\(^{43}\)
Our far-infrared power measurements imply that the smaller value of $r_e$ is appropriate so that $r_{42} = 2.59 \times 10^{-8}$ esu at 5145Å. Fortunately, for this choice of $r_e$ the linear combination of $r_e$ and $r_q$ that determines the DFG susceptibility changes by only 2% between 5145 and 6471Å; thus, we can use this shorter wavelength data to estimate the DFG susceptibility at our dye laser wavelengths (8100 to 8400Å).

**B. Relative Merits of Forward-, Backward-, and Non-Collinear Phase Matching**

Since BCPM occurs at a larger angle $\theta$ than FCPM, DFG of a far-infrared o-ray in crystals of the point groups 4, 4, 4mm, 42m, 3m (when $\sin 3\phi = 0$), and 6mm has a larger effective nonlinear susceptibility, $d_{\text{eff}}$, for BCPM than for FCPM, as can be seen in Table III. For o-ray generation in 32, 6, and 6m2 crystals and e-ray generation in 3m, 6, and 6m2 crystals $d_{\text{eff}}$ is smaller for BCPM than for FCPM. For the other combinations of the optically uniaxial point groups and the generated polarization in Table III, the size of $d_{\text{eff}}$ for BCPM relative to $d_{\text{eff}}$ for FCPM at the same difference frequency depends on the nonlinear susceptibility tensor elements of the particular crystal and the angle $\phi$ at which it was cut, or it depends on the difference frequency through the factor $\cos \theta \sin \theta$ (e.g., e-ray generation in 4, 4, 422, 42m, 6, and 622 crystals). However, collecting the far-infrared radiation is more difficult in a BCPM than in a FCPM configuration; thus, even when it has a larger $d_{\text{eff}}$, BCPM will probably be less useful than FCPM in any far-infrared source based on difference frequency generation.
Noncollinear phase matching offers three practical advantages over collinear phase matching. First, it adds cubic crystals in point groups 43m and 32 to the list of phase matchable materials. Second, a single crystal sample can be used to cover a frequency range that would require several separately cut samples for collinear phase matching. And third, in some crystals, such as LiNbO$_3$, it provides a more efficient far-infrared source by eliminating the angular dependence of $d_{\text{eff}}$, using a larger nonlinear susceptibility tensor element than collinear phase matching, or using the far-infrared polarization with the smaller absorption coefficient. Nevertheless, in the final analysis each crystal must be evaluated independently to determine the best phase matching configuration for it.

C. Difference Frequency Generation as a Far-infrared Source

We have also verified that the dual-frequency dye laser system shown in Fig. 24 can be operated with a flashlamp-pumped Rhodamine 6G dye laser, although the laser output was insufficient to generate detectable far-infrared radiation in the difference frequency experiment. However, a flashlamp-pumped dye laser with 100 kW peak power (50 kW at each frequency) and a 1 µsec pulsewidth would generate the same far-infrared signal for each pulse as our Ruby laser-pumped system with its 600 kW peak power and 30 nsec pulsewidth. Because flashlamp-pumped dye lasers with 100 kW peak power can be operated at repetition rates above 1 Hz and beam expanding telescopes can be used to narrow the linewidth, difference frequency generation using the dual-frequency dye laser system of Fig. 24 is an attractive source for far-infrared
spectroscopy. With the same conversion efficiency as in our experiment, the system would generate a few nanoWatts of time-average far-infrared power at a 1 Hz repetition rate.

There are at least two opportunities for significant research in developing difference frequency generation as a far-infrared source: (1) a search for new and better nonlinear crystals with larger linear electro-optic coefficients, lower far-infrared absorption coefficients, and high damage thresholds; (2) an experiment with a less than 1 cm$^{-1}$ linewidth TEM$_{00}$ mode laser to verify the theory of Chapter II and establish the maximum attainable optical to far-infrared conversion efficiency as a function of input power for each crystal.
Table I. Summary of the mixing experiments on five different crystals.

<table>
<thead>
<tr>
<th>Crystals</th>
<th>Tunable range</th>
<th>Power (Frequency observed)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiNbO₃</td>
<td>20 to 127 cm⁻¹ (FCPM)</td>
<td>See Fig. 28</td>
</tr>
<tr>
<td></td>
<td>20 to 95 cm⁻¹ (BCPM)</td>
<td>≈ twice that of FCPM</td>
</tr>
<tr>
<td></td>
<td>40 to 160 cm⁻¹ (NCPM)</td>
<td>See Fig. 28</td>
</tr>
<tr>
<td>ZnO*</td>
<td>≤ 190 cm⁻¹ (FCPM)</td>
<td>14 mW (190 cm⁻¹)</td>
</tr>
<tr>
<td>CdS*</td>
<td>≤ 180 cm⁻¹ (FCPM)</td>
<td>3 mW (180 cm⁻¹)</td>
</tr>
<tr>
<td>ZnS*</td>
<td>≤ 91 cm⁻¹ (FCPM)</td>
<td>0.74 mW (91 cm⁻¹)</td>
</tr>
<tr>
<td>CdSe*</td>
<td>≤ 150 cm⁻¹ (FCPM)</td>
<td>&lt; 0.15 mW⁺ (150 cm⁻¹)</td>
</tr>
</tbody>
</table>

*Crystal thickness 1 mm
⁺Less than the detector noise level
Table II. Absorption Coefficients of LiNbO₃ (used for calculations in Chapter IV)

<table>
<thead>
<tr>
<th>Frequency</th>
<th>o-ray</th>
<th>Source</th>
<th>e-ray</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td></td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>.50</td>
<td>a</td>
<td>.31</td>
<td>a</td>
</tr>
<tr>
<td>10</td>
<td>2.1</td>
<td>a</td>
<td>1.25</td>
<td>a</td>
</tr>
<tr>
<td>15</td>
<td>5.54</td>
<td>a</td>
<td>2.8</td>
<td>a</td>
</tr>
<tr>
<td>20</td>
<td>8.68</td>
<td>a</td>
<td>5.0</td>
<td>a</td>
</tr>
<tr>
<td>25</td>
<td>12.5</td>
<td>a</td>
<td>7.5</td>
<td>a</td>
</tr>
<tr>
<td>30</td>
<td>17.1</td>
<td>a</td>
<td>11.0</td>
<td>a</td>
</tr>
<tr>
<td>35</td>
<td>49.6</td>
<td>a</td>
<td>13.0</td>
<td>a</td>
</tr>
<tr>
<td>40</td>
<td>110.</td>
<td>b</td>
<td>18.0</td>
<td>a</td>
</tr>
<tr>
<td>45</td>
<td>150.</td>
<td>b</td>
<td>23.0</td>
<td>a</td>
</tr>
<tr>
<td>50</td>
<td>200.</td>
<td>b</td>
<td>30.0</td>
<td>a</td>
</tr>
<tr>
<td>55</td>
<td>230.</td>
<td>b</td>
<td>39.0</td>
<td>a</td>
</tr>
<tr>
<td>60</td>
<td>300.</td>
<td>b</td>
<td>50.0</td>
<td>a</td>
</tr>
<tr>
<td>65</td>
<td>425.</td>
<td>b</td>
<td>70.0</td>
<td>a</td>
</tr>
<tr>
<td>70</td>
<td>460.</td>
<td>b</td>
<td>100.</td>
<td>a</td>
</tr>
<tr>
<td>75</td>
<td>480.</td>
<td>b</td>
<td>130.</td>
<td>a</td>
</tr>
<tr>
<td>80</td>
<td>500.</td>
<td>b</td>
<td>175.</td>
<td>a</td>
</tr>
<tr>
<td>85</td>
<td>520.</td>
<td>b</td>
<td>230.</td>
<td>a</td>
</tr>
<tr>
<td>90</td>
<td>530.</td>
<td>b</td>
<td>260.</td>
<td>a</td>
</tr>
<tr>
<td>95</td>
<td>545.</td>
<td>b</td>
<td>288.</td>
<td>a</td>
</tr>
<tr>
<td>100</td>
<td>610.</td>
<td>b</td>
<td>319.</td>
<td>a</td>
</tr>
<tr>
<td>105</td>
<td>720.</td>
<td>b</td>
<td>352.</td>
<td>a</td>
</tr>
<tr>
<td>110</td>
<td>845.</td>
<td>b</td>
<td>386.</td>
<td>a</td>
</tr>
<tr>
<td>120</td>
<td>610.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>125</td>
<td>1500.</td>
<td>c</td>
<td></td>
<td></td>
</tr>
<tr>
<td>140</td>
<td></td>
<td></td>
<td>1047.</td>
<td>c</td>
</tr>
<tr>
<td>160</td>
<td></td>
<td></td>
<td>1510.</td>
<td>c</td>
</tr>
</tbody>
</table>

a) D. R. Bosomworth, ref. 28
b) our nonlinear measurements
c) Barker and Loudon, ref. 26
Table III. Angular Dependence\textsuperscript{a} of Second-Order Nonlinear Susceptibilities for Type II Collinear Phase Matching in Uniaxial Crystals.

(a) Ordinary-Ray Generation

<table>
<thead>
<tr>
<th>Symmetry group</th>
<th>Effective nonlinear susceptibility\textsuperscript{b,c}</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>$d_{15}\sin\theta$</td>
</tr>
<tr>
<td>4</td>
<td>$(d_{14}\sin2\phi - d_{15}\cos2\phi) \sin\theta$</td>
</tr>
<tr>
<td>422</td>
<td>0</td>
</tr>
<tr>
<td>4mm</td>
<td>$d_{15}\sin\theta$</td>
</tr>
<tr>
<td>42m</td>
<td>$d_{14}\sin2\phi \sin\theta$</td>
</tr>
<tr>
<td>32</td>
<td>$-(d_{11}\cos3\phi + d_{22}\sin3\phi) \cos\theta + d_{24}\sin\theta$</td>
</tr>
<tr>
<td>3m</td>
<td>$-d_{22}\sin3\phi \cos\theta + d_{24}\sin\theta$</td>
</tr>
<tr>
<td>6</td>
<td>$d_{24}\sin\theta$</td>
</tr>
<tr>
<td>6mm</td>
<td>$d_{24}\sin\theta$</td>
</tr>
<tr>
<td>622</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>$-(d_{11}\cos3\phi + d_{22}\sin3\phi) \cos\theta$</td>
</tr>
<tr>
<td>6m2</td>
<td>$-d_{22}\sin3\phi \cos\theta$</td>
</tr>
</tbody>
</table>
Table III. Continued.

(b) Extraordinary-Ray Generation

<table>
<thead>
<tr>
<th>Symmetry group</th>
<th>Effective nonlinear susceptibility$^b,c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>$d_{14}\sin\theta \cos\theta$</td>
</tr>
<tr>
<td>4</td>
<td>$[(d_{14}+d_{36})\cos2\phi + (d_{15}+d_{31})\sin2\phi] \sin\theta \cos\theta$</td>
</tr>
<tr>
<td>422</td>
<td>$d_{14}\sin\theta \cos\theta$</td>
</tr>
<tr>
<td>4mm</td>
<td>0</td>
</tr>
<tr>
<td>42m</td>
<td>$(d_{14}+d_{36}) \cos2\phi \sin\theta \cos\theta$</td>
</tr>
<tr>
<td>3</td>
<td>$(d_{11}\sin3\phi - d_{22}\cos3\phi) \cos^2(\theta) + d_{14}\sin\theta \cos\theta$</td>
</tr>
<tr>
<td>3m</td>
<td>$d_{11}\sin3\phi \cos^2(\theta) + d_{14}\sin\theta \cos\theta$</td>
</tr>
<tr>
<td>3m</td>
<td>$-d_{22}\cos3\phi \cos^2(\theta)$</td>
</tr>
<tr>
<td>6</td>
<td>$d_{14}\sin\theta \cos\theta$</td>
</tr>
<tr>
<td>6mm</td>
<td>0</td>
</tr>
<tr>
<td>622</td>
<td>$d_{14}\sin\theta \cos\theta$</td>
</tr>
<tr>
<td>6</td>
<td>$(d_{11}\sin3\phi - d_{22}\cos3\phi) \cos^2(\theta)$</td>
</tr>
<tr>
<td>6m2</td>
<td>$-d_{22}\cos3\phi \cos^2(\theta)$</td>
</tr>
</tbody>
</table>

a) All walk-off angles have been neglected.
b) $\theta$ is the angle between the normal, \( \hat{n} \), and the axis of symmetry, \( \hat{3} \).
c) $\phi$ is the angle between \( \hat{2} \) and \( \hat{n} \times \hat{3} \).
REFERENCES

14. H. S. Pillof, Appl. Phys. Lett. 21, 15 (1972) has described a similar, independently developed system.
15. P. P. Sorokin et al. (ref. 2) have reported and we have also observed that longitudinally pumped DTTC iodide lasers lase with a linear polarization parallel to the linear polarization of a ruby pump laser.
22. During this absolute calibration both detectors were used without black polyethylene filters, the Golay cell was connected to the output light pipe of the Michelson interferometer with a length
of light pipe to match the experimental collection optics of the Ge:Ga detector (except the focusing cone), and the Ge:Ga detector output was fed directly into a lock-in amplifier instead of through the operational amplifier that we used for our difference-frequency measurements. Both detectors were operating significantly above their respective noise levels.

30. To obtain this curve we used the TO phonon oscillator strengths, frequencies, and linewidths of Barker and Loudon (ref. 26) and the Raman cross sections of Kaminow and Johnston (ref. 33) to compute $|\chi_{24}^{(2)}|^2$ (see refs. 31 and 32); the far-infrared refractive indices of Bosomworth (ref. 28) and of Barker and Loudon (ref. 26); and our experimental values of the far-infrared o-ray absorption coefficient.

ACKNOWLEDGEMENTS

I would like to thank my thesis advisor, Professor Y. R. Shen, for his expert guidance of this research and for his encouragement, especially at discouraging moments. I also wish to thank Professor P. L. Richards for sharing his vast knowledge of far-infrared physics and techniques and for the use of his experimental equipment.

I would especially like to thank K. H. Yang for his assistance in the mixing experiments with two dye lasers and for many fruitful discussions, especially about his picosecond pulse experiments and the calibration of the far-infrared detectors for our dye laser experiment. Patrick's knowledge of far-infrared measurement techniques was a key to the successful completion of our dye laser experiments. I would also like to thank the graduate and post-doctoral students -- especially A. J. Schmidt, M. Loy, and G. Wong -- in Professor Shen's group for many educational discussions about nonlinear optics.

I also wish to thank the following individuals and organizations at Lawrence Livermore Laboratory: T. Wainwright, my division head, and J. A. Fleck, Jr., my supervisor, for their patience with the time it has taken me to complete this thesis; R. L. Pexton for providing a complex error function and a double integration subroutine; M. D. Feit for his comments on Chapter III; and T Division for providing computer time.
And last, but certainly not least, I wish to thank my wife, Louise, for her love, understanding, and encouragement during this long project. This work was performed under the auspices of the U.S. Energy Research and Development Administration.
APPENDIX A

The extraordinary ray assumed in Sec. IIA of Chapter II actually has the form

\[ \hat{E}_2(r,t) = \frac{\xi_2 \hat{e}_2 \sqrt{1 + \xi_A^2}}{\sqrt{(1 + i \xi_{2x}) (1 + i \xi_{2y})}} \exp \left[-\frac{(x - z_0)^2}{w_2^2 (1 + i \xi_{2x})} - \frac{y^2}{w_2^2 (1 + i \xi_{2y})}\right] \]

\[ \exp[i(k_2 z - \omega_2 t)] \]  

(A1)

where

\[ \xi_\Delta = (z_0,2x - z_0,2y) \frac{4}{n_2^2 / k_2^2 w_{2n_{\text{em},2}}^2 (n_{o,2}^2 n_{o,2}^2)} \]

\[ \xi_{ex} = 2(z - z_0,2x) \frac{4}{n_2^2 / k_2^2 w_{2n_{\text{em},2}}^2 n_{o,2}^2} \]

\[ \xi_{ey} = 2(z - z_0,2y) \frac{4}{n_2^2 / k_2^2 w_{2n_{\text{em},2}}^2} \]

\[ \hat{e}_2 = \text{unit vector parallel to the electric field of the e-ray for a normally incident laser beam} \]

and the remaining parameters are as defined for Eq. (1). This expression with \( z_0,2x = z_0,2y \) is essentially the same as the one given in Appendix I of Ref. 31, but there the factors in the square brackets in the definition of \( \xi_{2x} \) and \( \xi_{2y} \) were approximated by 1.

The nonlinear polarization \( \hat{P}^{(2)}(\hat{r},\omega) \) is obtained from Eq. (2) using the expressions of \( \hat{E}_1 \) in Eq. (1) and \( \hat{E}_2 \) in Eq. (A1). The transverse Fourier transform of \( \hat{P}^{(2)}(\hat{r},\omega) \) gives \( \hat{P}^{(2)}(\hat{k}_1, z) \). To obtain
the expression of $\hat{P}^{(2)}(k, z)$ in Eq. (3), we made the following
simplifying assumptions. First, we assumed $w_1 = w_2 = w$. Second, we
assumed a common focus for the two beams, $z_{0,1} = z_{0,2x} = z_{0,2y} = z_0$.
Finally, we assumed $\xi_1 - \xi_{2x} = \xi_1 - \xi_{2y} = 0$. This last assumption is
reasonable as long as $3|\xi_1 - \xi_{2x}|, 3|\xi_1 - \xi_{2y}| \ll \pi/2$. In our
calculations, the largest value of $3|\xi_1 - \xi_{2x}|$ or $3|\xi_1 - \xi_{2y}|$ is 1 for
the case of $\omega = 10 \text{ cm}^{-1}, \omega = 25 \text{ cm}$ and $\zeta = -0.02$. For all the other
cases, $3|\xi_1 - \xi_{2x}|$ or $3|\xi_1 - \xi_{2y}|$ is much smaller than 1.
To derive Eq. (6), we first Fourier transform Eq. (4) and obtain

\[ [\mathbf{k} \cdot \mathbf{E}^r + \frac{2\omega^2}{c^2} \mathbf{E}^r - \omega^2 \mathbf{E}^r] \cdot \mathbf{E}(\mathbf{k}) = -4\pi (\omega^2/c^2) \hat{P}^{(2)}(\mathbf{k}) \]  

(B1)

\[ \mathbf{k} \cdot \mathbf{E} \cdot \mathbf{E}(\mathbf{k}) = -4\pi \mathbf{k} \cdot \mathbf{P}^{(2)}(\mathbf{k}) \]

The particular solution of Eq. (B1) can be written in the form

\[ \mathbf{E}^p(\mathbf{k}) = \mathbf{k} \cdot \mathbf{E} \cdot \mathbf{E}(\mathbf{k}) \]

From the inverse transform on \( k_z \), we then have

\[ \mathbf{E}^p(\mathbf{k}_z, z) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}k_z \int_{-\infty}^{\infty} \mathrm{d}z' e^{ik_z(z-z')} \mathbf{k} \cdot \mathbf{E} \cdot \mathbf{E}(\mathbf{k}) \cdot \mathbf{P}^{(2)}(\mathbf{k}_z, z') \]  

(B2)

A straightforward, but tedious, application of the residue theorem finally leads to Eq. (6).

We also notice that Eqs. (6) and (7) are not the results of slowly varying envelope approximation. This is in fact generally true for the solution of optical mixing in the parametric approximation. For example, consider the simple case where the nonlinear process can be described by the wave equation

\[ \left( \frac{\partial^2}{\partial z^2} + \frac{k^2}{c^2} \right) E(z) = 4\pi (\omega^2/c^2) P^{NL}(z) \]  

(B3)

where \( P^{NL}(z) \neq 0 \) only if \( 0 \leq z \leq \lambda \). Then, in the region \( 0 \leq z \leq \lambda \), the solution of the equation is
\[ E(z) = \frac{2\pi \omega^2}{ik_0 c^2} \left[ \int_0^z P_{NL}(z') e^{ik_0 (z-z')} dz' + \int_0^\lambda P_{NL}(z') e^{-ik_0 (z-z')} dz' \right] \quad (B4) \]

No slowly varying envelope approximation was made in the derivation. In fact, one can easily show that, with the complete expression of \( E(z) \) in Eq. (B4), the terms \( \partial^2 |E(z)|/\partial z^2 \) and \( 2k_0 \partial |E(z)|/\partial z \) are generally comparable in magnitude. The usual slowly varying envelope approximation is actually equivalent to neglecting waves propagating in the opposite direction.
APPENDIX C

Computation Methods for Chapter II

The results presented in section III of chapter II were obtained by numerically integrating Eq. (32) after substituting for $|\hat{E}_{T+}|^2$ the quantity $\left\{(T_{+}^{-} P_{+}) + (T_{+}^{-} P_{-})\right\}_{1,1} |E_{o+}^P|^2$ where the term in braces is the upper left matrix element of Eq. (30) evaluated for the special case $\hat{c} = \hat{x}$ and $E_{o+}^P$ is obtained from Eq. (22) for the special case $\hat{e}_{1} = \hat{y}$, $\hat{e}_{2} = \hat{x}$, $\chi(2) = yX_{\text{eff}}$:

$$E_{o+}^P(k_{T+}, \xi) = \frac{2\pi i \omega}{k_{oz}^2} \hat{c}_{\text{eff}} \chi_{\text{eff}}(2) \frac{w^2}{4} \exp(-\delta) Q(\gamma, \rho, \xi) \quad (C1)$$

where

$$\gamma = [(k_x - \zeta k_1)^2 + k_Y^2]/2k_1^2w^2$$

$$\rho = (2\gamma)^{-1}[-k_T^2z_0/k_1^2w^2 + a_v(k_x - \zeta k_1)/k_1w^2$$

$$+ \frac{1}{2} i\zeta k_x(1-2iz_0/k_1w^2) - i(k_1^2-k_2^2-k_{oz}^2)]$$

$$\delta = a_1^2/2w^2 + \frac{1}{8} w^2k_T^2(1 + 4z_0^2/k_1^2w^4)$$

$$+ \frac{1}{2} ik_xa_1(1+2iz_0/k_1w^2) + 2\text{Im}(k_{oz})$$

and

$$Q(\gamma, \rho, \xi) \equiv \exp(\gamma \rho^2) \int_0^\xi dz \exp[-\gamma(\rho+z)^2] \quad (C2)$$
\[ Q(\gamma, \rho, \ell) = \frac{1}{2} \sqrt{\pi/\gamma} \left[ W\left(i\gamma^{1/2}\rho\right) - \exp\{-\gamma \ell (2\rho + \ell)\} \right] W\left(i\gamma^{1/2}(\rho + \ell)\right) \]  

with \( W(\xi) = \frac{2}{\sqrt{\pi}} \int_{-i\xi}^{\infty} \exp(-u^2) du. \) [See W. Gautschi, SIAM J. Numer. Anal. 7, 187 (1970).]

Except for the factor \( |Q(\gamma, \rho, \ell)|^2 \), all of the quantities in the integrand are simple to evaluate numerically. The function \( Q(\gamma, \rho, \ell) \), however, has some mathematical properties that must be circumvented to successfully integrate Eq. (32) on a digital computer. The first of these properties is that although \( W(Z) \) is bounded and analytic in the upper half of the complex plane, it diverges in the lower half-plane as \( \text{Im}(Z) \to -\infty \). Such \( Z \) values can occur when \( Q(\gamma, \rho, \ell) \) is evaluated for laser beams that are focused behind the exit face of the nonlinear slab; fortunately, for such problems \( Q(\gamma, \rho, \ell) \) can be re-expressed as

\[ Q(\gamma, \rho, \ell) = \exp\{\gamma \ell (2\rho + \ell)\} \left[ Q(\gamma^*, -(\rho^* + \ell), \ell) \right]^*. \]

In this form the signs of the imaginary parts of the arguments of \( W(Z) \) are changed; by choosing this alternate form when \( \text{Re}(\gamma^{1/2}) \) and \( \text{Re}[\gamma^{1/2}(\rho + \ell)] \) are both negative, the magnitude of the largest negative imaginary part of an argument of \( W(Z) \) is reduced and often both of the \( W(Z) \) arguments are moved into the upper half-plane. The derivation of the above symmetry relation is straightforward:
\[ Q(\gamma, \rho, \ell) = \exp(\lambda\rho^2) \int_0^\ell \exp[-\gamma(\rho^2)z^2] \, dz \]

\[ = \exp[-\gamma(2\rho^2+\gamma^2(\rho^2+\ell^2))] \int_0^\ell \exp[-\gamma(z^2-(\rho^2+\ell^2))] \, dz' \]

\[ = \exp[-\gamma\ell(2\rho^2+\gamma^2(\rho^2+\ell^2))] Q[\gamma,-(\rho^2+\ell^2)] \]

\[ = \exp[-\gamma\ell(2\rho^2+\gamma^2(\rho^2+\ell^2))] \{Q[\gamma^*,-(\rho^2+\ell^2)] \}^* \]

There are two other parameter regions in which \(Q(\gamma, \rho, \ell)\) is difficult to evaluate on a digital computer. The first of these is the region in which both \(|\gamma^{1/2}|\) and \(|\gamma^{1/2}(\rho^2+\ell^2)|\) are very small. Because \(W(0) = 1\) and \(Q(\gamma, \rho, \ell)\) involves the difference between two values of \(W(z)\), Eq. (C.3) cannot be used to evaluate \(Q(\gamma, \rho, \ell)\) in this region on a digital computer with finite precision (14 significant digits on a CDC 7600). However, in the region defined by

\[ |\text{Re}(\gamma^{1/2})| + |\text{Im}(\gamma^{1/2})| + |\text{Re}[\gamma^{1/2}(\rho^2+\ell^2)]| + |\text{Im}[\gamma^{1/2}(\rho^2+\ell^2)]| < .001 \]

the series expansion \(Q(\gamma, \rho, \ell) \approx \ell[1 - \frac{1}{3} \gamma\ell(\ell^2+3\rho)]\) is accurate to better than 1 part in \(10^6\). (This region is simply one of many choices which keep the loss of significance [round-off error] and the inevitable difference between the series and the results of Eq. (C.3) [truncation error] at the boundary of the region to less than \(10^{-6}|Q(\gamma, \rho, \ell)|\).)

The final parameter region in which \(Q(\gamma, \rho, \ell)\) is difficult to evaluate is the region where \(\gamma^{1/2} \ell << 1\) but \(\gamma^{1/2}|\rho|\) is large. Again round-off error is the source of the problem and a series expansion is the solution. When \(\gamma\ell^2 << 1\), the exponential function in the integrand of Eq. (C.2) can be written \(\exp[-\gamma(\rho^2+\ell^2)] \approx \exp(-\gamma\rho^2) \exp(-2\gamma\rho\ell)(1-\gamma\ell^2+...)\).
When \( \gamma > 1/2 \), we evaluate \( Q(\gamma, \rho, \kappa) \) via the first term of the asymptotic expansion that is obtained from a term-by-term integration of Eq. (C.2) with \( \exp[-\gamma(\rho+z)^2] \) replaced by this series expansion:

\[
Q(\gamma, \rho, \kappa) \approx \frac{\exp(-2\gamma \rho \kappa) - 1}{-2\gamma \rho} \]

The double integral in Eq. (32) was re-expressed as an iterated integral with constant limits via the change of variables for the inner \((k_y)\) integral:

\[ v = k_y \left[ (\omega/c)^2 - k_x^2 \right]^{-1/2} \]

Both the inner and outer integrals were evaluated with an adaptive three point Gaussian quadrature algorithm. For each \( k_x \), the inner \((v)\) integral was adaptively refined until at least three levels of subdivision occurred and two successive estimates of its value differed by less than

\[ .001 \cdot [(\omega/c)^2 - k_x^2]^{-1/2} \int_{-\omega/c}^{\omega/c} dk_x \frac{c}{2\pi} \left( \frac{ck_x}{\omega} \right) |\hat{E}_{\gamma}(k_x^1, 0)|^2, \]

which was estimated with a trapezoidal rule on 100 intervals. The outer \( k_x \) integral was adaptively refined until two successive estimates differed by less than .1% of its initial estimate. The existence of a phase matched or Cerenkov cone whose angular width was in some cases much smaller than its opening angle at large far-infrared frequencies (like 100 cm\(^{-1}\) for a 1 cm long crystal) made this careful check necessary to be sure the peak of this hollow cone was sampled.

A listing of the computer program follows:
PROGRAM INFRAT1, INPUT, OUTPUT, TAPE2 = INPUT, TAPE3 = OUTPUT, TAPE6 = INPUT
1
C U M P L E X H3, H4, H5, H6, H7, Hb, H9, H10, H11, S M A L E, P 5
C U M P L E X E 5, E 6, E 7, E 8, E 9, E C 1, E C 2, E C 3, E C 4, AN 2, W A C
L U G I C A L P
C U M U N / G R K (11), X C G, Y C G, Z C G, W I D E, X T I L L, D F R E Q, F R G L, P 1, P 2, D K 0, H D P G,
1 A X I, P N L S (1), X N U L (2), X N E L (2), X U N O (39, 4), N T, X D O N E (39, 4), N T 1,
4 H 3, H 4, H 5, H 6, H 7, H 8, H 9, H 10, H 11, P 1, P 2, D K 0, H D P G,
5 A K X C
1 A N 2, W A C
C U M U N / C L N S T / C, P I, S P U R E, C C M P R (13)
C U M U N / A R A G 2 / L 2, U 2, E K, M A, N N, A I, H 1, E P, M P, N, H T
C U M U N / G H / H O R I Z (1000), V E R T (1000), V E R T (10000)
D I M E S S I O N P H S (12), P H 4 (2), P H 5 (2), P H 6 (2), P H 7 (2), P H S (2), P S M O N E (2)
E Q U I V A L E N C E (P H 3, H 3 J, P H 6, H 4), (P H 6, H 5), (P H 6, H 6), (P H 7, H 1 1),
1 (P H 5, H 5 J), (P S M O N E, S M O N E)
D A T A C, P I, C C M P R / 2.2, 97 92 25 * 10, 3, 1 4 1 5 9 2 7, S H P A R A M, S H N T B L U, S H N T B L E,
2 4 H L C G, 3 H E N D,
D A T A P S M O N E / U * 1 / D A T A I s * 0 / D A T A F I E L D 3 / 0.0 /
D I M E S S I O N P 1, 2, 3
H 1, H 2, ... C L A I R P H E C L M P O T E C VALUES TO BE USED BY R O U T I N E F I E L D
T H E S E V A L U E S D O N T D E P E N D O N A R X OR AKY AND T H U S I N F R
P O W E R G E N E R A T E D
R A Y O N T H E F A R S I D E O F T H E C R Y S T A L
C A L L D E V I C E (6, C R E A T E, 4, F C O U T, 5 0 0 0 0 C)
C A L L D E V I C E (6, C R E A T E, S H M P E R, 5 0 0 0 0, 0, 0 , M M )
N R = L U L (A K X C) - L U C (H O R K (1)) + 1
D 1 1 1 0 = 1, N R
W O R K (1) = 0.0
C U N R = P I / 1 8 0
C U N I W = 2. * P I * C
W R I T E (3, 2 0 0 1 ) N R, I, L C M P R
C A L L D D D O D D I D (10, H B O X, T 4 4 , J M , 1 )
C A L L K E E P ( 8 0 )
2 0 A S S I G N 2 5 T O M 1
A S S I G N 3 1 T O M 1
2 1 R E A D (2, 1 0 0 1 ) A N A M E, I S T A R, N R, W O R K ( I ) , I = 1,9
G O T O M 1
2 5 D O 3 0 I = 1, 1 3
I F ( A N A M E = C O M P R (11), I = 3 0, 3 5, 3 0
3 0 C O N T I N U E
A S S I G N 2 0 T O M 1
W R I T E (3, 1 0 0 1 )
3 5 W R I T E (3, 1 0 0 2 ) A N A M E, I S T A R, N R, W O R K ( J ) , J = 1,9
G O T O M 1
3 1 I F ( I = 2) 4 0, 1 3 C, 3 2
3 2 I F ( I = 3 ) 4 2, 2 3 0, 4 0
4 0 F R Q L = 2. * P I / W O R K (I) * ( 1. 0 5 + 8 ) * C
D F R E Q = C U N I * W O R K (2)
W I D E = W O R K (3)
P 1 = W O R K (4)
P 2 = W O R K (5)
X00 = W0RK(6)
Y00 = W0RK(7)
Z00 = W0RK(8)
XTLL = W0RK(9)
ISW = W0RK(10)
IF(NR=2) G04,46,20
40 ASSIGN 3 TO M1
ASSGN 47 TO M
GO TO 21
47 XNUL(1) = W0RK(1)
XNCL(2) = W0RK(2)*.5
XNFL(1) = W0RK(1) - W0RK(3)
XUP0 = W0RK(3)
D0 = W0RK(9)
XNFL(2) = W0RK(2)*.5
XX1 = W0RK(4)*1.E-6
PNL5(3) = SwRTIm0RK(5)**2+mWRK(6)**2+W0RK(7)**2
PNL5(1) = W0RK(9)/PNL5(3)
PNL5(2) = W0RK(6)/PNL5(3)
PNL5(3) = W0RK(7)/PNL5(3)
G0J = W0RK(8)
ISW = W0RK(10)*280
WJ TO 20
130 K = 3
J1 = 3*NR
ISW = W0RK(4)*E1
ASSGN 3 TO P1
ASSGN 131 TO M
131 DJ 132 I=1,3
L = K+1-4
IF(W0RK(3*I-2)) I55,136,135
135 X0NUL(1) = W0RK(3*I-2)*0N1W
X0NUL(2) = W0RK(3*I-1)
132 X0NUL(3) = W0RK(5*I-1)*.5
NT = J1
K = K+3
IF(IK-J1) 21,21,137
136 NT = L-1
137 WRITE(6) COMPR(2),NT,HCRLZ(1),I=1,19
CALL SPLICE(NT,X0NU(1,1),X0NG(1,1),X0NC(1,4))
WRITE(6) (X0NG(1,J),I=1,NT),J=1,4)
P = ,FALSE.
KK = 0
DO 138 I=2,NT
D0 138 J=1,5
KK = KK+1
U3 = X0NG(1,1)-X0NC(1,1,1)
U1 = .2*U3*FLCAI(1)
U2 = .2*U3*FLCAI(5-J)
HCRLZ(KK) = (X0NC(1,1,1)+U1)/0N1W
138 VERTIKK) = (X0NG(1,1,1)+U1)/0N1W
128 HO=RAY AB50RTI3 Cgef (CM-1)
GO TO 20
230 K = 3
J1 = 3*NR
ISW = W0RK(10)*I08
ASSGN 35 TO M1
ASSGN 231 TO M
231 DO 232 I=1,3
L = K+1-3

70 CALL FREQ
C NON-PHASE MATCHABLE FREQUENCIES HAVE THE TOTAL RADIATED POWER
C COMPUTED FOR THE OPTIC AXIS PARALLEL TO THE LASER BEAMS
70 CONTINUE
   DU 72 I=1,NR
   DFRQ = OLCH*CGN1m
   HORIZ(I) = OLQm
   CALL SETEPS
   CALL CRKENT
   CALL SETCON(I)
   VERT(I) = POWER(P)
   VERT(I+1) = PWER1
   72 DLQm = OLCH*STEP
   ASSIGN 74 TO M
   GU TO 168
79 CONTINUE
   CALL GRAPHNR,LUHP,M,FREQ.,
   1 17HFREQUENCY (1/CM),23HPower GENERATED (mWatts)
   GU TO 20
   C 80 CALL WIDTH
80 CONTINUE
   BN = DLQm
   DU 81 I=1,NK
   HORIZ(I) = BM
   CALL SETCON(I)
   VERT(I) = POWER(P)
   VERT(I+1) = POWER1
   81 BN = BM+STEP
   ASSIGN 89 TO M
   GU TO 168
89 CONTINUE
   CALL GRAPHNR,10SPOT SIZE,
   1 35H LASER BEAM WIDTH (E**-2 POINT) (CM),
   2 23HPower GENERATED (mWatts)
   GO TO 20
   C 90 CALL ABSRO
90 CONTINUE
   DU 91 I=1,NR
   HORIZ(I) = DLQm
   XDA = 0.5*OLQm
   CALL SETCON(I)
   VERT(I) = POWER(P)
   VERT(I+1) = POWER1
   91 DLQm = OLCHm+STEP
   ASSIGN 94 TO M
   GU TO 163
99 CONTINUE
   CALL GRAPHNR,10HABSORPTION
   1 23HABSORPTION COEF. (1/CM),23HPower GENERATED (mWatts)
   GC TO 20
   C 100 CALL ANGJL
100 CONTINUE
   P = .FALSE.
   CALL SETCON(I)
   AI = ABS(DFRQ/C)
   GU TO 20
   C 101 I=1,NR
   AKX = AI+5IN(OLQ11*CONDR)
   GU TO 161
   C AVOID SINGULARITY IN TERMS CALCULATED BY FIELD.
   IF(ABS(AKX)>1.1*0.006) AKX=1.06E-04
HORIZ(I) = DLOW1
VERT(I) = FIELD(P)*H1
VERT(I) = VERT(I)*ICFRQ**2/CON1H*DIRK
101 DLOW1 = DLOW1*STEP
ASSIGN 136 TO M
GO TO 168
10b CONTINUE
CALL GRAPHRK,10HPWR DIST ...
142HANGLE IN PLANE OF INC. (DEGREES) ,20HPWR PER STERADIAN) ..
AXK = 1.E-40
DO 105 I=1,NR
AKY = AI*Sin(DLCK*CCDK)
C AVOID SINGULARITY IN TERMS CALCULATED BY FIELD.
IF (ABS(AKY).LT.1.0E-40) AKY=1.0E-40
HORIZ(I) = DLOW
VERT(I) = FIELD(P)*H1
VERT(I) = VERT(I)*ICFRQ**2/CON1H*DIRK
103 DLOW = DLOW+STEP
ASSIGN 109 TO M
GO TO 168
109 CONTINUE
CALL GRAPHRK,10HPWR DIST ...
153HANGLE PERP. TO PLANE OF INC. (DEGREES) ,20HPWR PER STERADIAN)
GO TO 20
C 110 CALL TALK
110 CONTINUE
FRAXT = WORK(4)
XLN = DLOW
DO 111 I=1,NR
IF (ISTAK.EQ.11+*) XLCC = -XLA*FRAXT
CALL SETCN(I)
HORIZ(I) = XLN
VERT(I) = POWER(P)
VERT(I) = POWER1
111 XLN = XLN+STEP
ASSIGN 119 TO M
GO TO 168
119 CONTINUE
CALL GRAPHRK,10LENGTH ...
1 L1LENGTH (CM) ,23HPWR GENERATED (WATTS)
GO TO 20
C X DISPLACEMENT
140 CONTINUE
XO = DLOW
DO 145 I=1,NR
CALL SETCN(I)
VERT(I) = POWER(P)
VERT(I) = POWER1
HORIZ(I) = XO
145 XO = XO + STEP
ASSIGN 146 TO M
GO TO 168
146 CALL GRAPHRK,10+X-CISPLACE,22HBEAP CENTER SHIFT (CM),
1 23HPWR GENERATED (WATTS)
GO TO 20
C Y DISPLACEMENT
150 CONTINUE
CY2 = -1.
YO = DLOW
DO 155 I=1,NR
CALL SETCN(I)
VERT(I) = POWER(P)
155 YO = YO + STEP
ASSIGN 156 TO M
GO TO 168
156 CALL GRAPHN(R,ICPY-DISPLACE,22HBHEMT CENTER SHIFT (CM), 1
23HPWER GENERATED (WATTS) )
GO TO 20
160 CONTINUE
XLOC = ULOC
DC 161 I=1,NR
HORIZ(1) = XLOC
CALL SETCON(I)
VERT(1) = POWER(P)
VERT(1) = POWER(F)
161 XLOC = XLOC + STEP
ASSIGN 162 TO M
GO TO 168
162 CALL GRAPHN(R,10FFOCUS LOC.,23HFCAL PT. LOCATION (CM), 1
23HPWER GENERATED (WATTS) )
GO TO 20
120 WRITE(3,10031) I,15h
END FILE 6
CALL PNI((IBETA)
C GET FIELD LENGTH
C LOCATE BLOCK FOR IO#'S ETC. AS AN INDEX FOR ARRAY IBETA
IBETA = I+IBETA*M-IBETA(0)-260B
C LOCATE THE WORD CONTAINING THE FIELD LENGTH
IBETA = (IBETA(IBETA)+ANX777777)+LCC(IBETA(0))
C GET FIELD LENGTH
IBETA(2) = ISR((IBETA(IBETA)+ANX7777777)
C ROUND LENGTH TO THE NEAREST .GE. 512 WORDS
IBETA(2) = ISL((IBETA(IBETA)+7777777),5),5)
C SET-UP THE FILE NAME IN BETA WORKS FOR FCN CALL
C THE LEADING BLANKS ARE NECESSARY - R FORMAT DOES NOT SUPPLY THEM
IBETA(1) = 10h
C SWITCH THE FILE NAME FROM CHIP DISPLAY CDE TO ASCII
CALL SWITCH(HCETA,IBETA,1)
CALL GUB(12008,BER,0,IBETA)
CALL DEVICE(16HCLOSER,5hGFILE)
CALL PLOUE
CALL EXIT
C ROUTINE TO PRINT THE CALCULATED CURVE PRIOR TO CRT PLOTTING
168 WRITE(3,1007) DO 169 I=1,NR,8
K = I+7
IF(K.GT.NR) K=NR
WRITE(3,10055) HORIZ(I),(VERT(J),J=I,K)
169 CONTINUE
WRITE(3,1007)
WRITE(6) COMPX(1PLT),NK=HORIZ(1),STEP,XO,YO,XLOC,BH,P1,P2,
1 FKOL,X1,PKL,X1X,XL,DFR,J,NDP,DK,
2 XDN,XCA,XDM,XEDA
WRITE(6) (VERT(I),I=1,NR)
IF(T.NCT,P) GC TC M
DO 170 I=1,NR,8
K = I+7
IF(K.GT.NR) K=NR
WRITE(3,10055) HORIZ(I),(VERT(I),I=I,K)
170 CONTINUE
WRITE(3,1007)
WRITE(6) (VERT(I),I=1,NR)
GO TO M

1000 FORMAT(AS, A1, 12, 9FU.1)           INF 3730
1001 FORMAT (17H CAKE JECHECK - 1)       INF 3750
1002 FORMAT (1H, A1, 11X, 12, 9E12.4)     INF 3760
1003 FORMAT (/// 11H END-OF-JOB 215/1H)           INF 3770
1004 FORMAT (15H STEP SIZE IS ,E11.4, 17H PROCEDURE IS ,A5/) INF 3780
1005 FORMAT (E12.4,8E13.4)        INF 3790
1007 FORMAT(/)                         INF 3800
2001 FORMAT(1220,9(2X,A5)/4CX,4(2X,A5)) INF 3810

END

* FORTRAN PL/ER

FUNCTION POWER(Q,w)

COMPLEX H3,H4,H5,H6,H11,SMCNE,H55

CJMPFLX E<.E,ECCZ,EOCX,EGS,ECEE1,E,ECCZ,EECZ,ANZ,MAC

LOGICAL P

COMMON WORK(I,1),XCC,YOC,ZO00,WIDE,XTLL,DREC,FRQL,PI,P2,UKD,WPOD,
XX1,PI:15(3),XN11(2),XN12(2),XN13(4),NT,XNNE(I5),VT1

2 CUT,XU,YY,XXD,LXN,DREC,CX,CP,XDN,XDNA,XEY,SEN,

3 CL.WXCFH,NR,STEP,ANAME,ISTAR,H1,H2,H2P,H2PP,H2PPP,

4 H3,H4,H5,H6,H7,H8,H9,H10,H11,H12,H13,H14,H411,CHR,SO,

5 AKX

COMMON / BC/ AKV,AKVl,FKCS,EG,ECCZ,EOCX,EGS,ECEE1,E,ECCZ,EECZ,

6 ANZ,MAC

COMMON / CCN01/ CP1,SYKE,CCPW(13)

COMMON / ARAG02/ C62,D62,ER,MM,NN,AN1,AN1,EP,MP,Nm

COMMON / PASS/ FIELS3,AS62,PLFR1,AKX,AKY,DIRK,P

COMMON / GRPH/ MORIZE(I0,0J),VERT(I00),VERT(I100)

DATA C62,C62,ER,MM,NN,EP,MP,N .0...L=3:0;20,1.E=3:0;20/

1 J= Ad3(DFRQ*(CLT))

AL = -BJ

MPREV = MM

M4 = 0

NEST1 = 2*WEST2

WEST = NEST1+1

AKY = 0.

AKX = AL

OAK = (H1-A1)/FLGAT(NEST)

WEST = .5*FIELD(P)

D(1) 100 1=1NEST1

AKX = AKX+OAK

100 WEST = UEST + FIELD(P)

AKX = BI

UEST = .5*(UEST + .5*FIELD(P))

EH = ERS*UEST

5 CON = C/(PI*(D62-D62))#1

POWER = CON*AGIC(CQ)

IF(MPREVNE.MM) WRITE(3,1000) MM

1000 FORMAT(2X,17,2F-PLAICTIC EVALUATIONS

RETURN

END

FUNCTION FAC(N)

COMPLEX H3,H4,H5,H6,H11,SMCNE,H55

COMPLEX ECCZ,EOCX,EGS,ECEE1,E,ECCZ,EECZ,ANZ,MAC

LOGICAL P

COMMON WORK(I),XCC,YCC,ZO00,HI,DREK,FRQL,PI,P2,UKD,WPOD,
XX1,PI:15(3),XN11(2),XN12(2),XN13(4),NT,XNNE(I5),VT1

2 CUT,XU,YY,XXD,LXN,DREC,CX,CP,XDN,XDNA,XEY,SEN,

3 OLWkX,OHIGH,NN,STEP,ANAME,ISTAR,H1,H2,H2P,H2PP,H2PPP,

4 H3,H4,H5,H6,H7,H8,H9,H10,H11,H12,H13,H14,H411,CHR,SO,

5 AKX

COMMON / BC/ AKV,AKVl,FKCS,EG,ECCZ,EOCX,EGS,ECEE1,E,ECCZ,EECZ,
1 (PH55,H55),(PSPLAE,SMUAE)
YMAX = 0.
U) 10 I=1,NG
YMAX = AMAX1(YMAX,VERT(I))
IF(P) YMAX = AMAX1(YMAX,VERT(I))
10 CONTINUE
CALL MPS(HORIZ(1),HORIZ(NG),0.,YMAX)
CALL SETCH(1.,1.,1.,0.,3.,0.,0.)
CALL LRTBCD(NG)
CALL SETCH(32.,1.,0.,3.,0.,0.,0.)
CALL LRTBCD(RX)
CALL SETCH(1.,32.,0.,0.,2.,1.,0.)
CALL CRTBCD(KY)
CALL TRACE(HORIZ,VERT,NC)
IF(P) CALL TRACE(HORIZ,VERT,NG)
CALL SETCH(65.,33.,1.,1.,0.,0.)
FRUD = UFRO/12.*PI*CI
FRRU = UFRO/12.*PI*CI
WRITE (100,100), P1,P2,DX,YO,XLOC,XLN,KGp,DK,XNOL,
1 FORMAT(HF1) = E11.3/6FP2 = E11.3/6HFw = E11.3
100 FORMAT(HF1) = E11.3/6FP2 = E11.3/6HFw = E11.3
1/10FI5590
1/10FI5600
1/10FI5610
1/10FI5620
1/10FI5630
1/10FI5640
1/10FI5650
1/10FI5660
1/10FI5670
1/10FI5680
1/10FI5690
1/10FI5700
1/10FI5710
1/10FI5720
1/10FI5730
1/10FI5740
1/10FI5750
1/10FI5760
1/10FI5770
1/10FI5780
1/10FI5790
1/10FI5800
1/10FI5810
1/10FI5820
1/10FI5830
1/10FI5840
1/10FI5850
1/10FI5860
1/10FI5870
1/10FI5880
1/10FI5890
1/10FI5900
1/10FI5910
1/10FI5920
1/10FI5930
1/10FI5940
1/10FI5950
1/10FI5960
1/10FI5970
1/10FI5980
1/10FI5990
1/10FI6000
1/10FI6010
1/10FI6020
1/10FI6030
1/10FI6040
1/10FI6050
1/10FI6060
1/10FI6070
1/10FI6080
1/10FI6090
1/10FI6100
1/10FI6110
1/10FI6120
1/10FI6130
1/10FI6140
1/10FI6150
1/10FI6160
1/10FI6170
1/10FI6180
1/10FI6190
1/10FI6200

SUBROUTINE SPLICE(NP,XT,YT,SP)
C
C GENERATION OF THE SPLINE INTERPOLATION COEFFICIENTS
C
C NP IS THE NUMBER OF (XT,YT) PAIRS
C XT IS THE IRA OF TABULATED INDEPENDENT VARIABLE ARRAY
C YT IS THE IRA OF TABULATED DEPENDENT VARIABLE ARRAY
C SP IS THE ARRAY OF SPLINE COEFFICIENTS
C
PARAMETER NI SHOULD BE SET TO MAXIMUM NP TO BE USED
BLOCK SSSS WILL THEN BE DIMENSIONED FOR 2N1(N1+2) WORDS
C
DIMENSION XT(2),YT(2),SP(2)
CUM4GN/SSS5/40(40,40),R(40,40),Y(40),S(40),T(40),V(40)
C
NT = NP
W(1,1) = 1./3.
W(1,2) = 1./6.
Y(I) = (YT(I-2)-YT(I))/(XT(I-2)-XT(I))*W(I) +
W(NT,NT-2) = 1./(XT(NT-1)-XT(NT-2))
W(NT,NT-1) = 1./(XT(NT)-XT(NT-2)+1./(XT(NT)-XT(NT-1))
Y(NT,NT) = 1./(XT(NT)-XT(NT-1))
Y(NT,NT) = 0.
NTP=NT-1
30 K=1,NTP
W(K,K+1) = (XT(K)+1-XT(K))/6.
W(K,K+1) = (XT(K)-XT(K+1))/6.
Y(K) = (YT(K+1)-YT(K))/(XT(K+1)-XT(K))-(YT(K)-YT(K-1))/(XT(K)-
1 XT(K-1))
CALL MLR(40,NT,AT,K,Y,SP,B,3,T,V)
RETURN
IF (ABS(G) .LT. 1.1E-10) GC TO 59
C FUDGE CRYSTAL ORIENTATION TO ELIMINATE POLARIZATION
C DEPENDENT EFFECTS FOR NCN-ZERO WALK-CFF ANGLES. FOR LARGE
C WALK-OF-ANGLE ANGLES THE E-RAY IS NEARLY PHASE-MATCHED AT LOW
C FREQUENCIES IN WHICH CASE THIS IS A BETTER APPROXI-
C MATION. FOR LARGE FREQUENCIES WHERE THE ORIEN-
C TATION ANGLE IS LARGE
C THE EFFECT OF THIS CHANGE SHOULD BE SMALL.
XORS = 1.
XORC = 0.
XNEL1 = XNUL1*SCRT(G+1.1)
GO TO 60
59 XORS = 1.
XORC = 0.
XNEL1 = XNCL1(1)
60 WRITE (13,1007) XNEL1(1),G,XORS
1007 FORMAT (31H OPTICAL E-RAY INDEX OF REF. = +E12.5,5H G = +
1 E13.5,9H X(.JC) = +E13.5)
RETURN
END
SUBROUTINE SETCEN(KKK)
C COMPLEX H3,H4,H5,H6,H11,SMCNN,H55
C COMPLEX HD,EE,EECZ,EUCC,EEZ,EECX,AN2,WAC
LOGICAL P
COMMON XCRK(11),XCCG,YOC,ZOC,XXL,T,U,LH,C,CDF,R,Q,PL,P2,DKO,WOPQ,
1 XX1,PNLS(3),XNOL(2),XNEL(2),XNUL(34,41),NT,XDNE(39,41),NTL,
2 CUX,YU,YLOC,EY, XLN,DFRQ,DK,XAD,/XDA,XEDN,XEN,
3 OLLW,DMH,NR,STEP,ANAME,ISTAR,H1,H2,H2P,H2PP,H2PPP,
4 H3,H4,H5,H55,H6,H7,H6,H9,H11,H12,H13,H14,XORS,XORC,
5 AKX
COMMON /BC/ AKV,AKVI,FKCS,ECC,ECCX,EOS,ECEE1,E11,EEX,EECX,
1 AN2,MAC
COMMON /CONST/ C,P1,SMONE,CCPR(13)
COMMON /ARGAB/ CG2,UG2,ER,WH,NN,AN,ANL,EP,MP,N,W,WT
COMMON /PASS/ FILLU3,A3G0,PLENRT,AYX,AKY,DIRK,P
COMMON /GPRH/ HURLI(1000),VREI(1000),VERTI(1000)
DIMENSION PH3(2),PH4(2),PH5(2),PH6(2),PH11(2),PH5S(2),PHSOME(2)
EQUIVALENCE (PH3,H3),(PH4,H4),(PH5,H5),(PH6,H6),(PH11,H11),
1 (PH5S,H5S)
1 (PHSOME,PSCONE,SPSNE)
AKV = UFRQ/C
AKVI = 1./AKV
FKQS = AKV*AKV
H1 = (P1*XX1*(DFRQ/C)**2*4.1C)**2*PI*PI+1.E+19
H2 = -25*(BW**2+2*(XLOC*C*(BW*DFRQ)**1)**2)
T7 = C/DFRQ+4**2)
H2 = -1*X0*X0*Y0*Y0+(8k*8k)
H2PP = -2.*T7*X0*Y0
H2PPP = -2.*T7*Y0*X0
H5P = Y0*T7*XNOL(1)
H7 = XLOC*T7*C/XNOL(1)*FRQL
H9 = T7*5.*C/DFRQ*XNUL(1)*2)
AK = XNDRFCRG/C
H11 = CMLXTAR*XLA
H12 = H11*CMLPLX(REAL(H11),A,IMAG(H11))
H4 = H11*C/ABS(CFRQ)
H3 = H11*H11
H4 = (C/CFRQ)**2
EL = CMLPLXEDN,XEDC/C/ABS(CFRQ)
EE = EE*EE
EO = H3*H14
EOE = EO*EO
RETURN
END
SUBROUTINE SETCEN(KKX
C COMPLEX H3,H4,H5,H6,H11,SMCNN,H55
C COMPLEX HD,EE,EECZ,EUCC,EEZ,EECX,AN2,WAC
LOGICAL P
COMMON XCRK(11),XCCG,YOC,ZOC,XXL,T,U,LH,C,CDF,R,Q,PL,P2,DKO,WOPQ,
1 XX1,PNLS(3),XNOL(2),XNEL(2),XNUL(34,41),NT,XDNE(39,41),NTL,
2 CUX,YU,YLOC,EY, XLN,DFRQ,DK,XAD,/XDA,XEDN,XEN,
3 OLLW,DMH,NR,STEP,ANAME,ISTAR,H1,H2,H2P,H2PP,H2P,
4 H3,H4,H5,H55,H6,H7,H6,H9,H11,H12,H13,H14,XORS,XORC,
5 AKX
COMMON /BC/ AKV,AKVI,FKCS,ECC,ECCX,EOS,ECEE1,E11,EEX,EECX,
1 AN2,MAC
COMMON /CONST/ C,P1,SMONE,CCPR(13)
COMMON /ARGAB/ CG2,UG2,ER,WH,NN,AN,ANL,EP,MP,N,W,WT
COMMON /PASS/ FILLU3,A3G0,PLENRT,AYX,AKY,DIRK,P
COMMON /GPRH/ HURLI(1000),VREI(1000),VERTI(1000)
DIMENSION PH3(2),PH4(2),PH5(2),PH6(2),PH11(2),PH5S(2),PHSOME(2)
EQUIVALENCE (PH3,H3),(PH4,H4),(PH5,H5),(PH6,H6),(PH11,H11),
1 (PH5S,H5S)
1 (PHSOME,PSCONE,SPSNE)
AKV = UFRQ/C
AKVI = 1./AKV
FKQS = AKV*AKV
H1 = (P1*XX1*(DFRQ/C)**2*4.1C)**2*PI*PI+1.E+19
H2 = -25*(BW**2+2*(XLOC*C*(BW*DFRQ)**1)**2)
T7 = C/DFRQ+4**2)
H2 = -1*X0*X0*Y0*Y0+(8k*8k)
H2PP = -2.*T7*X0*Y0
H2PPP = -2.*T7*Y0*X0
H5P = Y0*T7*XNOL(1)
H7 = XLOC*T7*C/XNOL(1)*FRQL
H9 = T7*5.*C/DFRQ*XNUL(1)*2)
AK = XNDRFCRG/C
H11 = CMLXTAR*XLA
H12 = H11*CMLPLX(REAL(H11),A,IMAG(H11))
H4 = H11*C/ABS(CFRQ)
H3 = H11*H11
H4 = (C/CFRQ)**2
EL = CMLPLXEDN,XEDC/C/ABS(CFRQ)
EE = EE*EE
EO = H3*H14
EOE = EO*EO
RETURN
END
$A_K = W_O P$

$A_N = 1.0 / (W_O S * # 2 / L E * X_O R C * # 2 / L O )$

$E_U E_1 = A_N * A_N * E_E_1 / E_C$

$W_A C = ( E_E_1 - 1 / E_C * X_O R S * X_G R C * A_N)$

$E_O C_X = E_C * X_O R S$

$E_U C_L = E * X_O R C$

$E_E C_X = E * X_O R S$

$E_L U = E * X_O R C$

$A_K X C = T / X_U L C / H_2 / A_M A X_1 (A_B_S (X_O), A_B_S (X_O + A_R * X_L N), A_B_S (Y_0))$

$H_S = A_R * T / X_N C L (1)$

$H_L U = 0.5 * (A_R / E _ B)^2$

$H_S S = A_K * (0.5 * S_M_O N E - T / T * X_L O C)$

$H_S = H_S S - X_O / T / X_N C L (1)$

$T _ N E L = T H E _ E X T R_A L _ A R R A W _ I N D E X _ C F _ R E F R A C T I O N _ F O R _ A _ W A V E$

$P K U P _ P A G I N G _ A T _ A N _ A N G L E _ K C / S I N (X_K C) _ F R O M _ T H E _ O P T I C _ A X I S$

$F O R _ T H E _ C A S E _ O F _ N E G L E G I B L E _ A b S o R P _ T I O N.$

$T _ N E L _ = S W_R T (1 / (W_O S / X_N C L (1) * # 2 / X_C R C / X_N C L (1) * # 2 / 1))$

$K = D _ K + K E A L _ (11)$

$A I = X_N O L (2) * X_N E L (2) + X_O / A R / (E _ B / X_W_B)$

$H _ O = C M P L X (K * A I)$

$H _ L I = - (P _ N L S (1) / X_O R C + P_N L S (3) / X_C R C)$

$H _ L I = H_L * (4 / (X_N C L (1) - 1) * (T _ N E L + 1) * # 2)$

$F L F K _ K K _ . L E _ . R E T U R N$

$K = D _ K + K E A L _ (11)$

$A I = X_N O L (2) * X_N E L (2) + X_O / A R / (E _ B / X_W_B)$

$H _ O = C M P L X (K * A I)$

$H _ L I = - (P _ N L S (1) / X_O R C + P_N L S (3) / X_C R C)$

$H _ L I = H_L * (4 / (X_N C L (1) - 1) * (T _ N E L + 1) * # 2)$

$F L F K _ K K _ . L E _ . R E T U R N$

$K = D _ K + K E A L _ (11)$

$A I = X_N O L (2) * X_N E L (2) + X_O / A R / (E _ B / X_W_B)$

$H _ O = C M P L X (K * A I)$

$H _ L I = - (P _ N L S (1) / X_O R C + P_N L S (3) / X_C R C)$

$H _ L I = H_L * (4 / (X_N C L (1) - 1) * (T _ N E L + 1) * # 2)$

$F L F K _ K K _ . L E _ . R E T U R N$

$K = D _ K + K E A L _ (11)$

$A I = X_N O L (2) * X_N E L (2) + X_O / A R / (E _ B / X_W_B)$

$H _ O = C M P L X (K * A I)$

$H _ L I = - (P _ N L S (1) / X_O R C + P_N L S (3) / X_C R C)$

$H _ L I = H_L * (4 / (X_N C L (1) - 1) * (T _ N E L + 1) * # 2)$

$F L F K _ K K _ . L E _ . R E T U R N$

$K = D _ K + K E A L _ (11)$

$A I = X_N O L (2) * X_N E L (2) + X_O / A R / (E _ B / X_W_B)$

$H _ O = C M P L X (K * A I)$

$H _ L I = - (P _ N L S (1) / X_O R C + P_N L S (3) / X_C R C)$

$H _ L I = H_L * (4 / (X_N C L (1) - 1) * (T _ N E L + 1) * # 2)$

$F L F K _ K K _ . L E _ . R E T U R N$

$K = D _ K + K E A L _ (11)$

$A I = X_N O L (2) * X_N E L (2) + X_O / A R / (E _ B / X_W_B)$

$H _ O = C M P L X (K * A I)$

$H _ L I = - (P _ N L S (1) / X_O R C + P_N L S (3) / X_C R C)$

$H _ L I = H_L * (4 / (X_N C L (1) - 1) * (T _ N E L + 1) * # 2)$

$F L F K _ K K _ . L E _ . R E T U R N$
EQUIVALENCt (PT2, R3), (PT2(2), S3)
DIMENSION PAKZEP(2), PAKZEM(2)
EQUIVALENCt (PT1, R4), (PT1(2), S4)
EQUIVALENCt (PAKZEP, R5), AKZEP(2), (PAKZEP(2), S5)
EQUIVALENCt (PAKZEM, R6), AKZEM(2), (PAKZEM(2), S6), (PT2, R3), (PT2(2), S3)
COMPLEX *OFZ
ICHAT = ICNT+1
DEBUG = (ICHAT. LE. &1)
J = AKX**2
AKX = U*AUXS
DINX = SQRT(1. - V**H14)
C T1 = THE Z COEFFICIENT CF K IN THE CRYSTAL
AK2UP = CSQRT(T1 - V)
C T2 = 2.*GAMMA*ETA
T2 = SMDE. *(T1 - Mo) + PT - V5 - H5*AUX + H5*AKX
IF (DEBUG) WRITE(13, 23011) L, V, PT1, PT2
C T3 = GAMMA
T3 = HE*AUX + H9*V + H10
C T5 = GAMMA**.5
T5 = SQRT(T15)
C T6 = GAMMA**.5*ETA
T6 = T2/(T2. + T3)
T10 = 2*P + H2**P*AUX + H2**P*AUX - 2.*U*IMAG(AK2CP)*XN
C FIELD = THE SQUARE OF THE MODULUS OF THE FIELD GENERATED
C AT THE EXIT OF THE CRYSTAL W/ BOUNDARY CONDITIONS
C (MATCHING LINEAR MEDIA) AND W/O THE EXP ABSORPTION TERM
IF (DEBUG) WRITE(13, 23041) T3, T4, T10
DEL = XLN*T3
IF (ABS(DEL). LT. .0011) CC TL 200
FIELD = T1/(4.*(REAL(T3)**2 + IAMAG(T3)**2))
T3 = DEL + T4
IF (SHIFT(1R1. ANG. K2. ANG. 4.0000000000000000001) .LT. EQ11) 3CO, 400
200 Tu = (1. - EXP(-K3*XLN))**CMLX(CCS1S3*XLN), -SIN(S3*XLN))/T11*T22
IF (ABS(S1) + ABS(S2) + ABS(R1) + ABS(R21)), LT. 1.E-3
1 T6 = XLN*(1. - DEL*(T4 + 33333333333**3)*DEL)) / T1
FIELD = EXP*(2*V+I10)*2*UAMAG(T6)**2
50 TO 210
300 T10 = T10 - 2.*DEL*DEL*+2.*REAL(T4))
T6 = -CUHJG(T3)
T3 = -CUNJG(T4)
T4 = T6
400 FIELD = FIELD*EXP*(H2*V+I10)
T20 = (T4 - T3)*T3 + T4
T6 = (KOFZ(-S1.R1)) - EXP(T21)**CMLX(CCS1S2**T22), SIN(T22))
1. + KOFZ(-S2.R21)/T1
IF (ABS(S31) + ABS(S2) + ABS(R1) + ABS(R21)), LT. 1.E-3
1 Tu = DEL*(T4 + 3333333333333**3)*DEL)) / T1
FIELD = FIELD*(REAL(T6)**2 + IAMAG(T6)**2)
210 IF (DEBUG) WRITE(3, 23041) K2, S2, R1, S1, T6
IF (DEBUG) WRITE(3, 2307) T3, T4, FIELD
TOP1 = XGHS*AUX
TOP2 = -XGCR*AUX
TOP3 = XGUR*AUX
OP(2) = TOP3 - XGHS*AUKDP
GPL = 1./CSEXIT(TCP2(2)*OP2) + U)
OP(1) = LPL**TCP2
OP(2) = OP1**TCP
OP(3) = GPL**TCP1
AKVP = AKY*CIRK
AKT = SQRT(V)
AKTV(1) = AKX/AKX
52 TEMP=R1 $ TEMM=R2 $ GO TO 55
58 TEMP=S1 $ TEMM=S2
59 IF(Y.EQ.0.) 54,5
55 RE=EXP(-X*X) $ GO TO 6
9 KE=1.1283791670551*TEMP
6 IM=1.1283791670551*TEM
IF(Y.LT.0.) MUZ = -CMPLX(IM*IM, IM*IM) + 2.*EXP(Y+Y-X*X) + Z*EXP(Y+Y-X*X)
1 CMPLX(IM*IM, IM*IM) + 2.*EXP(Y+Y-X*X)
IM = IM*SIGN(1.,U)
RETURN
END
APPENDIX D

The plane wave square pulse laser fields are given by

\[ \tilde{E}_p(z,t) = \sum_p \tilde{E}_{\lambda p}(z,t) \text{ with} \]

\[ \tilde{E}_{\lambda p}(z,t) = \tilde{E}_{\lambda p} \cos[\omega_0(t-zn_p/c)] \quad 0 < t-zn_p/c < \tau \]

If we make the simplifying assumptions that only the beating of optical polarizations \( j \) and \( k \) to generate far-infrared polarization \( i \) has non-zero tensor elements and that the optical pulses still overlap at \( z = \lambda \), then the solution to Eq. (42) is

\[ \tilde{E}_i(t,\lambda) = (1-R_i) \sum_{m=0}^{\infty} \left[ E_i(s)(t_m, z=\lambda) - R_i E_i(s)(t_m - n_i \lambda/c, z=0) \right] \]

with \( t_m \equiv t-m(2n_i \lambda/c) \),

\[ E_i(s)(t, z=\lambda) = \]

\[ 0 \quad \text{for } t_r < t_j \]

\[ -E_i(n_i-n_j)^{-1} \cos(\omega_j t_r) \quad t_j < t_r < \min(t_i, t_k+\tau) \]

\[ 0 \quad t_i < t_r < t_k+\tau \]

\[ -E_i \left\{ (n_i-n_j)^{-1} \cos(\omega_j t_r) \right\} \quad t_k+\tau < t_r < t_i \]

\[ \quad -E_i \left\{ -(n_i-n_k)^{-1} \cos(\omega_k(t_r-\tau)) \right\} \quad \max(t_i, t_k+\tau) < t_r < t_i+\tau \]

\[ \tilde{E}_i(n_i-n_k)^{-1} \cos(\omega_k(t_r-\tau)) \quad t_i+\tau < t_r , \]
and

\[ E_{1}^{(s)}(t, z=0) = \]

\[
\begin{align*}
0 & \quad t < 0 \\
-\tilde{E}_{1} \left( n_{i} + n_{j} \right)^{-1} \cos(\omega_{j} t) & \quad 0 < t < \min(\tau, t_{i} + t_{j}) \\
0 & \quad t_{i} + t_{j} < t < \tau \\
-\tilde{E}_{1} & \left\{ \left( n_{i} + n_{j} \right)^{-1} \cos(\omega_{j} t) \right\} \left\{ -\left( n_{i} + n_{k} \right)^{-1} \cos[\omega_{k} (t-\tau)] \right\} \\
\tilde{E}_{1} \left( n_{i} + n_{k} \right)^{-1} \cos[\omega_{k} (t-\tau)] & \quad \tau < t < t_{i} + t_{j} \\
0 & \quad \max(\tau, t_{i} + t_{j}) < t < t_{i} + t_{k} + \tau \\
 & \quad t_{i} + t_{k} + \tau < t
\end{align*}
\]

where

\[ E_{1} \equiv \frac{8\pi}{n_{i}^{+NL}} \tilde{E} \tilde{E}_{1} \tilde{E}_{k} \tilde{E}_{k} (1-\delta_{jk}/2) \]

\[ \omega_{p} \equiv \omega \frac{(n_{i} - n_{k})}{n_{i} + n_{p}} \quad (p = j \text{ or } k) \]

\[ t_{r} \equiv t - n_{i} \ell/c \]

\[ t_{q} \equiv n_{q} \ell/c \quad (q = i, j, \text{ or } k) \]

\[ n_{i} \equiv n_{i}(\omega); \quad n_{j} \equiv n_{j}(\omega); \quad n_{k} \equiv n_{k}(\omega) \]

and \( n_{i} > n_{j} \geq n_{k} \) has been assumed.
APPENDIX E

Determination of the Least Squares Fitting Parameters $\hat{a}$ and Their Covariance Matrix

The minimization of $\chi^2$ was based on a local linearization of

$$P(\omega, \theta, \hat{a})$$

on the three components of $\hat{a}$:

$$P(\omega, \theta, \hat{a}_{n+1}) \approx P(\omega, \theta, \hat{a}_n) +$$

$$\nabla_{\hat{a}} P(\omega, \theta, \hat{a}_n) \cdot (\hat{a}_{n+1} - \hat{a}_n).$$

When $\hat{a}_n$ was sufficiently close to the solution $\hat{a}^*$, the standard linear solution of the resulting equations was used. Whenever this procedure gave a step, $\hat{a}_{n+1} - \hat{a}_n$, at an angle greater than

$$\cos^{-1}(0.1) = 89.4^\circ$$

to the gradient (after scaling the components of $\hat{a}$)

$$\nabla_{\hat{a}} P(\omega, \theta, \hat{a}_n),$$

a step along the above gradient was taken instead of the normal calculated step. After the solution, $\hat{a}^*$, was found by iterating the above procedure, the variance of the absorption coefficient was obtained by the standard linear least squares fitting method (see B. W. Lindgren, Statistical Theory, (MacMillan, N.Y., 1962), p. 386f) from the local linearization of $P(\omega, \theta, \hat{a})$ about $\hat{a}^*$ using the assumption that the variances of all our power measurements $P_p$ at a given far-infrared frequency were equal to $\sigma_p^2(\omega)$, a value measured near the peak of each phase matching function.
Let $M_y = (Y \tilde{Y})$

$$Y = \begin{pmatrix} p_1 - p(\omega, \theta_1, \tilde{a}^*) \\ p_2 - p(\omega, \theta_2, \tilde{a}^*) \\ \vdots \\ p_m - p(\omega, \theta_m, \tilde{a}^*) \end{pmatrix}$$

Then following Lindgren:

$$M^+ = [(\tilde{X}X)^{-1}X]M_y[X(\tilde{X}X)^{-1}]$$

or if $M_y = \sigma_p^2(\omega) I$, then

$$M^+ = \sigma_p^2(\omega) (\tilde{X}X)^{-1}$$

where $\tilde{X}$ and $\tilde{Y}$ are the transposes of $X$ and $Y$, respectively. The matrix $\tilde{X}X$ is adequately approximated by the matrix inverted at the last step of the iterative solution since $\nabla_{\tilde{a}^*}^2(\tilde{a}^*) = 2\tilde{X}X \cdot (\tilde{a}^* - \tilde{a}^*)$. 
FIGURE CAPTIONS

Fig. 1. Wavevector diagram for reflection of a plane wave incident from the vacuum side on the plane interface between vacuum and uniaxial crystal half-spaces: (a) crystal fills the right half-space; (b) crystal fills the left half space and (c) an equivalent diagram with the crystal on the right (Therefore, Fig. 1(b) is equivalent to Fig. 2(c) below).

Fig. 2. Wavevector diagrams showing (a) boundary transmission and reflection of ordinary and extraordinary waves and its decomposition into (b) and (c) which describe two simpler cases of linear transmission and reflection of waves at an interface.

Fig. 3. Angular distribution of far-infrared power output at $\omega = 100 \text{ cm}^{-1}$ with the axial phase mismatch at its optimum value $\Delta k_a = -5.1 \text{ cm}^{-1}$, a near optimum focal spot radius $w = 25 \mu\text{m}$, a zero walk-off angle $\zeta = 0$, and a crystal length $\ell = 1 \text{ cm}$.

Fig. 4. Far-infrared power output at $\omega = 100 \text{ cm}^{-1}$ as a function of $\Delta k_a \ell$, assuming $\alpha = 0$, $\zeta = 0$, $w = 25 \mu\text{m}$ and $\ell = 1 \text{ cm}$.

Fig. 5. Far-infrared power output at $\omega = 100 \text{ cm}^{-1}$ as a function of $\Delta k_a \ell$ assuming $\alpha = 0$, $\zeta = 0$, $w = 0.2 \text{ mm}$, and $\ell = 1 \text{ cm}$.

Fig. 6. Far-infrared power output at $\omega = 100 \text{ cm}^{-1}$ as a function of the focal spot radius $w$ for various walk-off angles $\zeta$, $\alpha = 0$, and $\ell = 1 \text{ cm}$. The calculation was done by always adjusting the axial phase mismatch $\Delta k_a$ to its optimum value for maximum power output.
Fig. 7. Angular distribution of far-infrared power output at $\omega = 10 \text{ cm}^{-1}$ for $\Delta k_a = -4.0 \text{ cm}^{-1}$, $w = 25 \mu\text{m}$, $\zeta = 0$, $\alpha = 0$, and $\ell = 1 \text{ cm}$. The azimuth $\phi$ is defined by $\phi = \tan^{-1}(y/x)$.

Fig. 8. Far-infrared power output at $\omega = 10 \text{ cm}^{-1}$ as a function of $\Delta k_a \ell$ assuming $\alpha = 0$, $\zeta = 0$, $w = 25 \mu\text{m}$, and $\ell = 1 \text{ cm}$.

Fig. 9. Far-infrared power output at $\omega = 10 \text{ cm}^{-1}$ as a function of the focal spot radius $w$ for various walk-off angles $\zeta$, $\alpha = 0$, and $\ell = 1 \text{ cm}$. The axial phase mismatch was always adjusted to its optimum value in the calculation.

Fig. 10. Angular distribution of the far-infrared power output at $\omega = 10 \text{ cm}^{-1}$ for various walk-off angles $\zeta$ assuming $w = 25 \mu\text{m}$, $\alpha = 0$, $\ell = 1 \text{ cm}$, and the optimum value of $\Delta k_a(\zeta)$. All curves were computed in the $\phi = 0$ plane.

Fig. 11. $P(\alpha)/P(\alpha = 0)$ versus $\alpha$ showing the reduction of output power due to far-infrared absorption. For each point on the curves for $\omega = 10 \text{ cm}^{-1}$ and $\omega = 100 \text{ cm}^{-1}$, $w = 25 \mu\text{m}$, an optimum value of $\Delta k_a$, and an optimum location of the focal plane were used in the calculation. A corresponding curve calculated from the plane wave model is also shown for comparison.

Fig. 12. Optimum values of $\Delta k_a \ell$ versus the absorption coefficient $\alpha$ for the case of Fig. 11 with $\omega = 100 \text{ cm}^{-1}$ where $\Delta k_a$ is the axial phase mismatch.

Fig. 13. Comparison of the results of the Gaussian distribution model, the plane wave model, and our present calculation for $\omega = 100 \text{ cm}^{-1}$, $\alpha = 0$, $\zeta = 0$, and $\ell = 1 \text{ cm}$.
Fig. 14. Comparison of the results of the Gaussian distribution model and our present calculation for $\omega = 10 \text{ cm}^{-1}$, $\alpha = 0$, $\ell = 0$, and $\ell = 1 \text{ cm}$.

Fig. 15. Phase-matched wavevector diagrams for (a) second harmonic generation and (b) difference frequency generation.

Fig. 16. Second harmonic power output as a function of $\Delta k \ell$ when $\Delta k R \ell = 100$. [After Boyd and Kleinman, Jour. Appl. Phys. 39, 3597 (1968)].

Fig. 17. Second harmonic power output as a function of $\Delta k \ell$ when $\Delta k R \ell = 5.68$. [After Boyd and Kleinman op. cit.]

Fig. 18. The far-infrared spectrum computed from Eq. (6) for a 2 psec. (full width at half-maximum) Nd laser pulse normally incident on a 1-mm LiNbO$_3$ slab. The crystal is oriented with the c-axis parallel to the plane surfaces of the slab and the laser pulse is polarized along the c-axis ($\chi_{33} = 1.57 \times 10^{-6} \text{ esu}$). The other parameters used in the calculation are $w_0 = 0.017 \text{ cm.}$ (corresponding to a 4-mrad. divergence of the laser beam), $L = 135 \text{ cm.}$, $n_i^{(\omega)} = 5.05$ and $n_i^{(\omega_0)} = 2.2$. The solid and the dashed curves are computed with and without boundary conditions respectively.

Fig. 19. The far-infrared spectrum computed from Eq. (6) with the same laser parameters as in Fig. 1. Here, the 1-mm slab is oriented with the c-axis tilted at $16.8^\circ$ away from the normal of the slab, and the a-axis is in the plane defined by the c-axis and the normal. The laser is polarized at $45^\circ$ to the
plane, so that only \( \chi_{24}^{NL} = 1.54 \times 10^{-6} \) esu is responsible for the difference-frequency signal with polarization perpendicular to the plane. With \( \hat{i} = \hat{j} \) (along the b-axis) and \( \hat{k} \) being the directions of polarizations of the ordinary and the extraordinary light propagating along \( \hat{z} \) respectively, we have \( n_{i}(\omega) = 6.6, n_{i}(\omega_o) = 2.2, \) and \( n_{k}(\omega_o) = 2.193. \)

**Fig. 20.** The electric field for the optical rectification case (as in Fig. 18). The optical input is a 2 psec square pulse and linear boundary reflections are ignored. Upper curve: field that travels back toward the laser \( (z < 0) \). Lower curve: field that propagates in the direction of the laser beam \( (z > \lambda) \).

**Fig. 21.** The spectrum of the sum of the lower and first reflection of the upper curves in Fig. 20 illustrating the interference of these two fields.

**Fig. 22.** The backward \( (z < 0; \) upper curve) and forward \( (z > \lambda; \) lower curve) wave electric fields for the o-ray, e-ray mixing case (as in Fig. 19). The optical input pulse is a 2 psec square pulse and linear boundary reflections are ignored.

**Fig. 23.** Wavevectors in non-collinear phase matching: (a) the phase matched case - \( k_1 = n_i \omega_1 / c; \omega_3 = \omega_1 - \omega_2 \equiv \omega \) for difference frequency generation. (b) The unmatched case.
Fig. 24. Dual-frequency dye laser system. The Ruby laser beam was circularly polarized with a quarter-wave plate. The two output frequencies could be independently tuned from 8100 to 8400 Å.

Fig. 25. Far-infrared bandwidth versus angular width of the phase matching function for 1.61 mm (solid line) and 1 cm (dashed line) thick LiNbO₃ samples used to generate o-ray far-infrared radiation in a type II collinear phase matching configuration at a 21 cm⁻¹ difference frequency.

Fig. 26. Experimental measurement of the phase matching function for our 1.61 mm thick LiNbO₃ sample at 21 cm⁻¹. The solid curve is the theoretical phase matching function for a 3 cm⁻¹ far-infrared bandwidth.

Fig. 27. O-ray absorption coefficient of LiNbO₃. The solid curve is a composite of Bosomworth’s (below 70 cm⁻¹) and of Barker and Loudon’s (above 70 cm⁻¹) o-ray absorption coefficients.

Fig. 28. Far-infrared peak power generated in LiNbO₃ versus difference frequency for three phase matching methods. The solid curves are the plane wave theory including the calculated dispersion of χ₂₄ (collinear) or χ₃₃ (non-collinear).

Fig. 29. Calculated far-infrared power vs. frequency for FCPM in LiNbO₃ with monochromatic lasers neglecting the dispersion of χ₂₄. The lower curve includes the exact boundary conditions discussed in chapter II; the upper curve is the power in the absence of all reflections. (a) Coaxial pump laser beams; (b) Laser beam axes .5 mm apart.
Fig. 30. Dispersion of $|x_{24}^{(2)}|^2$ in LiNbO$_3$. The circles are our experimental measurements. The solid curve is a multiple simple-harmonic-oscillator calculation based on the Raman cross sections of Kaminow and Johnston and the TO phonon oscillator strengths, frequencies, and linewidths of Barker and Loudon.

Fig. 31. Dye laser system used for non-collinear phase matching experiments in LiNbO$_3$.

Fig. 32. Schematic of the LiNbO$_3$ cube used for our non-collinear phase matching experiments.

Fig. 33. Dispersion of the electronic, $r_e$, and ionic, $r_\text{i}$, contributions to the linear electro-optic coefficient $r_{42}$ of CdS from the Raman scattering data of Ralston et al.
Fig. 1.
Fig. 2.
Fig. 3.
Fig. 4.
Fig. 5.
Fig. 6.
Fig. 7.
Fig. 8.
Fig. 9.
Fig. 10.
Fig. 11.
Fig. 12.
Fig. 13. Graph showing the relationship between power (arb. units) and focal spot radius ($10^{-2}$ cm). The graph compares the present calculation with a Gaussian distribution model and a P.W. (presumably a specific model or reference). The x-axis represents the focal spot radius in units of $10^{-2}$ cm, ranging from 0 to 2, while the y-axis represents power in arbitrary units, ranging from 0.2 to 1.4.
Fig. 14.

- Gaussian distribution model
- Present calculation
Fig. 15.
Fig. 16.
Fig. 17.
Fig. 18.
Fig. 19.
Fig. 20.
Fig. 21.

Spectral density, arb units

Frequency, cm$^{-1}$
Fig. 22.
Fig. 23.
Fig. 24.
Fig. 25.
Fig. 26.
Fig. 27.
Fig. 28.
Fig. 29b.
Fig. 30.
Fig. 31.
Fig. 32.
Fig. 33.
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