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Coherent Phonon Generation by Optical Mixing in a One-Dimensional Superlattice

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ABSTRACT

With the help of the lattice momentum, phase-matched optical generation of coherent phonons with frequencies higher than 50 GHz appears feasible. We have considered two cases: 1. Direct conversion\(^1\) of millimeter or sub-millimeter photons in a piezoelectric superlattice.

2. Optical mixing of two laser beams in a non-piezoelectric superlattice.
The generation of coherent hypersonic waves by driving a piezoelectric crystal with microwaves has very low efficiency as the frequency\(^2\) approaches 50 GHz. Phonon experiments done at frequencies in excess of 50 GHz have generally used incoherent heat pulse techniques\(^3,4\) or, more recently, phonon fluorescence\(^5\) in superconductors.

In this Letter, we examine the feasibility of generating high frequency coherent phonons in a superlattice by optical means. Because of the periodicity of a superlattice, coherent phase-matched generation of phonons by umklapp processes can occur at frequencies which would be prohibited in a homogeneous material. Direct conversion of photons into phonons is possible if the superlattice is piezoelectric, as suggested by Bloembergen and Sievers.\(^1\) This is the case in multi-layer epitaxially grown crystals\(^6,7\) or in partially oriented sputtered films.\(^8\) Phonons can also be generated by optical mixing in a superlattice via the electrostrictive effect.\(^9\) Single crystals are not required for this process, so that superlattices constructed by sputtering\(^8\) or vacuum deposition\(^10\) can be used.

Consider first the case of direct conversion of photons into phonons in a superlattice with alternating crystal layers of thickness \(d_1\) and \(d_2\) respectively. High frequency transverse phonon waves propagating along \(\hat{x}\) perpendicular to the layers can be generated at discrete frequencies by a far-infrared field \(E\) through the piezoelectric effect which is governed by the equation,

\[
\rho \frac{\partial^2 u}{\partial t^2} - \frac{\gamma}{2} \frac{\partial u}{\partial t} - T \frac{\partial^2 u}{\partial x^2} = \frac{3}{3x} (e \cdot E)
\]  

(1)
where \( i = 1 \) or \( 2 \) indicates the two different crystal layers, and \( \rho_i \), \( \gamma_i \), \( T_i \) and \( e_i \) are the density, the acoustic damping, the elastic stiffness and the piezoelectric stress constants respectively. We shall assume that the overall thickness of the superlattice is small enough that the far-infrared power is not depleted appreciably in the conversion. We also assume that the photon-phonon coupling is sufficiently weak compared with the phonon linewidth that the polariton effect \(^{11}\) can be neglected.

The undepleted far-infrared field in a periodic medium should have the Block form \( E = \&(x) \exp(ikx-i\omega t) \) with \( \& (x) = \& (x+nd) \) where \( d = d_1 + d_2 \) and \( n \) is an integer. This function \( \& (x) \) and the reduced wave vector \( k \) can be obtained from the homogeneous wave equation with the boundary conditions for the given layer structure.\(^ {12}\) If the wavelength of the far-infrared field is much larger than \( d \), then \( \& (x) \approx \) constant. Similarly, without the driving field the acoustic wave should also have the Block form \( U = U(x) \exp(iqx-i\omega t) \) where \( U(x) = U(x+d) \). With the driving field in Eq. (1), the acoustic wave can be written as \( U = A(x) U(x) \exp(iqx-i\omega t) \) where the amplitude function \( A(x) \) is a monotonically increasing function of \( x \). If \( A(x) \) is slowly varying so that \( q \left| \frac{\partial A}{\partial x} \right| \gg \left| \frac{\partial^2 A}{\partial x^2} \right| \) then Eq. (1) can be written as

\[
\frac{\partial A}{\partial x} = -\frac{3eE}{2T[iqU(x)+\partial U/\partial x]} \equiv f(x). \tag{2}
\]

The solution of the above equation is
\[ A(nd) = M(F_1 + F_2) \]  

\[ M = \frac{1 - \exp(i(k-q)nd)}{1 - \exp(i(k-q)d)} \]

\[
F_1 = \begin{cases} 
\int_{d_1}^{d_1+\varepsilon} f(x)dx + \int_{d_1+\varepsilon}^{d_1+d_2-\varepsilon} f(x)dx \\
\varepsilon + \int_{d_1-d_2+\varepsilon}^{d_1+\varepsilon} f(x)dx
\end{cases}
\]

\[
F_2 = \begin{cases} 
\int_{d_1}^{d_1+\varepsilon} f(x)dx + \int_{d_1+\varepsilon}^{d_1+d_2-\varepsilon} f(x)dx \\
\varepsilon + \int_{d_1-d_2+\varepsilon}^{d_1+\varepsilon} f(x)dx
\end{cases}
\]

where \( F_1 \) is the contribution from the bulk and \( F_2 \) is the contribution from the discontinuous boundaries between layers.

From Eq. (3), it is seen that if \( k = q \), then \( M = n \) is a maximum.

This phase-matching condition can be satisfied at many different phonon frequencies in a superlattice, whenever in the reduced zone scheme the photon dispersion curve crosses the phonon dispersion curve. Physically, the phase-matched generation of phonons at various frequencies with approximately the same reduced wave vector corresponds to different orders of umklapp processes. With a small phase mismatch \( \Delta k = k-q \), we have \( M = \frac{\sin(\Delta kd)}{\sin(\Delta nd)} \).

As an example, we consider a superlattice of 100 epitaxial GaAs:GaP layers normal to the [111] axis. The incoming em field is propagating along [111] and is polarized along [011]. We choose \( d_1 = 280 \) Å and \( d_2 = 354 \) Å so that \( d_1/v_1 = d_2/v_2 \) where \( v_1 \) and \( v_2 \) are the sound velocities in GaAs and GaP respectively. In this case, the sound dispersion and \( U(x) \) are easily calculated. We have calculated the conversion efficiency as a function of/discrete phonon frequencies, as shown in Fig. 1. Here, unless \( d > 50 \mu m \), \( F_1 \) in Eq. (3) can be neglected in comparison with \( F_2 \).
From Fig. 1, it is seen that the odd-order umklapp processes have larger conversion efficiencies \(10^{-4}\). This is because the phonon waves generated at the successive boundaries between layers are in phase. In the even-order umklapp processes, they are out of phase. We have assumed in the calculation that the piezoelectric constant is independent of frequency and the discontinuities between layers are infinitely sharp. Therefore, the conversion efficiencies for the odd-order and the even-order umklapp processes separately have no appreciable dependence on frequency. If the discontinuities between layers spread over a distance comparable to the phonon wavelength, then phase cancellation occurring within the boundaries will decrease the conversion efficiency.

We have also calculated the first-order effect on the conversion efficiency due to ± 2% random errors in the periodicity of the superlattice. The results are also given in Fig. 1. The random variation tends to decrease the phonon power at points of high conversion efficiency in a perfect superlattice, and increases the phonon power at points where the efficiency is low due to cancellation. The high-order umklapp processes which involve shorter phonon wavelengths are more sensitive to such imperfection as one would expect.

Consider next the generation of high-frequency longitudinal acoustic phonons in a superlattice by beating two laser beams at frequencies \(w_1\) and \(w_2\) via the electrostrictive effect. In this case, no crystalline structure of the films is necessary. We assume that the two beams propagating along \(\hat{x}\) have the same polarization. Then, the phonon generation is again described by Eq. (1), but with the driving
term replaced by \[ \frac{2}{2\pi} (\epsilon_0^2 PE_1 E_2^*/4\pi) \] where \( \epsilon_0 \) is the optical dielectric constant and \( P \), the stress-optical constant. A solution similar to Eq. (3) can also be obtained by assuming negligible depletion of laser power.

As an example, we consider a superlattice composed of 100 vacuum-deposited KCl: CdS layers with \( d_1 = 420\AA \) and \( d_2 = 450\AA \) so that again \( d_1/v_1 = d_2/v_2 \). The calculated phonon power is shown in Fig. 2 as a function of the discrete phonon frequencies corresponding to various umklapp processes. For odd-order umklapp processes, \( F_2 \) in Eq. (3) is the dominant effect as in the piezoelectric case. We notice that even for higher-order umklapp processes, the phonon power generated by two 10 MW/cm² laser beams can be as high as 5 mW provided that the discontinuities between layers spread over a distance much smaller than the sound wavelength. For even-order umklapp processes, the dominant contribution to the sound power is from \( F_1 \) in Eq. (3) since the two terms in \( F_2 \) are out of phase. The effect of a ± 2% random variation in the period of the superlattice is also shown in Fig. 2. Again, the high-order umklapp processes are more sensitive to such imperfection.

From our calculations, it appears that a superlattice can be used as a practical device to generate coherent hypersonic waves at very high frequencies. In both cases we have discussed, the acoustic power generated under phase-matching conditions is proportional to the square of the total number of unit cells in the superlattice.

The generation of hypersonic waves at frequencies higher than 50 GHz with a power larger than 1 mW should be possible in a superlattice.
about 10 \mu m thick.

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9. High order umklapp processes have larger phonon attenuation constants and so need larger laser threshold power for stimulated Brillouin scattering than zero order umklapp process. Unless specially designed cavities are used to discriminate against the zero order umklapp process, stimulated Brillouin scattering in a superlattice would thus produce only the same low frequency phonons as in a uniform medium.
   In this paper, the phonon attenuation was neglected in the calculation of the polariton modes.


FIGURE CAPTIONS

Fig. 1. The circular points are the calculated conversion efficiencies for various orders of umklapp process using a piezoelectric superlattice consisting of 100 epitaxial layers of GaAs:GaP with a total thickness of 6.34 μm. The triangular points give the conversion efficiency if the periodicity of the superlattice has a ± 2% random variation. Note the break in the vertical scale.

Fig. 2. The circular points are the calculated coherent phonon power (divided by the product of the powers of two mixing ruby laser beams\(^\text{13}\)) for various orders of umklapp process in a superlattice of 100 alternating vacuum evaporated layers of KCl-CdS with a total thickness of 8.7 μm. The triangular points give the phonon power if the periodicity of the superlattice has a ± 2% random variation. Note the break in the vertical scale.
Fig. 1
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