Nonlinear Light-Matter Interactions in Metamaterials

by

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A dissertation submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Physics in the Graduate Division of the University of California, Berkeley

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Abstract
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Metamaterials possess extraordinary linear optical properties never observed in natural materials such as a negative refractive index, enabling exciting applications such as super resolution imaging and cloaking. In this thesis, we explore the equally extraordinary nonlinear properties of metamaterials. Nonlinear optics, the study of light-matter interactions where the optical fields are strong enough to change material properties, has fundamental importance to physics, chemistry, and material science as a non-destructive probe of material properties and has important technological applications such as entangled photon generation and frequency conversion. Due to their ability to manipulate both linear and nonlinear light matter interactions through sub-wavelength structuring, metamaterials are a promising direction for both fundamental and applied nonlinear optics research.

We perform the first experiments on nonlinear propagation in bulk zero and negative index optical metamaterials and demonstrate that a zero index material can phase match four wave mixing processes in ways not possible in finite index materials. In addition, we demonstrate the ability of nonlinear scattering theory to describe the geometry dependence of second and third harmonic generation in plasmonic nanostructures. As an application of nonlinear metamaterials, we propose a phase matching technique called “resonant phase matching” to increase the gain and bandwidth of Josephson junction traveling wave parametric amplifiers. With collaborators, we demonstrate a best in class amplifier for superconducting qubit readout – over 20 dB gain with near quantum limited noise performance with a bandwidth and dynamic range an order of magnitude larger than alternative devices. In conclusion, we have demonstrated several ways in which nonlinear metamaterials surpass their natural counterparts. We look forward to the future of the field where nonlinear and quantum metamaterials will enable further new physics and new applications.
To my family, friends, and mentors without whom this would not have been possible.
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Chapter 1

Introduction

Photonics touches every aspect of modern life. Optical fiber, laser, and detector technology is crucial for the global telecommunication network. Advances in lighting efficiency are important to reduce energy consumption and reduce our carbon footprint. Nonlinear optical processes such as three and four wave mixing and pump probe spectroscopy are important tools for non destructive material characterization. Terahertz and mid-infrared spectroscopy operating in the fingerprint region, the frequency range in which many chemical bonds vibrate, is important for chemical analysis and medical diagnostics. Linear optical systems and those coupled to quantum systems such as atoms or ions are a promising path towards developing quantum computers. Advances in materials lie at the heart of many of these impressive achievements. For example, transoceanic optical fiber communication would have not have been feasible without the development of ultra-transparent glass by Charles Kao and colleagues, for which he received half of the 2009 Physics Nobel prize.

The focus of this thesis is exploring the nonlinear properties of a class of man-made materials called metamaterials. In the last several decades, advances in engineered materials such as metamaterials and photonic crystals have revolutionized photonics.

1.1 Metamaterials

A metamaterial is a manmade composite material which is structured on a sub-wavelength scale\cite{125,72}. The properties of a metamaterial are derived both from the electromagnetic properties of the constituents and their structure. Much interest in metamaterials stems from their ability to manipulate both the electric properties, described by the permittivity, and the magnetic properties, described by the permeability. As shown by Victor Veselago in 1968\cite{135}, a simultaneously negative permittivity and permeability imply a negative refractive index; which has the surprising consequence that the phase velocity and the Poynting vector (the energy flow), point in opposite directions. Importantly, this has applications for super-resolution imaging as realized by John Pendry in his 2000 work “Negative Refraction Makes a Perfect Lens”\cite{97}. An ideal perfect lens would have the ability to image both the propagating waves used in a typical lens and the evanescent waves, enabling such a lens to break the diffraction limit. In the last 15 years, negative index met
materials have been demonstrated at both microwave\cite{122} and optical frequencies\cite{119, 133} as well as proof of concept demonstrations of perfect lenses\cite{49, 41}.

The ability to control the permittivity and permeability allows one to create designer materials, which exhibit previously unachievable effects such as negative indices, super-resolution imaging, and cloaking\cite{116, 93}. However, the metamaterials discussed so far have a limitation: they are linear time invariant systems (LTIs) and thus subject to a number of constraints which preclude many desired applications such as amplification and frequency conversion. To break free from these constraints, nonlinear metamaterials are required as we will detail in the next section.

1.2 Nonlinear Metamaterials

Nonlinear optics is broadly defined as light-matter interactions where the electromagnetic field is strong enough to change material properties. Modern nonlinear optics dates back to the development of the laser in the early 1960s\cite{77}. The availability of an intense coherent light source enabled Franken et al. to observe frequency doubling of a ruby laser by a quartz crystal in 1961\cite{45}. From that point, the field grew rapidly due to the development of new materials, new sources, and new techniques\cite{19, 121, 18}.

From 2003 to 2004, shortly after the development of metamaterials, several groups published theoretical studies of metamaterials incorporating a nonlinear element\cite{65, 148, 1}. It was suggested that these artificial nonlinear materials could exhibit all of the nonlinear processes found in natural materials, such as frequency mixing, parametric amplification, and phase conjugation while benefiting from the field enhancement and full control over the permittivity and permeability obtainable with metamaterials.

One of the strengths of nonlinear metamaterials is their ability to manipulate the phase matching conditions for nonlinear processes. Phase matching refers to a matching of the phase velocities of both the pump and the products of a nonlinear mixing process and is required for efficient conversion\cite{18}. An example of phase matching from daily life is operating a swing, a type of parametric amplifier. If the swing is driven, by moving one’s center of mass, at the wrong phase relative to the motion, then the amplitude of the oscillations will decrease. If the phase of the drive is correctly chosen, then the amplitude of the oscillations will increase. Through manipulating the sign of the refractive index, metamaterials can generate novel phase matching conditions. For example, for second harmonic generation, the phase matching conditions and the Manley-Rowe relations predict that when the refractive index at either the pump or the second harmonic frequency is negative then one can obtain a “nonlinear mirror”\cite{118, 98} in which the second harmonic is emitted back towards the pump. This effect was demonstrated at microwave frequencies\cite{111} and very recently at optical frequencies in plasmonic waveguides\cite{64}. The drive to study nonlinear metamaterials is motivated by the fact that nonlinear metamaterials can exhibit phenomena not possible in a linear passive metamaterial such as parametric amplification\cite{100}, switching\cite{149}, and a multitude of other nonlinear processes\cite{66}. 
1.3 Thesis overview

A key advantage of nonlinear metamaterials over natural materials is the ease with which their properties can be tailored. To take advantage of this design freedom, one needs the capability to predict the nonlinear properties of metamaterials. In Chapter 2, we discuss calculating the nonlinear properties of nanostructures using nonlinear scattering theory. We describe nonlinear scattering theory as well as our implementation in the commercial finite element method solver COMSOL.

In Chapter 3, we describe our experimental tests of nonlinear scattering theory on second harmonic generation from arrays of plasmonic nanostructures whose geometry changes from inversion symmetric bars to highly asymmetric U-shaped structures. These experiments indicate that nonlinear scattering theory provides an accurate description of the geometry dependence of metamaterial nonlinearities.

In Chapter 4, we detail our experimental setup for broadband measurements of the refractive index of metamaterials. Such a capability is important for characterizing zero and negative index metamaterials. We develop the phase measurement setup based on spectrally and spatially resolved interferometry using entirely reflective optics. This system can measure a refractive index variation of $\lambda/300$ over the visible and near infrared (400 nm - 1700 nm). We use this system to characterize of phase response of a $\pi$ shaped nanostructure which has an EIT-like transmission spectrum.

In Chapter 5, we describe our experimental measurement of the first nonlinear propagation effects in bulk zero and negative index optical metamaterials. First, we design and fabricate fishnet metamaterials, a type of metamaterial previously used to observe the first bulk negative index at optical frequency. We then characterize the refractive index using our setup based on spectrally and spatially resolved interferometry as detailed in the previous chapter. Finally, we perform nonlinear experiments using single-shot four wave mixing. We find that in the zero index region, forward and backward four wave mixing are both phase matched, while in the positive or negative index regions, only forward propagating four wave mixing is phase matched. The observed phase matching of both forward and backward nearly degenerate four wave mixing is not possible in finite index media.

At optical frequencies, nonlinear metamaterials have a number of interesting properties; however, losses pose a challenge for many applications. At microwave frequencies, and particularly in superconductors, minimal losses and large nonlinearities create opportunities for nonlinear metamaterials devices to surpass the performance of semiconductor technology. In Chapter 6, we describe the Josephson traveling wave parametric amplifier (JTWPA), a traveling wave parametric amplifier using Josephson junctions as the nonlinear element. Crucially, we propose a scheme called “resonant phase matching” to phase match the nonlinear propagation in this amplifier and significantly increase the gain and bandwidth.

In Chapter 7, we solve for the strong field dynamics of the Josephson traveling wave parametric amplifier (JTWPA). We find a traveling wave solution which propagates without distortion and identify the higher order signal and idler sidebands as a potential cause of the non-unity quantum efficiency. We propose a simple method to increase the quantum efficiency by decreasing the junction plasma frequency.
In the final chapter, Chapter 8, we summarize and give concluding remarks on the results presented in this thesis. We also give perspectives on potential future directions.

1.4 Summary of key results

The key results of this thesis are twofold: we perform the first demonstration of phase matched nonlinear propagation in a nonlinear zero index optical metamaterial[127]. Second, we propose[96] and demonstrate[76] the technique of resonant phase matching to increase the gain and bandwidth of a traveling wave parametric amplifier. This low noise amplifier has broad applicability to single microwave photon level microwave measurements in circuit quantum electrodynamics, opto-mechanics, and astrophysics.
Chapter 2

Predicting the nonlinear properties of metamaterials with nonlinear scattering theory

The principle advantage of nonlinear metamaterials over natural nonlinear materials is that the nonlinear properties of metamaterials can be controlled through design of the sub-wavelength components. To realize the benefits of this design freedom, one requires a rapid and robust simulation technique to predict both the qualitative and quantitative properties of nonlinear metamaterials. Treating the nonlinear interaction between light and nanostructures requires solving Maxwell’s equations for a linear and a nonlinear polarization over arbitrary and often complicated geometries. A number of numerical techniques are capable of solving this problem such as the boundary element method (BEM)\cite{78, 48} or nonlinear finite-difference time-domain (FDTD)\cite{28, 29} and the finite element method (FEM)\cite{6}; however, we find that nonlinear scattering theory has several practical advantages. We briefly summarize the advantages and disadvantages below:

- Nonlinear scattering theory is both conceptually and computationally simple to implement, requiring only the evaluation of an overlap integral between the linear and nonlinear polarizations.

- The overlap integral can be evaluated with commercial electromagnetic solvers. This allows us to take advantage of their ability to simulate electrodynamics over complex geometries, without the substantial resource investment required to develop custom EM simulation software. Although we use a finite element method (FEM) solver; other methods which provide surface fields such as BEM or multiple multipole method (MMM) are suitable. For problems involving a bulk rather than a surface nonlinear polarization; simulation methods such as FDTD which do not provide surface fields are acceptable.

- A somewhat subtle point is that a surface nonlinear polarization can result in a discontinuity of both the tangential and normal components of the electric and magnetic fields at an interface\cite{121}. Implementations of the finite element method, such as
COMSOL Multiphysics, assume continuity of either the tangential electric or magnetic fields. This renders the software incapable of directly simulating the electromagnetic fields resulting from such a surface nonlinear polarization for some elements of the surface nonlinear susceptibility tensor. The inability to simultaneously solve for discontinuous tangential electric and magnetic fields poses no obstacle to simulating the far field electromagnetic response using nonlinear scattering theory.

- The disadvantage of nonlinear scattering theory is that it only provides the electromagnetic field at a single position per calculation. Typically, this position is chosen to be in the far field, where a detector would be placed in an experiment. For example, to investigate the nonlinear response of an array of nanostructures, then the position is chosen to be in the far field at the desired angle relative to the array. Due to the requirement of a separate evaluation of the electromagnetic fields for each position, nonlinear scattering theory is capable of, but inefficient for mapping the near field nonlinear response of nanostructures. We find that for predicting the far field response of nonlinear metamaterials, the reduction of computational complexity is well worth sacrificing knowledge of the nonlinear near fields.

Here we describe nonlinear scattering theory and the details of our implementation. In the next chapter, we compare the predictions of nonlinear scattering theory to our experimental results.

2.1 Nonlinear scattering theory

The goal here is to calculate the nonlinear emission from a nanostructure when pumped by a strong electromagnetic field using nonlinear scattering theory and compare the results to experiments. In nonlinear scattering theory, the problem of calculating the field from a distribution of nonlinear dipoles is converted into finding the overlap integral between the nonlinear polarization and the electric field radiated by a dipole placed at the detector. We first describe the nonlinear polarization, then derive the nonlinear scattering theory, and demonstrate the use in a simple model systems and for nanostructures with arbitrary geometries.

The polarization term in Maxwell’s equations can be divided into linear terms which are independent of intensity and nonlinear terms which depend on the intensity of the incident field:

\[ P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkm}^{(3)} E_j E_k E_m + \ldots \]  
\[ = \chi_{ij}^{(1)} E_j + P_i^{NL} \]  

The nonlinear polarization leads to processes which are intensity dependent where the photon energy can change such as wave mixing. In this treatment, we take the undepleted pump approximation, or in other words assume the nonlinear polarization is much smaller than the linear polarization at all times. This allows us to decouple the two problems: first calculating the nonlinear polarization from the linear response of the system, then in a separate calculation determine the emission from the nonlinear polarization.
Nonlinear scattering theory is a general theory for nonlinear emission from nanostructures \[110, 33\]. The assumptions are (1) Lorentz reciprocity and (2) the nonlinearity can be described by a local nonlinear susceptibility tensor.

Consider two current sources \( j_1(r, \omega) \) and \( j_2(r, \omega) \) which emit optical frequency electromagnetic fields \( E_1(r, \omega) \) and \( E_2(r, \omega) \) in a reciprocal medium. We consider materials with losses, \( \epsilon \) and \( \mu \) may be complex, but without magneto-optical properties so the \( \epsilon \) and \( \mu \) tensors are symmetric. Lorentz reciprocity yields the following relation between the localized sources and the fields they emit\[73\]:

\[
\int j_2(r', \omega) \cdot E_1(r', \omega) dV' = \int j_1(r, \omega) \cdot E_2(r, \omega) dV
\]  

We select one current source to be an ensemble of nonlinear dipoles on the surface or in the volume of a nanostructure which are excited by the pump field:

\[
j_1(r, \omega) = \frac{\partial P(r, \omega)}{\partial t} = i\omega P(r, \omega)
\]  

which emit an unknown electric field \( E_1 \). The second current source, a current dipole with a polarization axis given by \( \hat{j} \) and a length \( \Delta l \) is placed at the detector position.

\[
j_2(r, \omega) = J_0 \delta(r - r_2) e^{i\omega t} \hat{j}
\]

By substituting these currents into the reciprocity relation and evaluating the integral on the left of Eq. 2.3, we obtain a relation for the unknown field from the nonlinear polarization \( E_1 \):

\[
\int E_1 \cdot J_0 \delta(r - r_2) e^{i\omega t} \hat{j} dV' = \int i\omega P \cdot E_2 dV
\]

Eq. 2.7 can be directly used to calculate the nonlinear emission; however, when using numerical solvers of Maxwell’s equations, it is often inconvenient to calculate the field from a dipole placed at a detector far from the nanostructure because of the large range of lengths scales involved. Instead, a plane wave source with amplitude \( E_{20} \) is used. The relation between the dipole moment \( J_0 \Delta l \) and the plane wave amplitude can be obtained by solving Maxwell’s equations for the current source. This is conveniently done by solving for the Green’s function for the Helmholtz equation:

\[
(\nabla^2 + k^2)G(r, r') = -\delta(r - r')
\]

In three dimensions, the Green’s function is:

\[
G(r, r') = \frac{1}{4\pi |r - r'|} e^{-ik|r - r'|}
\]

The vector potential for the current dipole \( j_2 \) is then:

\[
A_2(r) = \mu \int j_2(r') G_0(r, r') dV' = \mu \Delta l J_0 \frac{e^{i\omega t} \hat{j}}{4\pi |r - r_2|} \frac{1}{e^{-ik|r - r_2|}}
\]
Figure 2.1: Illustration of nonlinear scattering theory. (a) the system consists of a nonlinear optical material and a detector. (b) a nonlinear polarization is excited by a pump field. In nonlinear scattering theory, the equality of the following integrals is used to calculate the nonlinear emission: (a) the integral of the field emitted by the nonlinear polarization and a current source placed at the detector \( \int j_2(r', \omega) \cdot E_1(r', \omega) dV' = \int j_1(r, \omega) \cdot E_2(r, \omega) dV \) and (b) the integral of the field emitted by the current source at the detector with nonlinear polarization \( \int j_1(r, \omega) \cdot E_2(r, \omega) dV \)

Thus dipole at the detector radiates an electric field in a direction perpendicular to the polarization axis of:

\[
E_{20}(r, \omega) = \frac{dA_2(r)}{dt} = i\omega A_2(r) = \frac{i\omega \mu A_2(r)}{4\pi r} e^{-ikr + i\omega t} \hat{j}
\]

where \( r = |r - r_2| \) is the distance between the nanostructure and the detector. Substituting in the magnitude of the field at the nanoparticle from the source at the detector, we obtain the nonlinear emission from the polarization \( P \) in terms of an overlap integral:

\[
E_1 \cdot \hat{j} = \frac{i\mu \omega^2 e^{-i\omega t}}{4\pi r} \int P \cdot \frac{E_2}{|E_{20}|} dV
\]

where \( E_2 \) is obtained from the interaction of the plane wave of amplitude \( E_{20} \) with the nanostructure.

This approach is computationally simple and accounts for the electrodynamic behavior of the structure including loss.
2.2 Example: Second harmonic generation in a one dimensional nonlinear crystal

We now demonstrate nonlinear scattering theory for a simple 1 dimensional system and compare to directly solving the nonlinear wave equation. In one dimension the Green’s function for the Helmholtz equation is:

\[ G_{x,x'} = -\frac{i}{2k} e^{-ik|x-x'|} \]  

(2.16)

The vector potential for the current dipole \( j_2 \) is then:

\[ A_2(x) = \mu \int j_2(x')G_0(x,x')dx' \]

\[ = -\mu \Delta l J_0 e^{i\omega t} \frac{i}{2k} e^{-ik|x-x_2|} \hat{j} \]  

(2.17)

Thus a dipole at the detector radiates an electric field in a direction perpendicular to the polarization axis of:

\[ E_{20}(r,\omega) = \frac{dA_2(r)}{dt} = i\omega A_2(r) \]

\[ = \frac{\mu\omega \Delta l J_0 e^{i\omega t}}{2k} e^{-ik|x-x_2|} \hat{j} \]  

(2.18)

The relation for the unknown field from the nonlinear polarization \( E_1 \) is then:

\[ E_1 \cdot \hat{j} = e^{-i\omega t} \int_{0}^{\Delta l} i\omega P \cdot E_2 dV \]

\[ = \frac{i\mu \omega^2 e^{-i\omega t}}{2k} \int P \cdot \frac{E_2}{|E_{20}|} dV \]  

(2.19)

We consider second harmonic generation in the undepleted pump approximation in medium with length \( L \), a refractive \( n(\omega) \), a wave vector \( k(\omega) = \frac{2\pi n(\omega)}{\lambda} \), and a nonlinear susceptibility \( \chi^{(2)} \). The material can have losses in which case \( n \) and \( k \) will be complex. The pump field \( E(\omega) = E_0 e^{-ikx+i\omega t} + c.c \) originates from \( x = 0 \) and propagates to the right. The pump field generates a nonlinear polarization in the medium of \( P^{NL}(2\omega) = \epsilon_0 \chi^{(2)} E_0^2 e^{i2k(\omega)x+2\omega t} + c.c \). The field from the detector at the second harmonic frequency, \( E'(2\omega) = E'_0 e^{-ik(2\omega)(x-L)+2\omega t} + c.c., \) originates from \( x = L \) and propagates to the left. We drop the complex conjugate terms in the calculation below. The amplitude of the generated second harmonic is then calculated from the overlap integral:

\[ E_{SH} = \frac{i\mu \epsilon_0 \cdot (2\omega)^2 e^{-2i\omega t}}{2k(2\omega)E_0} \int_{0}^{L} P^{NL}(2\omega) \cdot E'(2\omega) dx \]

\[ = \frac{2i\mu \epsilon_0 \omega^2 \chi^{(2)} E_0^2 e^{i(2k(\omega)L+2\omega t)}}{k(2\omega)} \int_{0}^{L} e^{i(2k(\omega)-ik(2\omega))x} dx \]  

(2.20)

\[ = \frac{2i\mu \epsilon_0 \omega^2 \chi^{(2)} E_0^2 e^{i(2k(\omega)L+2\omega t)}}{k(2\omega)} \frac{e^{i(2k(\omega)-ik(2\omega))L} - 1}{2i(k(\omega) - ik(2\omega))} \]  

(2.21)
defining $\Delta k = 2k(\omega) - k(2\omega)$, we can write this in a more familiar form:

$$E_{SH} = \frac{2i\mu\epsilon_0\omega^2 \chi^{(2)} E_0^2 e^{ik(2\omega) L + 2i\omega t} e^{i\Delta k L} - 1}{i\Delta k}$$  

(2.27)

$$= \frac{2i\mu\epsilon_0\omega^2 \chi^{(2)} E_0^2}{k(2\omega)} L e^{ik(2\omega) L + ik(2\omega) L/2 + 2i\omega t} \text{sinc} \left( \Delta k L/2 \right)$$  

(2.28)

Separating the wave vector and refractive index into real and imaginary parts

$k = k' + ik''$, $n = n' + i n''$, the second harmonic intensity is then

$$I_{SH} = 2\epsilon_0 n'(2\omega) |E_{SH}|^2$$  

(2.29)

$$= \frac{\omega^2 |\chi^{(2)}|^2}{2\epsilon_0 n'(2\omega) \epsilon_0 n'^2(\omega)} I_0^2(\omega) L^2 \times$$  

(2.30)

$$e^{-2k''(\omega) L - k''(2\omega) L} \text{sinc} \left( \Delta k L/2 \right)^2$$  

(2.31)

When the medium has no losses, $k''(2\omega) = k''(\omega) = 0$, then the formula reduces to the textbook result for second harmonic generation[5, 18]:

$$I_{SH} = \frac{\omega^2 |\chi^{(2)}|^2}{2\epsilon_0 n'(2\omega) \epsilon_0 n'^2(\omega)} I_0^2(\omega) L^2 \text{sinc}^2 \left( \Delta k L/2 \right)$$  

(2.32)

To compare with literature results for loss, we assume perfect phase matching by setting $\text{Re}[2k(\omega) - k(2\omega)] = 0$ in Eq. 2.26, then the second harmonic amplitude is:

$$E_{SH} = \frac{-2i\mu\epsilon_0\omega^2 \chi^{(2)} E_0^2 e^{2i\omega t} e^{-2k''(\omega) L} - e^{-k(2\omega) L}}{2k''(\omega) - k''(2\omega)}$$  

(2.33)

in agreement with the solution for second harmonic generation in a medium with loss[2].

Figure 2.2: (a) Second harmonic intensity as a function of material loss, calculated from nonlinear scattering theory (blue) and a direct solution of the nonlinear wave equation (black dashed). (b) Second harmonic intensity as a function of phase mismatch, calculated from nonlinear scattering theory (blue) and a direct solution of the nonlinear wave equation (black dashed). The nonlinear scattering theory calculations are in agreement with the direct solution of the nonlinear wave equation.

We see that nonlinear scattering theory accurately predicts the nonlinear emission taking into account both phase matching and material absorption in this simple 1d model,
in perfect agreement with the direct solutions of the nonlinear wave equation. A detailed comparison between the direct solution and the scattering theory solution for a nanosphere can be found in Ref. [33]. In the following section we apply nonlinear scattering theory to complex nanostructures in which a direct solution of Maxwell’s equations would be more challenging.

2.3 Numerical implementation for second harmonic generation in nanostructures

We consider second harmonic generation in nanostructures composed of centrosymmetric materials. In this case, the second harmonic originates from the interface between the two materials. For a process such a second harmonic generation which occurs only on the surface, the integral will reduce to a surface integral. The surface susceptibility is described in terms of the vectors normal ($n$) and parallel ($t$) to the metal-air interface. In a 3 dimensional simulation, most finite elements solvers will provide the normal and two tangential unit vectors and the fields in the chosen coordinate system, typically cartesian coordinates. From these quantities, the normal and tangential fields can be constructed by taking the appropriate dot products. The fields are evaluated outside of the metal. The nonlinear susceptibilities of gold from experimental data[142] are $\chi_{nnn} = 4.42 \cdot 10^{-22} m^2/V$, $\chi_{ntt} = 4.94 \cdot 10^{-21} m^2/V$, and $\chi_{un} = 1.77 \cdot 10^{-21} m^2/V$.

The linear electric field is described in the local coordinate system of the surface of the nanostructure (Fig. 2.3):

$$E(\omega) = E_n \hat{n} + E_{t_1} \hat{t}_1 + E_{t_2} \hat{t}_2$$

(2.34)

In our calculations these linear electric fields are calculating with the finite element solver
COMSOL multiphysics. These are used to calculate the surface nonlinear polarization:

\[ \mathbf{P}^{NL} = \chi_{nnn} E_n E_n \mathbf{n} + \chi_{ntt} E_{t1} E_{t1} \mathbf{n} + \chi_{ntt} E_{t2} E_{t2} \mathbf{n} \]

\[ + \chi_{ttn} E_{t1} E_{t1} \mathbf{t1} + \chi_{ttn} E_{t2} E_{t2} \mathbf{t2} \]

(2.35)

(2.36)

We then calculate the field on the nanostructure from a plane wave at the second harmonic frequency traveling from the detector:

\[ \mathbf{E}'(2\omega) = E'_n \mathbf{n} + E'_{t1} \mathbf{t1} + E'_{t2} \mathbf{t2} \]

(2.37)

We then calculate the second harmonic in the far field (Eq. 2.15) by performing the overlap integral between the field from the detector and the nonlinear polarization:

\[ E_{SH} \propto \int \mathbf{P} \cdot \mathbf{E}' d^2r \]

(2.38)

\[ \propto \int \chi_{nnn} E'_n E_n E_n \]

\[ + \chi_{ntt} E'_{t1} E_{t1} + \chi_{ntt} E'_{t2} E_{t2} \]

\[ + \chi_{ttn} E'_{t1} E_{t1} E_n + \chi_{ttn} E'_{t2} E_{t2} E_n d^2r \]

(2.39)

(2.40)

(2.41)

One must be careful to specify a side of the metal-dielectric interface when the extracting fields. Many finite element solvers will return the average of the fields on both sides of the interface if one side is not specified, which will lead to incorrect results. It does not matter which side is chosen, as long as it is consistent with the definition for the nonlinear susceptibility\cite{142}. In the simulation software we use, the side on which fields are evaluated can be specified with the up() and down() commands. One should not that the choice of which side to evaluate the nonlinear susceptibility will change the proportion of the total energy arising from the different elements of the nonlinear susceptibility tensor, but will not change the overall nonlinear response.

### 2.4 Conclusions

In conclusion, we have detailed the method used to calculate the nonlinear emission from plasmonic nanostructures. In the next chapter, we will compare the predictions of nonlinear scattering theory to experimental results for U-shaped plasmonic nanostructures of varying morphology.
Chapter 3

Experimental tests of nonlinear scattering theory

The discovery of optical second harmonic generation in 1961 started modern nonlinear optics[45, 5, 11, 46]. Soon after, R. C. Miller found empirically that the nonlinear susceptibility could be predicted from the linear susceptibilities. This important relation, known as Millers Rule[46, 88], allows a rapid determination of nonlinear susceptibilities from linear properties: revolutionizing the discovery of nonlinear optical materials. In recent years, metamaterials, which exhibit intriguing linear optical properties not found in natural materials like negative refractive indices[119], have novel nonlinear properties such as phase mismatch free nonlinear generation[127], new quasi-phase matching capabilities[111, 112], and large nonlinear susceptibilities[111, 112, 128]. However, understanding of nonlinear metamaterials is still in its infancy, with no general conclusion on the relationship between linear and nonlinear properties. The key question is whether Miller’s Rule applies to nonlinear metamaterials or equivalently, can one determine the nonlinear behavior of these artificial materials from their exotic linear behavior? Here we show that Miller’s Rule does not apply in general to nonlinear metamaterials. In contrast to natural materials, in plasmonic metamaterials, the maximum nonlinear susceptibility does not correlate with the maximum linear susceptibilities measured in far field due to the larger size of the metamolecules and the geometry dependent mode overlap. We show, instead, that it is possible to predict the relative nonlinear susceptibility of large classes of metamaterials using a more comprehensive nonlinear scattering theory, which allows efficient design of metamaterials with strong nonlinearity for important applications such as coherent Raman sensing, entangled photon generation, and frequency conversion.

The broad applicability of Millers rule, which holds for a wide variety of natural materials, stems from the fact that it is a direct result of the Lorentz oscillator description of material nonlinearity in which a charge is moving in a non-quadratic potential[46]. An estimate for Millers delta, The proportionality between the nonlinear susceptibility and the linear susceptibilities, can be obtained by assuming that the linear and nonlinear restoring forces will be of similar magnitudes when the displacement of the charge is on the order of the inter-atomic distance, leading to an approximate value of Miller’s delta of $\Delta M \approx a^2/e$, where $a$ is the inter-atomic distance and $e$ is the electron charge. Experimentally, Millers
delta was found to be nearly constant with a value of $0.45 \pm 0.07 m^2/C$ for many semiconductor crystals even though the linear and nonlinear susceptibilities spanned over four orders of magnitude[19, 115]. The model was shown to apply to a wide variety of materials from semiconductor crystals to atomic vapors[25, 87] and noble metals[81]. In a quantum mechanical treatment, the derivative of the potential is replaced with an average over the ground state[104]. The development of metamaterials in the last decade allows the exploration of the nonlinear properties of such materials at optical[53, 86] and microwave[101] frequencies. Researchers have found that in some cases, such as the third harmonic emission from bow tie and double-bar nanostructures[53, 86] Millers rule or its equivalent harmonic oscillator model[9] quite accurately predicts the nonlinear susceptibilities. However, the general validity of Miller’s rule in optical metamaterials for arbitrary nonlinear processes, and specifically for second order susceptibilities is not known. Here we show experimentally that Miller’s rule fails to describe the second order susceptibility of metamaterials and predicts an incorrect optimum geometry for generating the most second harmonic. We however demonstrate the optimal geometry can be correctly predicted with a more general nonlinear scattering theory. This general principle describes not only the second but also higher order nonlinear optical responses of plasmonic nanostructures over a broad wavelength range. The predictive capability of nonlinear scattering theory enables rapid design of optimal nonlinear nanostructures for sensing and integrated photonics.

3.1 Sample design

Predicting the optimal metamaterial for nonlinear generation is a critical test of Miller’s rule and microscopic theories of metamaterial nonlinearity. We study the second harmonic generation from metamaterial arrays in which the geometry varies gradually from a symmetric bar to an asymmetric shape. A schematic of the sample design is shown in Fig. 3.1, where the morphology changes along one axis and the length changes along the other. A dramatic increase in the second order susceptibility is expected due to the extreme sensitivity of second harmonic generation to symmetry.

![Figure 3.1: Schematic of the nanostructure array design. (a) A 2D array of different nanostructures is generated by discretizing the parameters specifying the geometry: the length and asymmetry ratio. (b) The length is varied along the vertical axis and the asymmetry ratio is varied along the horizontal axis of the array.](image-url)
3.2 Sample preparation

The samples were prepared using a standard electron beam lithography and metal lift-off process. The array of metamaterials is fabricated on top of a quartz substrate where a 2 nm thick layer of Indium-Tin-Oxide (ITO) is deposited by sputtering (Auto 306, Edwards) as an EBL conductive layer. After defining the nanometer scale metamaterial patterns in the bi-layer photoresist with 250 nm thick methyl methacrylate (MMA-EL8) and 40 nm thick polymethyl methacrylate (PMMA-A2) with high-resolution EBL (CABL-9000C, Crestec), an electron beam evaporation system (Solution, CHA) is then used to deposit chromium and gold thin film followed by gentle soaking in acetone to lift-off the photoresist layer. The width of the nanostructures was approximately 40 nm, the thickness of the gold and chromium was 35 nm and 2 nm, respectively. The chromium layer is used to enhance adhesion between the quartz surface and the gold layer.

3.3 Experimental setup

A confocal microscope was used to measure the linear transmission and the nonlinear emission from each geometry. Stage scanning confocal microscopy allows us to investigate the geometry dependent nonlinear properties of metamaterials, while avoiding the challenges associated with changing the laser wavelength such as differences in pulse shape, focal volume size, or transmission[24]. The light source is an optical parametric oscillator (Spectra Physics Opal) pumped by a 100 fs Ti:Sapphire oscillator (Spectra Physics Tsunami). The pulse width is approximately 200 fs. The light source is focused to a spot size of 1.25 micrometers and scanned across the sample. A PMT is used for detection and color filters are used to isolate the second and third harmonic. The filters used were HG450/65 for the third harmonic and HG675/55 for the second harmonic. A KG3 filter was used to absorb the infrared. The power level was measured to be 100 microwatts of incident power at the back aperture of the objective. Spectral measurements show that at the power levels used no observable continuum emission is present. We work in transmission mode with a Zeiss LD-Plan-NEOFLUAR 63x 0.75NA objective for excitation and 40x 0.65 NA objective for collection. The collection half angle for the objective is 40.5 degrees. The detector is a Hamamatsu H7421-40 photon counting PMT. For linear transmission measurements we use a fianium supercontinuum source. Experiments performed on sparse arrays (500 nm period) and for isolated nanostructures (2000 nm period), yield similar results, suggesting that particle-particle and long range interactions are not important in this system. In addition, the 1.25 micron focal spot only illuminates order of 10 particles, limiting the interactions to a short range.

3.4 Simulation method

The linear and nonlinear response of each nanostructure was simulating using the finite element solver COMSOL using nonlinear scattering theory as detailed in the previous chapter. Periodic boundary conditions with a 500 nm periodicity in both lateral dimensions and a 3000 nm length in the propagation dimension were used. The geometry was rounded
Asymmetry Ratio $\mathcal{R} = \frac{L_y}{L_{\text{eff}}}$

Figure 3.2: Schematic of the metamaterial array used to examine Miller’s rule in nonlinear metamaterials. We study the nonlinear generation from metamaterial arrays in which the geometry varies gradually from a symmetric bar to an asymmetric U-shape. The second order susceptibility is expected to be extremely sensitivity to symmetry of the metamaterial. (a) The definition of the parameter space: the total length and asymmetry ratio of the nanostructures is changed throughout the array. (b) Second and third harmonic generation as a function of the nano-structure length and morphology. A confocal microscope is used to excite individual nanostructures with infrared laser pulses (1305 nm). The nonlinear emission is measured in transmission. Also, the linear transmission is characterized using a supercontinuum laser. (c) Scanning electron microscope image of the nanostructures. To avoid particle-particle coupling a period of 500 nm was used. This spectroscopy method allows us to separate the intrinsic variations in the nanostructure nonlinearities from the frequency dependent linear and nonlinear susceptibility of the metal constituents.
to a 5 nm radius of curvature. A uniform dielectric environment was used to account for the
2 nm ITO layer without introducing numerical instabilities. The simulations were performed
for normal incidence. The reciprocity calculations were performed by first exciting the
nanostructure with a plane wave at the pump wavelength, then calculating the nonlinear
polarization at every point on the nanostructure surface for second harmonic and bulk for
third harmonic. Another simulation was then performed by sending in a second wave from
the detector at the emission wavelength. The overlap integral was then performed, thus
calculating the effective nonlinear susceptibility\[110\]. The linear properties of the gold in
the visible and near infrared are described using an analytic model\[39, 40\] which accounts
for interband transitions by adding two additional poles to the standard Drude model and
agrees well with the experimental data from Johnson and Christy\[59\].

3.5 Experimental results and analysis

From the nonlinear confocal microscopy data, we find that the nanostructure with
the most obvious asymmetry, the highly curved U shaped structure, does not yield the
maximum second order nonlinear susceptibility as seen in Fig. 3.3. Instead, the largest
second harmonic emission is observed for an intermediate morphology. The transition be-
tween bar to U-shaped nanostructures and thus the degree of asymmetry is quantified
by an asymmetry ratio, defined as the ratio of the vertical protrusion to the total length
of the nanostructure. The asymmetry ratio varies from 0-0.3 along the horizontal axis
of the array (Fig. 3.2) with a constant volume to avoid volume dependent changes in the
nonlinearity\[69\]. We study resonant and non-resonant interactions of the metamaterial with
the pump pulse by varying the length of the nanostructure from 150-300 nm. This method
allows us to separate the intrinsic variations in the metamaterial nonlinearities from the
frequency dependent linear and nonlinear susceptibility of the metal constituents\[81\]. The
two dimensional arrays of nanostructures are then illuminated at normal incidence with 100
fs laser pulses at 1305 nm using confocal stage scanning microscopy. We find the maximum
of the second harmonic emission for a specific geometry that corresponds to (length, ratio) = (292 nm, 0.18) as shown in Fig. 3.3a. In stark contrast, we find that Millers rule fails
to predict the optimal nonlinear metamaterial, found to be at a much higher ratio in the
experiment (0.18 ± 0.02) than predicted by Miller’s rule (0.12 ± 0.02), as seen in Fig. 3.3b.
In addition, this result deviates from the intuition that the most asymmetric structure will
yield the most second order nonlinearity. The optimum nonlinear metamaterial can be fully
predicted by applying nonlinear scattering theory, which as we will show allows an accurate
prediction of the correct far field nonlinear susceptibility using the microscopic rather than
far field linear response of the metamaterial.

While the far field linear response fails to predict the nonlinear properties of the
metamaterial as Miller’s rule prescribes, we find that the microscopic description can ef-
fectively predict the nonlinear susceptibility. In nonlinear scattering theory, the nonlinear
emission is described by assuming a local nonlinear susceptibility tensor on the surface of the
noble metal. This relationship can be quantified using the Lorentz reciprocity theorem\[110\]
as

\[
E_{nl}(2\omega) \propto \int \chi_{nnn} E_n^2(\omega) \hat{E}_n(2\omega) dS
\] (3.1)
Figure 3.3: Nonlinear scattering theory vs. Miller’s rule: The experimental second harmonic emission for different geometries of nanostructure for $\lambda_{pump} = 1305\text{nm}$ normal incidence. The nanostructure with the most obvious asymmetry, the highly curved U shaped structure, does not yield the maximum second order nonlinear susceptibility: the largest second harmonic emission is observed for an intermediate morphology. (a) Experimental result of the SH emission as a function of the nano-structure length (y-axis) and morphology (x-axis). (b) The predicted SH emission following Miller’s rule as a function of the nanostructure length and asymmetry ratio. From the transmission spectrum, we calculate the log of the transmission at the fundamental and second harmonic wavelengths, yielding a quantity proportional to the extinction cross section. We then predict the relative nonlinear susceptibilities using Miller’s rule, $\chi^{(2)}_{miller} \propto \sigma_{ext}(\omega)^2 \sigma_{ext}(2\omega)$ (c) Comparison of the emission intensity as a function of the asymmetry ratio for the nonlinear experiment (red dots), Miller’s rule (blue dots), and the nonlinear scattering theory (black line). Miller’s rule predicts a different optimum geometry than seen in the experiment, while nonlinear scattering theory correctly predicts the optimum geometry.
where $E_{nl}(2\omega)$ is the nonlinear emission, $\chi_{nnn}$ is the local nonlinear susceptibility, and $E_n(\omega)$ and $E_n(2\omega)$ are the linear field of the fundamental mode and the mode at the second harmonic frequency normal to the surface of the nanostructure. This process is illustrated schematically in Fig. 3.4a. The value of the overlap integral depends on the local field and the relative sign of the contributions: destructive interference between contributions can easily occur, resulting in low nonlinear emission. On the contrary, good mode overlap, meaning constructive interference between the nonlinear polarization mode and its harmonics, will lead to very high far field nonlinear emission, resulting in an intimate connection between the microscopic linear polarization and the far field emission. By examining the microscopic origin of the far field nonlinear emission, we can quantify this relationship and get an intuitive physical understanding of violation of Miller’s rule. In contrast, the numerical simulation using nonlinear scattering theory predicts an asymmetry ratio (0.19) that is consistent with the experimental results as shown in Fig. 3.3c.

The microscopic contributions, which are complex valued, add up from each region of the nanostructure creating constructive or destructive interference in the far field emission. A physically intuitive understanding of the net nonlinear emission in metamaterials can be obtained by plotting the value of $P \dot{E}$ on a path around the nanostructure in a complex plane representation. The net nonlinear emission is proportional to the magnitude of the vector sum of the microscopic nonlinear sources. A large final magnitude corresponds to high nonlinear emission, which is a consequence of large polarizability and good overlap between the modes, while poor nonlinear emission can result from weak local fields as well as poor overlap or destructive interference between nonlinear generations in different regions. Four nanostructures with varying asymmetry ratios are plotted in Fig. 3.4b. For "bar-like" nanostructures with a low asymmetry ratio (purple), the trajectory in the complex plane is not straight, illustrating the destructive interference of the microscopic nonlinear sources. In the opposite limit, for the greatest asymmetry ratio of 0.33 (dark green), the line integral follows a straight path in the complex plane, but the overall length of the vectors is smaller than the optimal nanostructure. The reason for this effect is the smaller absorption cross section of the highly curved SRR compared to the bar. For the optimal nanostructure with an asymmetry ratio of 0.19 (brown), the path integral displays some curvature, but has the largest final radius. The optimal nanostructure for second harmonic emission represents a compromise between the absorption at the fundamental, the second harmonic, and the ability for the microscopic sources to combine constructively.

In the experiment, the second harmonic has a single peak along the vertical axis, indicating that the wavelength dependence is dominated by the resonance of one specific eigenmode at the fundamental frequency. To test the wavelength scaling, the nonlinear response was measured for different pump wavelengths. The second harmonic vs position on the array is shown in Fig. 3.5a, b and c for pump wavelengths of 1160 nm, 1220 nm, 1305 nm. The relation between the position of second harmonic emission peak and the excitation wavelength is linear (Fig. 3.5d), which corresponds to the linear relation between particle length and resonant wavelength. We find that although the optimum length shifts with the pump wavelength, the optimum asymmetry ratio remains fixed. This optimum asymmetry ratio depends on a delicate balance between the ability of the nanostructure to absorb light, which decreases roughly linearly with the length of the horizontal arm and its
Figure 3.4: The nonlinear emission into the far field originates from the overlap integral of the microscopic nonlinear polarization and the mode at the second harmonic. The multiplication (overlap) between the nonlinear polarization, which is calculated from the linear fields at the fundamental frequency \( \omega \) (for normal incidence) and the mode at the second harmonic yields the amount of far field emission. High second harmonic emission is a consequence of large polarizability and good mode overlap, while poor overlap or destructive interference between nonlinear generations in different parts of the nanostructure results in low SH emission. (b) The microscopic contributions, which are complex valued, add up from each region of the nanostructure creating constructive or destructive interference in the far field emission. Physically intuitive understanding of the net nonlinear emission is obtained by plotting the value of \( \mathbf{P} \mathbf{E} \) on a path around the nanostructure on a complex plane. The net nonlinear emission is proportional to the magnitude of the vector sum of the microscopic nonlinear sources. A large final radius corresponds to high nonlinear emission. Four nanostructures with varying asymmetry ratios pulse are plotted in (b). For bar-like nanostructures with a low asymmetry ratio (purple), the trajectory in the complex plane is not straight, illustrating the destructive interference of the microscopic nonlinear sources. In the opposite limit, for the nanostructure with the greatest asymmetry ratio with ratio=0.33 (dark green), the line integral follows a straight path in the complex plane, but the overall length of the vectors is smaller than the optimal nanostructure. The reason for this effect is the smaller absorption cross section of the highly curved SRR compared to the bar. For the optimal nanostructure with ratio=0.19 (brown), the path integral displays some curvature, but has the largest final radius. The optimal nanostructure represents a compromise between the absorption at the fundamental, the second harmonic, and the ability for the microscopic sources to combine constructively.
Figure 3.5: Wavelength dependence of the optimal nanostructure for second harmonic generation. Spatial dependence of second harmonic generation for different excitation wavelengths (a–c). The same arrays of nanostructure is illuminated with pulses of varying wavelength, and the geometry which produces the maximum nonlinearity is determined. The effective length and asymmetry ratio which produce the maximum second harmonic are plotted in (d) and (e), respectively. The optimum nanostructure length varies linearly with the wavelength, in agreement with the wavelength scaling of Maxwell’s equations. The asymmetry ratio of the nanostructure which produces the maximum second harmonic remains fixed as the length is varied; illustrating the wavelength independence of the optimum asymmetry ratio. To determine the optimum geometry a centroid was fitted to the spatial maps of the second harmonic from the arrays of nanostructures.
ability to generate an asymmetric near field current pattern, which depends on the length in the vertical direction ($L_Y$ in Fig. 3.2). The constant relation that was observed in Fig. 3.5e, suggests that such scalability will be valid also at different wavelength as long as the linear relation between the resonance frequency and the length of the nanostructure holds. This conclusion adds considerable intuition to the design of nonlinear metamaterials.

There are some special cases where Miller’s rule can serve as an approximation to guide the design of nonlinear nano-structures. One example is third order nonlinearities, such as third harmonic generation, which do not have a symmetry breaking requirement, so for dipole modes, the electric field and nonlinear polarization will add up constructively when integrated over the nanostructure. This eliminates one mechanism for the failure of Miller’s rule. To demonstrate this, we have measured the third harmonic emission (Fig. 3.7a) from the same array of nanostructures. The third harmonic generation was analyzed in the same manner as done for the second harmonic case, i.e. according to Millers rule based on far field polarizabilities. In the THG case, we find that Miller’s rule (Fig. 3.7a,c) predicts a similar dependence on geometry as seen in the experiment. The implication of such a result is that the emission as a function of particle geometry can be reasonably described by the far field radiation properties, largely because third harmonic is a dipole allowed process and the nonlinear polarization for such modes will add up constructively. This explains the good agreement that was achieved in prior research on the nonlinear oscillator descriptions of experimental results for a wide range of geometries[53, 86, 47]. One small difference is that the far field theory (Fig. 3.7c) predicts a steeper drop in third harmonic intensity with the ratio than what is observed in the microscopic theory and the experiment. The third harmonic emission from the U-shape is larger than expected from the nonlinear oscillator model due to the enhanced local fields associated with the internal corners of the nanostructure as well as the changing ratio of absorption to scattering for the different structures[57].

### 3.6 Single nanostructure experiments

In order to verify that the linear-to-nonlinear predictions are indeed a single particle property, we have also performed the same experiments on another nanostructure array with larger particle spacing (2000nm). In this experiment, the particles were addressed individually, in contrast to the simultaneous illumination of 10 particles as in the experiments of 500nm. The results show how the conclusions drawn from the nonlinear scattering theory do not change with increased particle spacing. The experimental data and the conclusions from the experiments on the 2000 nm period sample are shown in Fig. 3.6.

### 3.7 Conclusions

In this Chapter, we have systematically studied the geometry dependence of metamaterial nonlinearity, demonstrating the validity of nonlinear scattering theory and thus the importance of the mode overlap between the nonlinear polarization and its harmonic modes for efficient nonlinear light emission. We show that due to varying mode overlap, Miller’s rule cannot describe metamaterial nonlinearities in all cases. We demonstrate the validity
Figure 3.6: (a) Experimental second harmonic (red) and third harmonic (blue) generation as a function of position for the arrays depicted in Fig. 3.1 with a (a) 3000 nm distance between particles and a (c) 500 nm distance between particles. (b,c) cross section along the horizontal axis (asymmetry ratio). We find that the highest second harmonic intensity occurs for the same aspect ratio for both inter-particle spacings.

of this approach for second and third harmonic generation with experiments on arrays of nanostructures in which the morphology is varied from elongated bars to U-shapes. Those were also analyzed in a geometrical approach, bringing a physical understanding to the violation of Miller’s rule in plasmonic nanostructures. These experimental and theoretical methods can be used to calculate, predict, and demonstrate other perturbative parametric processes, including multiple wave mixing and parametric down conversion in arbitrary nanostructures.
Figure 3.7: Analysis of third harmonic emission for different geometries of nanostructure for $\lambda_{pump} = 1305$ nm at normal incidence. (a) Experimental result for the TH emission as a function of the nano-structure length (y-axis) and morphology (x-axis). (b) The experimental third harmonic intensity (blue dots) and the Miller’s rule prediction (green plus) as a function of the ratio parameter. (c) The predicted third harmonic emission following Miller’s rule as a function of the nanostructure length and ratio, which is calculated from the cube of the linear extinction cross section. (d) The linear extinction cross section ($\sigma_{ext}(\omega)$) at the pump wavelength (for a horizontal polarization) as a function of the nanostructure length and ratio. (e) The experimental third harmonic intensity (blue dots) and the Miller’s rule prediction (green plus) as a function of the nanostructure length ($L_{eff}$).
Chapter 4

Phase and refractive index characterization of metamaterials

Characterizing the linear response of optical metamaterials is an important step before investigating the nonlinear properties. The linear refractive index plays a central role in determining the phase matching of nonlinear processes and is required to validate metamaterial designs. Time domain phase measurements with electronic detectors are not possible at optical frequencies due to the relatively slow speed of electronic detectors. The fastest response time for electronic detectors is on the order of picoseconds while a single cycle at a wavelength of 800 nm is 2.7 femtoseconds. To resolve the phase of a single cycle of the optical field, one must interfere light with itself in a homodyne measurement. The methods for performing phase measurements range in complexity from a Michelson interferometer which measures phase through linear interference of a sample and reference path to state of the art ultrafast pulse characterization techniques such as frequency-resolved optical gating (FROG)[61] or multiphoton intrapulse interference phase scan (MIIPS)[141] which use wave mixing in nonlinear crystals. Linear techniques such as interferometry cannot measure the phase of a complex electric field since they measure the intensity autocorrelation which is equivalent through the Wiener-Khinchin theorem to a spectrally resolved intensity measurement. However, linear techniques are capable of measuring the relative phase between two optical fields. This enables a full characterization of the refractive index of materials using linear interference, without the challenges associated with nonlinear pulse measurement techniques such as phase matching and power requirements inherent in nonlinear phase measurement techniques.

In this chapter we detail the development of a phase measurement setup based on spectrally and spatially resolved interferometry (SSRI)[103, 84, 85, 17, 16]. Our work is the first application of SSRI to nanostructures and metamaterials. We developed an easy to use setup using reflective optics which can rapidly measure optical phases as small as lambda/300 over more than an octave of bandwidth. In the next chapter, the phase measurement setup will be used to measure the phase response of negative and zero index optical metamaterials.
4.1 Spectrally and spatially resolved interferometry

The unambiguous determination of optical refractive indices of metamaterials is a challenging task for device applications and the study of new optical phenomena. We demonstrate here simple broadband phase measurements of metamaterials using spectrally and spatially resolved interferometry. We study the phase response of a -shaped metamaterial known to be an analog to electromagnetically induced transparency. The measured broadband interferograms give the phase delay or advance produced by the metamaterial in a single measurement. The presented technique offers an effective way of characterizing optical metamaterials including nonlinear and gain-metamaterial systems.

Metamaterials have demonstrated phenomena not thought possible decades ago, such as optical super-resolution and cloaking [119]. As metamaterials are used to study more nonlinear, ultrafast, and gain phenomena, the extraction of index information from intensity measurements and simulations will grow more challenging and error prone, so there is a need for accurate and fast optical phase measurements. Various approaches exist to measure the phase behavior of metamaterials. Researchers have fabricated structures out of or next to the metamaterial allowing them to observe the effects of the phase in the far field[133, 62]. Polarization walkoff interferometry was used for metamaterial and single nanoparticle phase measurements[35, 102, 126, 134]. A Michelson interferometer was used to measure the index of a fishnet metamaterial[34]. In this work, we demonstrate a simple, fast, broadband and accurate method for measuring the phase of the light transmitted by a metamaterial array based on spatially and spectrally resolved interferometry. We report a white light interferometer using reflective optics which can measure the phase change induced by a metamaterial across a broad wavelength range, limited only by the light source bandwidth and detector sensitivity. We pass white light through the sample and a reference path then recombine the light on the input slit of an imaging spectrometer with different vertical angles (Fig. 4.2b) in order to produce spatial fringes along the vertical axis of the image plane (Fig. 4.3a). The positions of the maxima and minima of this interference
Figure 4.2: Top (a) and side (b) views of the experimental setup for broadband interferometry. BS: beamsplitter cube. M1, M2: 25 mm focal length 90 degree off axis parabolas. M4 and M5: folding mirrors for the reference path, to produce the angle ($\alpha$) between the sample and reference paths.

The fringe pattern are proportional to the phase of each wave and the periodicity is proportional to the angle [15]. A change in the optical length of either beam path causes the fringe pattern to shift vertically. This technique is known as spectrally and spatially resolved interferometry (SSRI) [103]. It has seen many applications in the field of ultrafast optics [84, 85, 17, 16]. A summary of the history and analysis of the accuracy of the technique is given in Ref [15]. The use of SSRI with reflective focusing optics allows broadband phase measurements of small scale (50 µm) metamaterial arrays.

4.2 Experimental Setup

We used a reflective Mach-Zehnder interferometer to perform the broadband optical phase measurement on a metamaterial. As the light source we use a supercontinuum laser (Fianium SC450) spanning a wavelength range of 400-2400 nm. In our experiment we split the incident light into a sample and a reference path using a non-polarizing beamsplitter cube. We focus the light from the sample path onto the metamaterial using a 20 mm effective focal length off axis parabolic mirror, recollimate the light using a similar parabolic mirror (M1 and M2 in Fig. 4.2a), then reflect the light into the spectrometer using M3. The reference beam is reflected into the spectrometer using mirrors M4 and M5. The difference between the height ($y$) of the two beams at M4 and the distance ($L$) from the spectrometer determines the angle, and thus the periodicity of the fringes. Previous works on SSRI have found that approximately 20 fringes give the highest accuracy [15]. The two beams are then overlapped without focusing onto the input slit of the imaging spectrometer. The image is acquired with an InGaAs infrared camera (SU640KTS-1.7RT, 640x512 pixels, with a pitch of 25 µm) placed at the output port of the spectrometer (angular dispersion of 150 lines/mm)
and linear dispersion of 0.5 nm/pixel). We set the length of the reference path equal to the length of the sample path and place an identical substrate in the reference path. Typical images of the fringes are shown in Fig. 4.3a,b. Cross sections of the interferograms are shown in Fig. 4.3c,d. We compare interferograms acquired on the sample and adjacent to the sample using a Fourier based technique. We take the Fourier transform (FT) along the vertical axis then find the maximum of the absolute value of the FT, which corresponds to the period of the fringes. The arc tangent of the imaginary divided by real components determines the phase. Subtracting the two results yields the relative phase.

4.3 Phase characterization of EIT metamaterials

The described technique was used to measure the phase change produced by a π-shaped metamaterial known to produce an optical response analogous to electromagnetically induced transparency (EIT) in atomic systems[147, 70] or equivalently a type of Fano resonance[75]. The metamaterial consists of a vertical bar and a pair of horizontal bars
which is spaced a varying distance from the vertical bar. Near infrared light can only couple to a bar through the longitudinal plasmon mode associated with the long axis of the bars. In this experiment, we use vertically polarized light which excites a plasmon mode in the vertical bar and in the horizontal bar through near field coupling. The coupling strength between the vertical and horizontal bars can be tuned by changing the distance, and therefore samples with different gap distances have drastically different phase responses. Two gap distances were measured: 10nm (Fig. 4.4a), corresponding to the blue curve in Fig. 4.4c-f, which exhibits a distinct mode splitting and 80nm (Fig. 4.4b), which has a nearly Lorentzian resonance due to the weak coupling between the bars. The samples were made of 35nm thick gold deposited with electron beam evaporation and patterned with a standard electron beam lithography and liftoff process. The structure dimensions are given in Fig. 4.4.

4.4 Theory-experiment comparison

For comparison with the experimental results, we performed numerical simulations of the phase in transmission using the finite element solver Comsol. The simulation was performed with a substrate index of 1.52 with periodic boundary conditions (sample plane) and scattering boundary conditions normal to the plane of the structures, with a domain size of 0.6x0.6x5 µm. In the simulation, we used refractive index data from Johnson and Christy[59] with the damping rate of the gold increased by a factor of three to account for the increased losses due to surface scattering from the rough edges of the nanostructures and grain boundaries in the gold. The structure geometry was scaled by +5% to compensate for differences between the designed and fabricated structure. Comparing the experimentally determined phases (Fig. 4.4c) and simulated phases (Fig. 4.4d) one finds a good agreement. A possible explanation for the differences between simulation and experiment is uncertainties in the material parameters and deviations from designed dimensions. The phase response of the metamaterial array can be understood from the classical light scattering theory. The scattered field has a phase change from one side of the resonance to the other, while the phase of the incident field is unchanged[14]. In the far field, the sum of the two fields is measured. The summation is weighted by the scattering cross section, giving a smaller measured phase change further from the resonance where the metamaterial scatters less efficiently. For the weakly coupled system (Fig. 4.4b) we find a zero crossing in the phase at the same point as the dip in the transmission spectrum. We see a positive phase shift at longer wavelengths and a negative phase shift at shorter wavelengths, consistent with the result for a Lorentz oscillator. In this case, the general shape of the phase curve is similar to the derivative of the transmission with wavelength. For the strongly coupled system, the phase change can no longer be qualitatively understood from the derivative of the transmission with wavelength. Such a model would predict three zero crossings in the phase, due to the three points where the derivative of the transmission is zero. We see only a single zero crossing in both the experiment and simulation. Decreasing the loss of the system would produce the three zero-crossings in the phase that are typically found in an EIT-like response[44]. Fabricating the sample from single crystal metals is a potential route to reducing the losses and producing the expected zero crossing[55]. The measured
Figure 4.4: Scanning electron microscope (SEM) images of the nanostructure arrays with a gap of 10 nm (a) and 80 nm (b). The dimensions of the vertical bars are 200 x 60 nm, and the horizontal bar 160 x 50 nm with a separation of 90 nm. The pitch is 600 nm with an array size of 50 x 50 µm. Measured (c) and simulated phase (d). The corresponding measured (e) and simulated transmission (f). The average standard deviation for the measured phase is 0.02 radians (λ/300)
phases allow the calculation of the group delay dispersion (GDD). The maximum and minimum GDD were found on the short and long wavelength sides of the transmission minima with values of $\pm 200\,fs^2$ and $\pm 100\,fs^2$ for the 80 nm and 10 nm gap samples, respectively. When performing multiple measurements, the main difference between consecutive scans is a vertical shift of the phase vs. wavelength. The errors for 3 consecutive measurements are shown in Fig. 4.4c and are 0.02 radians on average. We found the following factors to be important for accurate results: a box around the entire setup to minimize changes in the optical path length of each arm due to air disturbances, smooth substrates, and a large sample area relative to the beam waist. The accuracy of the phase measurements is also influenced by factors such as fringe visibility and uniformity, spatial and spectra resolution, camera noise, and beam uniformity which are discussed in Ref[15].

4.5 Conclusions

In conclusion, we have demonstrated broadband phase measurements of a metamaterial exhibiting EIT in the near infrared with an accuracy of $\lambda/300$. This measurement technique will allow rapid and accurate phase characterization of active and nonlinear metamaterials, improving the understanding of these systems and aiding the development of practical metamaterial devices.
Chapter 5

Nonlinear optics in zero index media

Phase-matching is a critical requirement for coherent nonlinear optical processes such as frequency conversion and parametric amplification. Phase-mismatch prevents microscopic nonlinear sources to combine constructively, resulting in destructive interference and thus very low efficiency. We report the experimental demonstration of phase-mismatch free nonlinear generation in a zero index optical metamaterial. In contrast to phase-mismatch compensation techniques required in conventional nonlinear media, the zero index eliminates the need for phase-matching, allowing efficient nonlinear generation in both forward and backward directions. We demonstrate phase-mismatch free nonlinear generation using intrapulse four-wave mixing; observing a forward to backward nonlinear emission ratio close to unity. The removal of phase matching in nonlinear optical metamaterials may lead to new applications, such as multi-directional frequency conversion and entangled photon generation.

Nonlinear optics, the study of phenomena occurring when optical properties of a material are modified by the presence of light, plays a critical role in frequency conversion, nonlinear spectroscopy, and in the generation of new light sources\cite{121, 18}. A major problem in nonlinear optics is the inherent phase-mismatch between the interacting waves propagating inside the nonlinear materials. This effect originates from material dispersion and causes a lack of optical momentum conservation between the photons involved in the nonlinear process. The phase-mismatch prevents the constructive addition of the nonlinear fields; resulting in destructive interference and poor generation efficiency. To increase the amounts of nonlinear light, a compensation technique must be used. The most widely used methods (Fig. 5.1A), include birefringence phase-matching, angle phase matching and quasi phase matching (QPM)\cite{5, 56, 4, 113}. Implementing each technique poses a number of challenges. The birefringence phase matching technique uses the polarization dependent indices to match the phase velocities of the interacting waves, but is limited to birefringent materials\cite{121, 18}. Angle phase matching uses geometrical alignments of the interacting waves to compensate the phases, but the non-collinear optical arrangement limits the interaction length\cite{121, 18}. Quasi phase matching cancels out the inherent phase-mismatch using artificial momentum introduced by periodic and/or aperiodic poling of nonlinear crystals,
which is restricted to certain nonlinear crystals and provides a limited range of mismatch that can be compensated. Moreover, all compensating schemes work only in a specific direction: either in the forward direction\cite{5, 56, 4, 113} or the backward direction\cite{50, 21}, but not both. This restriction arises because the phase matching process represents a balance between the momenta of the photons involved in the nonlinear interaction; a balance that is disturbed when the momentum of one photon changes sign due to a direction change. We show that the requirement for phase matching can be eliminated using a metamaterial with a zero refractive index. In a zero index material\cite{133, 3, 136}, the photons carry zero momentum and satisfy momentum conservation for any combination of photon directions, thereby allowing the nonlinearly generated waves to coherently build up in both forward and backward directions. We demonstrate the nonlinear dynamics through four wave mixing (FWM) in a metamaterial with zero refractive index. Equal amounts of nonlinearly generated waves are observed in both forward and backward propagation directions, matching well with the predictions from nonlinear scattering theory\cite{110}. In contrast, the forward and backward FWM generations are drastically different for metamaterials with a positive or a negative index, as expected from phase matching considerations. Metamaterials allow us to tailor the linear electromagnetic response and introduce new regimes of interaction between radiation and matter\cite{135, 97, 120, 119, 41}. The nonlinear properties of negative index metamaterials have been explored both theoretically\cite{98, 100, 99, 114} and experimentally\cite{111, 130, 32, 89, 105}. The rich nonlinear dynamical behavior in metamaterials promises the realization of novel effects such as backward mirrorless parametric amplification\cite{98, 100}, novel quantum switches\cite{99} and cavity-free microscopic optical parametric oscillators\cite{114}.

5.1 Theory

To illustrate the phase-mismatch free nonlinear wave interactions in a zero refractive index material, without loss of generality, we consider a degenerate FWM process, in which the pump, signal, and idler photons have approximately the same wavelength $\omega_{\text{signal}} \approx \omega_{\text{pump}} \approx \omega_{\text{idler}}$. In this process, the phase (momentum) mismatch in the forward and backward propagation directions are $\Delta k_\pm = |2k_{\text{pump}} - k_{\text{signal}} \mp k_{\text{idler}}|$. The subscript + and - represent the forward and backward directions, respectively. In a conventional degenerate FWM system, when one of the propagation directions, say the forward direction, is perfectly phase-matched, i.e. $(\Delta k_+ \to 0)$, the phase is then poorly matched in the backward propagation direction where $\Delta k_- \approx 2|k_{\text{idler}}|$. For a metamaterial with a zero refractive index at the idler frequency, however, the phase-mismatch in both the forward and backward directions is zero. This important distinction between forward and backward nonlinear propagation allows us to explore nonlinear generation for negative, positive and zero index regimes. We expect that in the case of negative or positive index, due to the fact that $(\Delta k_+ \to 0)$, while $(\Delta k_- \neq 0)$, the generated forward light is accumulated in phase, while the generated backward light is not, thus causing the degenerated FWM intensity to grow monotonically for the forward direction and to oscillate for backward direction (Fig. 5.1, B and D). On the contrary, in the zero index regime, $(\Delta k_+ \to 0)$, and also $(\Delta k_- \neq 0)$, making both the generated forward light and backward light accumulate
Figure 5.1: The role of phase mismatch. Avoiding phase mismatch is critical for obtaining efficient nonlinear conversion. (A) The most widely used methods for phase matching are: birefringence phase matching, angle phase matching, and quasi-phase-matching (QPM), which allow compensation either in the forward or backward directions, but not both. In contrast, a zero index metamaterial creates a phase-mismatch free environment for nonlinear propagation, eliminating the requirement for phase matching. In the microscopic picture of nonlinear generation, each source coherently emits equally in both directions, acquiring a phase which is proportional to the refractive index (B,C) as it propagates. All of these sources add up coherently to generate the net nonlinear emission. In a zero index medium (C), the radiation from all nonlinear sources acquire no phase as they propagate, guaranteeing a constructive interference and an increase of the signal in both directions with propagation length (E). In contrast, in a finite index medium (B), the emission acquires phase as it propagates, leading to destructive interference when the process is not phase matched. For example, in a degenerate four wave mixing process in which \( \omega_{\text{signal}} \approx \omega_{\text{pump}} \approx \omega_{\text{idler}} \), the sources destructively interfere in the backward direction (D) resulting in weaker backward emission.
in phase, having the same yield in the forward and backward directions (Fig. 5.1, C and E). This is in stark contrast to nonlinear generation in negative index materials, where the phase-mismatch parameter has a finite value[98, 100, 99, 114]. Phase-mismatch free zero index material allows the nonlinear process to be efficient regardless of directionality, and the need to carefully balance between the momenta of the waves involved in the nonlinear interaction is eliminated. In our experimental realization, the pump pulse has a positive Poynting vector in the material so the direction of energy flux is the same as in free space. In contrast, the nonlinear emission has an energy flux in both directions, with the relative amounts influenced by the phase matching. The refractive index controls the direction of the phase velocity and canonical momentum relative to the direction of the energy flux[135, 8], which influences the direction of energy propagation through the phase matching. In a zero index medium, as seen, the energy flux in both directions is equal. Compared to the poorly phase-matched case, more energy will be extracted from the pump in a zero index material.

5.2 Experimental setup - intra-pulse four wave mixing

Figure 5.2A shows the experimental apparatus for single-shot FWM which allows intrapulse wave mixing between the different spectral components of ultrashort pump laser pulses[36]. Before impinging on the samples the transform limited pump pulse is first amplitude shaped to remove the long wavelength tail in the spectral domain. The generated intrapulse FWM is measured in both forward and backward directions within this filtered spectral regime. This method eliminates the needs to overlap two laser pulse temporally and spatially, and maximizes the nonlinear yield. An example measurement spectra (Fig. 5.2B) shows both the generated FWM signals, with their relative strengths. Note that the pump is far from depleted by the nonlinear process as evident by the much weaker FWM signal than that of the pump. Operating in the weak field regime (the generated intensity of the idler is on the order of $10^{-5}$ of the pump intensity) allows us to analyze the experimental observations with a perturbative approach.

We verify the nonlinear origin of the emission by measuring the cubic scaling with pump power and quadratic spectral phase dependence. Scaling the power of the pump will influence the generated nonlinear signal in a nonlinear manner (power of 3), and the remaining pump in a linear manner. As can be seen in the figure below, the signal at the short wavelengths, which is the remaining pump wavelength components, after using a long pass filter (step 6 in the experimental flow), scales linearly with the pump power. In contrast, the long wavelengths, which were generated by the nonlinear interaction, increase in a nonlinear manner with a cubic relation with the increasing of the pump power.

We have also used a heterodyne detection scheme, which is based on a pulse shaper manipulation, in order to accurately resolve the coherent parametric contribution. This was done by subtracting a spectral trace obtained with a stretched pulse from one obtained with a transform limited pulse. By doing so, the background, which originated from linear scattering and diffraction, could be removed and the nonlinear coherent response could be retrieved accurately. In the following, we show an example of spectral measurements as a function of the pump chirp value. In Fig. 5.4a, we present a two dimensional map of the measured spectra (vertical lines) as a function of the linear chirp that was added with
Figure 5.2: Experimental apparatus for nonlinear measurements (A) A transform limited 100 fs laser pulse centered at 1315 nm or 1510 nm was amplitude shaped through a spatial light modulator (SLM) to remove the long wavelength tail of the pulse (red). The pulse was passed through a fishnet metamaterial, which generates a four wave mixing nonlinear signal (green) in the backward and forward directions due to a third order nonlinearity. The four wave mixing signals were measured in an infrared spectrometer after filtering the pump pulse. The forward emission was measured without the flip mirror (M4). An analyzer was used before the spectrometer to control the detected polarization and a half wave plate (not shown) was used to control the pump polarization. (B) An example measurement of the emission spectrum, showing both the generated four wave mixing signal (green) and the filtered pump (red). The pump is undepleted by the nonlinear process as evident by the much weaker four wave mixing signal than that of the pump (the magnitude of the four wave mixing is $10^{-5}$ times the pump intensity), justifying the use of the perturbative approach in our analysis. (C) SEM image of the cross fishnet structures. Inset: angled view.
Figure 5.3: Power Scaling of the pump and four wave mixing in a single-shot FWM spectral measurement. Both the FWM signal and remaining pump are observed in the spectral measurements, and can be differentiated by their spectral location. At the bottom panel, several spectral measurements are plotted for different pump intensities. The remaining pump (at shorter wavelengths) is found to scale linearly with the pump power (upper left panel in red), while the four wave mixing signal is proportional to the cube of the pump power as expected for a third order nonlinear process (upper right panel in green).
the SLM. Only when the pulse is close to transform limited, a nonlinear FWM signal was generated. For a stretched pulse the nonlinear signal is significantly reduced. In Fig. 5.4b, we plot two measured spectral traces, one when the pump pulse was transform limited (solid red) and one when we stretch it using the pulse shaper (dashed black). The subtraction between the two measurements yields a clean nonlinear four wave mixing signals.

Figure 5.4: Heterodyne detection using pulse shaper. (A) Measured spectral trace as a function of linear chirp. As seen, only when the pulse is close to transform limited (meaning that its spectral phase was flat, i.e. shortest pulse), a nonlinear FWM signal was generated. In a stretched pulse the nonlinear signal is significantly reduced. (B) Two measured spectral traces when with optimal pump pulse (solid red) or stretch pulse (dashed black). We have used the subtraction between the two, in order to retrieve a clean nonlinear four wave mixing signals.

5.3 Sample

We have chosen the fishnet metamaterial structure[133], a stack of metal-dielectric multilayers with perforated holes (Fig. 5.2). Fishnet metamaterials are widely used negative index materials at optical frequency, due to their low loss, well understood linear properties, and robust fabricability. Our fishnet metamaterial consists of 20 alternating layers of 30 nm gold and 50 nm Magnesium Fluoride on a 50-nm-thick silicon nitride membrane. The magnetic moments, created by the antiparallel currents in neighboring conductive layers, and the electric responses of the perforated metallic thin films provide either a positive, zero, or negative refractive index regime depending on the wavelength.

5.4 Phase characterization

The refractive index of the fishnet structures were measured experimentally by using spectrally and spatially resolved interferometry. The measurement technique, detailed in Ref.[95], is performed using a white light interferometer and reflective optics, which can measure the phase change induced by a metamaterial across a broad wavelength range, limited only by the light source bandwidth and detector sensitivity (shown to have an
accuracy of greater than $\lambda/300$ [95]). The white light is passed through the sample and a
reference path then recombines on the input slit of an imaging spectrometer with different
vertical angles in order to produce spatial fringes along the vertical axis of the image plane.
The positions of the maxima and minima of this interference pattern are proportional to
the phase of each wave and the periodicity is proportional to the angle. A change in
the optical length of either beam path causes the fringe pattern to shift vertically. A
measurement of the interferogram is taken with and without the sample. The phase shift in
the transmitted light caused by the sample is determined from a Fourier analysis of the two
interferograms. In Fig. 5.5 we present the retrieved experimental refractive index from the
measured transmission phase. The zero crossing of the index is approximately 1325-1340
nm for the sample with period of 750nm and cross lateral holes of dimensions 475 nm x 175
nm (Fig. 5.2C).

Figure 5.5: Refractive index measurements. (A) The measured refractive index of the sam-
ples, using the spectrally and spatially resolved interferometry technique. A measurement of
the interferogram is taken with and without the sample. The phase shift in the transmitted
light caused by the sample is determined from a Fourier analysis of the two interferograms

Using the measured refractive index values of the fishnet structures, the phase-
mismatch values can be calculated as a function of wavelength for forward and backward
propagation (Fig. 5.6A). The forward phase-mismatch is ($\Delta k_+ \rightarrow 0$) for all wavelengths,
while $\Delta k_- \text{ has different values across the zero index wavelength. However, in the wavelength}
regime of zero refractive index (1330 nm), both the forward and backward directions are
phase-mismatch free.

5.5 Experimental results

The intrapulse FWM signal in the zero index regime (where the refractive index
changes its sign) is shown for both the forward (solid purple) and the backward (dashed blue)
directions (Fig. 5.6B). The observed nonlinear yield is about the same in both directions.
In contrast, in the negative index regime, the intensity of the degenerate FWM signals
in the opposite propagation directions are distinctly different (Fig. 3C). Due to the low
transmission at 1530 nm, a different fishnet metamaterial with 800nm period and perforated
Figure 5.6: Four wave mixing in metamaterials with and without phase mismatch (A) The phase-mismatch of the forward (purple) and backward (blue) four wave mixing as a function of wavelength, based on experimentally measured refractive indices. The forward phase-mismatch is near zero ($\Delta k_+ \rightarrow 0$) for all wavelengths, while the backward has a large phase mismatch ($\Delta k_- \neq 0$) except when the index is near zero. (B) The measured four wave mixing process in the zero index regime has almost the same yield in both directions, illustrating the phase mismatch-free properties of zero index materials. (C) In contrast, the forward propagating four wave mixing (solid purple) is much stronger than the backward in the negative index regime, due to the phase mismatch. The upper inset in (B) and (C) shows the dependence of the nonlinear emission (with a horizontally polarized analyzer) on pump polarization, which has the characteristic $\cos^6 \theta$ of $\chi^{(3)}$ dynamics.
hole size of 560 nm by 250 nm was used. The zero crossing of the refractive index is at 1460 nm and the refractive index $\text{Re}[n] = -0.5$ at 1530 nm (See Fig. 5.5). The relative strength of the forward and backward FWM waves correlates with the predicted phase matching difference for zero and negative indices, as illustrated in Fig. 5.1. In particular, at the zero refractive index, the observation of a forward/backward FWM ratio of unity, indicates phase-mismatch free nonlinear interaction for both directions. Low transmission loss is critical for observing the effects of phase matching. High losses can also strongly affect the ratio between the forward and backward nonlinear emission for positive, negative, and zero index regimes. It decreases the overall nonlinear emission, and due to the shorter propagation length of backward emission, increases the backward emission relative to the forward. The fabricated fishnet has a relatively low loss in the zero-index regime, allowing us to observe the effects of phase matching rather than absorption.

The phase-mismatch free nonlinear generation can be further demonstrated by measuring the nonlinear emission as a function of the polarization angle of the pump for the forward and backward FWM, shown in insets of Fig. 3, B and C for the zero index and the negative index materials, respectively. The relative strength between the forward and backward nonlinear waves in the zero index region is again about the same for all polar angles, while in the negative index regime, the forward nonlinear waves remain stronger than the backward for all polarizations. The polarization dependent FWM (with a horizontally polarized analyzer) matches well the analytical curve of $\cos^6 \theta$, a characteristic response of a third order nonlinearity intensity signal. This angular dependence is particularly important as it means that the nonlinearity originates from the light that is coupled into the metamaterial.

5.6 Numerical simulations

The numerical simulations of the nonlinear emission in the paper is based on a microscopic theory for nonlinear generation and propagation called nonlinear scattering theory[110]. The linear electric fields are calculated from the material parameters and microscopic structure using the finite element method. These are then used to derive the far field nonlinear response, without relying on standard wave equations with simplified approximations for bulk materials. We first calculate the linear electric fields in the frequency domain using the full wave finite element solver COMSOL. From these fields we then calculate the nonlinear polarization. We calculate the far field nonlinear emission using the following integral form of the reciprocity theorem[110]:

$$E_{FBM}^{FWM}(\omega) \propto \int \int \int_{\text{metal volume}} P_{i}^{NL}(\omega) E_{b,f,i}(\omega) \, d^3r$$

where $P_{i}^{NL}(\omega) = \chi^{(3)}_{iii} E_{i} E_{i} E_{i}$. We assume the $\chi^{(3)}_{iii}$ of gold to be dominant, which has 3 order of magnitude higher nonlinearity than MgF$_2$ [7, 106, 94]. This procedure allows us to simulate the far field nonlinear response from the near field linear response of the fishnet metamaterial. In the simulation we use a geometry composed of 50 nm silicon nitride ($n = 2$), and N pairs of 30 nm gold ($\omega_p = 1.34 \times 10^{16} \text{Hz}$, $\gamma_p = 1.1 \times 10^{14} \text{Hz}$) and 50 nm MgF$_2$ ($n = 1.37$) with a sidewall angle of 4.3 degrees, where N is the number of functional layers.
5.7 Comparison with numerical simulations

The numerical simulation of the dynamics as a function of thickness around the zero index predicts a ratio of unity for forward/backward nonlinear emission (see Fig. 5.7a); whereas in the negative index regime, the forward generated nonlinear signal has a much higher yield than that of the backward signal (Fig. 5.7b) for a thick fishnet of 20 layers. In contrast, for thin fishnets (less than 5 layers), the forward/backward ratio is unity regardless of the index of refraction, indicating that phase matching is not important in these structures[130, 32, 89, 105]. The nonlinear simulation is performed with nonlinear scattering theory[110], which calculates the nonlinear properties using the linear results from a full wave simulation of the metamaterial, taking into account the fabricated structure geometry and material losses. To further confirm that phase plays a dominant role in the dynamics and not the loss or surface effects, we artificially remove the phase-mismatch in the nonlinear simulation while keeping loss, thus forcing all waves to add up constructively. We find that the behavior in the zero index region is unchanged, indicating that phase matching plays little role. In contrast, in the negative index region, the forward and backward emissions are now nearly equal, indicating that phase matching is critical in this region. It is worth noting that perfect phase matching using a zero refractive index is different than quasi-phase matching[56, 111], in which the effective momentum supplied by a periodic structuring is used to compensate for a phase mismatch. In these experiments, the sub-wavelength spacing of the layers (80 nm) does not provide sufficient effective momentum to phase-match the backward nonlinear generation.

5.8 Conclusions

The concept of phase-mismatch free nonlinear interaction in zero index materials provides a new degree of freedom in controlling the nonlinear dynamics in a metamaterial and can be further explored in other nonlinear processes such as coherent Raman for remote sensing applications or spontaneous parametric down-conversions for entangled photon generation. The design of multi-valued or broadband zero index materials opens the opportunity to achieve phase-mismatch free dynamics for simultaneous nonlinear processes.
Figure 5.7: Numerical simulations of nonlinear emission as a function of metamaterial thickness. (A) for zero index materials the nonlinear yield in the forward and backward directions is similar and both increase with material thickness due to constructive interference. (B) In the negative index regime, only the forward emission increases with material thickness, while the backward emission decreases due to the large phase-mismatch of the finite index material. As the metamaterial thickness increases, eliminating phase mismatch with zero index metamaterials becomes important for maximizing the nonlinear yield. The nonlinear simulations, performed with nonlinear scattering theory, calculate the nonlinear properties from a linear full wave simulation of the metamaterial, taking into account the real geometry and material losses.
Chapter 6

Resonant phase matching of Josephson traveling wave parametric amplifiers (JTWPs)

Josephson parametric amplifiers\cite{23, 10, 52, 109, 37} routinely approach quantum-noise-limited performance \cite{74, 67, 22, 79}, and are currently used in sensitive experiments requiring high-fidelity detection of single-photon-level microwave signals, such as the readout and feedback control of superconducting quantum bits \cite{92, 137, 58, 138, 51, 20, 107, 108}, and magnetometry with the promise of single-spin resolution \cite{52}. To obtain a large parametric gain, the interaction time with the material nonlinearity—the order-unity nonlinear inductance of the Josephson junction—must be maximized. Current Josephson parametric devices increase the interaction time by coupling the junction to a resonant cavity albeit at the expense of instantaneous bandwidth. In contrast, traveling wave parametric amplifiers\cite{30, 31, 124, 131} (TWPAs) achieve long interaction times by utilizing long propagation lengths rather than employing multiple bounces in a cavity, thereby avoiding the inherent gain-bandwidth tradeoff associated with cavity based devices. A major challenge in the design of TWPAs, however, is that optimum parametric gain is only achieved when the amplification process is phase matched. TWPAs based on Josephson junctions have been investigated theoretically\cite{42, 129, 143, 144} and experimentally\cite{139, 145}, but have not demonstrated sufficient gain, in part due to phase-matching limitations, to replace existing semiconductor amplifier technology. TWPAs based on the weaker nonlinear kinetic inductance of thin titanium nitride wires and phase matched through periodic loading have also been demonstrated\cite{54, 13}, but require significantly longer propagation lengths and higher pump powers to achieve comparable gain. In this chapter, we show that by adding a resonant element into the transmission line, phase matching and exponential gain can be achieved over a broad bandwidth.
6.1 Derivation of nonlinear wave equation

First we derive the nonlinear wave describing the dynamics of the JTWPA. The JTWPA is an LC ladder of Josephson junctions as shown in Fig. 7.1a. To derive the equation describing the propagation of a strong pump flux, we first calculate the Lagrangian for the array of Josephson junctions[140, 80, 38]. We then convert this to a Lagrangian density[91] by taking the limit of a small unit cell size \( a \) compared to the wavelength:

\[
\mathcal{L} = \frac{E_J}{a} \cos \left( \frac{a}{\phi_0} \phi_x \right) + \frac{C}{2a} \phi_t^2 + \frac{C_J a}{2} \phi_{xt}^2
\]  

(6.1)

where \( C_J \) is the junction capacitance, \( E_J = \phi_0 I_0 \) is the Josephson energy, \( I_0 \) is the junction critical current, \( C \) is the capacitance to ground, \( a \) is the unit cell size, and \( \phi_0 = \hbar/(2e) \) is the reduced flux quantum. We then derive the dynamical equation of motion from the Euler-Lagrange equation (see Appendix 1 for further details):

\[
\frac{a E_J}{\phi_0^2} \cos \left( \frac{a}{\phi_0} \phi_x \right) \phi_{xx} - \frac{C}{a} \phi_{tt} + C_J a \phi_{xxtt} = 0
\]  

(6.2)

We calculate the Lagrangian for the array of Josephson junctions[140, 80, 38] to be:

\[
L = \sum_n \left[ E_J \cos \left( \frac{\phi_{n+1} - \phi_n}{\phi_0} \right) + \frac{C_J}{2} \left( \frac{d\phi_{n+1}}{dt} - \frac{d\phi_n}{dt} \right)^2 + \frac{C}{2} \left( \frac{d\phi_n}{dt} \right)^2 \right]
\]  

(6.3)

where \( \phi \) is the magnetic flux in each unit cell, with the reduced flux quantum \( \phi_0 = \hbar/(2e) \), and the Josephson energy \( E_J = \hbar I_0/(2e) \) where \( I_0 \) is the junction critical current. We re-write this as a Lagrangian density[91]:

\[
L = \sum_n \left[ \frac{E_J}{a} \cos \left( \frac{\phi_{n+1} - \phi_n}{\phi_0} \right) + \frac{C_J}{2a} \left( \frac{\partial \phi_{n+1}}{\partial t} - \frac{\partial \phi_n}{\partial t} \right)^2 + \frac{C}{2a} \left( \frac{\partial \phi_n}{\partial t} \right)^2 \right] = \sum_n a \mathcal{L}
\]  

(6.4)

Taking the limit \( a \to 0 \), the system can be described as an integral over the Lagrangian density \( L = \int_0^x \mathcal{L} dx \). Approximating the differences as differentials the Lagrangian density is:

\[
\mathcal{L} = \frac{E_J}{a} \cos \left( \frac{\phi_{n+1} - \phi_n}{\phi_0} \right) + \frac{C_J a}{2} \left( \frac{\partial \phi_{n+1}}{\partial t} - \frac{\partial \phi_n}{\partial t} \right)^2 + \frac{C}{2a} \left( \frac{\partial \phi_n}{\partial t} \right)^2
\]  

(6.5)

We can derive the dynamical equations of motion from the Euler-Lagrange equations:

\[
\frac{\partial \mathcal{L}}{\partial \phi} - \frac{d}{dx} \left( \frac{\partial \mathcal{L}}{\partial (\partial \phi/\partial x)} \right) - \frac{d}{dt} \left( \frac{\partial \mathcal{L}}{\partial (\partial \phi/\partial t)} \right) + \frac{d^2}{dx dt} \left( \frac{\partial \mathcal{L}}{\partial (\partial^2 \phi/\partial x \partial t)} \right)
\]  

(6.6)

The exact nonlinear wave equation is then:

\[
\frac{a E_J}{\phi_0^2} \cos \left( \frac{a}{\phi_0} \phi_x \right) \frac{\partial^2 \phi}{\partial x^2} - \frac{C}{a} \frac{\partial^2 \phi}{\partial t^2} + C_J a \frac{\partial^4 \phi}{\partial t^2 \partial x^2} = 0
\]  

(6.7)

From this exact nonlinear wave equation, one can obtain a more easily solved approximate form by Taylor expanding the \( \cos \) term to the first nonlinear order, \( \cos(x) = 1 - x^2/2 \). One then obtains a nonlinear wave equation identical to the result of Yaakobi et al[143, 144].
To account for dissipation in the capacitors, one can add a term proportional to the time derivative of the flux.

\[
\frac{aE_J}{\phi_0} \cos \left( \frac{a}{\phi_0} \frac{\partial \phi}{\partial x} \right) \frac{\partial^2 \phi}{\partial x^2} - \frac{C}{a} \frac{\partial^2 \phi}{\partial t^2} + C_J a \frac{\partial^4 \phi}{\partial t^2 \partial x^2} - \frac{1}{aR} \frac{\partial \phi}{\partial t} = 0 \quad (6.8)
\]

### 6.2 Derivation of coupled wave equations for parametric amplification

Here we derive the coupled wave equations for a traveling wave parametric amplifier. Starting with the approximate form of the nonlinear wave equation derived above (or Eq. 22 from Ref. [24]):

\[
C_0 \frac{\partial^2 \phi}{\partial t^2} - \frac{a^2}{L} \frac{\partial^2 \phi}{\partial x^2} - C_J a^2 \frac{\partial^4 \phi}{\partial x^2 \partial t^2} = \frac{a^4}{2 L_0^4 L^4} \frac{\partial^2 \phi}{\partial x^2} \left( \frac{\partial \phi}{\partial t} \right)^2 \quad (6.9)
\]

we take the ansatz that the solutions will be forward propagating waves of the form:

\[
\phi = \frac{1}{2} \left[ A_p(x) e^{i(k_p x + \omega_p t)} + A_s(x) e^{i(k_s x + \omega_s t)} + A_i(x) e^{i(k_i x + \omega_i t)} + c.c. \right] \quad (6.10)
\]

Where \( A_m \) is the slowly varying amplitude, \( k_m \) is the wave vector, and \( \omega_m \) is the angular frequency. We substitute the above expression into the nonlinear wave equation then make the following approximations:

1. Neglect the second derivatives of the slowly varying amplitudes using the slowly varying envelope approximation: \( \left| \frac{\partial^2 A_m}{\partial x^2} \right| \ll \left| k_m \frac{\partial A_m}{\partial x} \right| \).

2. Neglect the first derivatives of the slowly varying amplitudes on the right side of the nonlinear wave equation (ie, in the nonlinear polarizability): \( \left| \frac{\partial A_m}{\partial x} \right| \ll \left| k_m A_m \right| \).

Considering only the left side of Eq. 6.9 and separating out the terms that oscillate at the pump, signal, and idler frequencies we get the following equation:

\[
\begin{align*}
C_0 & \frac{\partial^2 \phi}{\partial t^2} - \frac{a^2}{L} \frac{\partial^2 \phi}{\partial x^2} - C_J a^2 \frac{\partial^4 \phi}{\partial x^2 \partial t^2} \\
&= \frac{a^4}{2 L_0^4 L^4} \frac{\partial^2 \phi}{\partial x^2} \left( \frac{\partial \phi}{\partial t} \right)^2 \quad (6.11)
\end{align*}
\]

where \( m = p, s, i \). Defining the wave vector as \( k_m = \frac{\omega_m \sqrt{C_0 L}}{a \sqrt{1 - C_J L \omega_m}} \), Eq. 6.11 simplifies to:

\[
- \frac{i C_0 \omega_m^2}{k_m} e^{i(\omega_m + k_m x)} \frac{\partial A_m(x)}{\partial x} \quad (6.12)
\]

Now we consider the nonlinear component (the right side of Eq. 6.9). The propagation equation for the pump is:

\[
\frac{\partial A_p}{\partial x} - \frac{ia^4 k_p^5}{16C_0 L_0^2 L^2 \omega_p^2} A_p^2 A_p^* = 0 \quad (6.13)
\]
where we have neglected the terms proportional to the amplitudes of the signal and idler as they are much smaller than the pump field. The propagation equation for the signal and idler, neglecting terms which are quadratic in the signal and idler amplitudes:

\[
\frac{\partial A_s}{\partial x} - i \frac{a^4 k_p^2 k_s^3}{8C_0 I_0^2 L^3 \omega_s^2} A_p A_p^* A_s - i \frac{a^4 k_p^2 (2k_p - k_i)k_s k_i}{16C_0 I_0^2 L^3 \omega_s^2} A_p^2 A_i e^{i\Delta k_L x} = 0
\]  
(6.14)

\[
\frac{\partial A_i}{\partial x} - i \frac{a^4 k_p^2 k_i^3}{8C_0 I_0^2 L^3 \omega_i^2} A_p A_p^* A_i - i \frac{a^4 k_p^2 (2k_p - k_s)k_s k_i}{16C_0 I_0^2 L^3 \omega_i^2} A_p^2 A_s^* e^{i\Delta k_L x} = 0
\]  
(6.15)

Now we solve for the pump propagation, assuming no loss, and obtain:

\[
A_p(x) = A_{p,0} e^{i \frac{a^4 k_p^2 A_p A_p^*}{16C_0 I_0^2 L^3 \omega_p^2} x}
\]  
(6.16)

We substitute the solution for the pump field (Eq. 6.16) into Eqs. 6.14 and 6.15:

\[
\frac{\partial A_s}{\partial x} - i \alpha_s A_s - i \kappa_s A_i e^{i(\Delta k_L + 2\alpha_p)x} = 0
\]  
(6.17)

\[
\frac{\partial A_i}{\partial x} - i \alpha_i A_i - i \kappa_i A_s e^{i(\Delta k_L + 2\alpha_p)x} = 0
\]  
(6.18)

where the couplings are defined as:

\[
\alpha_s = \frac{2\kappa k_s^3 a^2}{LC_0 \omega_s^2}
\]

\[
\alpha_i = \frac{2\kappa k_i^3 a^2}{LC_0 \omega_i^2}
\]

\[
\alpha_p = \frac{\kappa k_p^3 a^2}{LC_0 \omega_p^2}
\]

\[
\kappa_s = \frac{\kappa (2k_p - k_i)k_s k_i a^2}{LC_0 \omega_s^2}
\]

\[
\kappa_i = \frac{\kappa (2k_p - k_s)k_s k_i a^2}{LC_0 \omega_i^2}
\]

\[
\kappa = \frac{a^2 k_p^2 A_p A_p^*}{16I_0^2 L^2}
\]  
(6.21)

To generalize these equations for arbitrary circuits, we make the substitution \( C_0 = 1/(i\omega Z_2) \) and express the pump amplitude in terms of the characteristic impedance and pump current: \( A_{p,0} = I_p Z_{\text{char}} / \omega_p \). The couplings are now:

\[
\alpha_s = \frac{2\kappa k_s^3 a^2 iZ_2(\omega_s)}{L\omega_s}
\]

\[
\alpha_i = \frac{2\kappa k_i^3 a^2 iZ_2(\omega_i)}{L\omega_i}
\]

\[
\alpha_p = \frac{\kappa k_p^3 a^2 iZ_2(\omega_p)}{L\omega_p}
\]

\[
\kappa_s = \frac{\kappa (2k_p - k_i)k_s k_i a^2}{L\omega_s}
\]

\[
\kappa_i = \frac{\kappa (2k_p - k_s)k_s k_i a^2}{L\omega_i}
\]

\[
\kappa = \frac{a^2 k_p^2 |Z_{\text{char}}|^2}{16L^2 \omega_p^2} \left( \frac{I_p}{I_0} \right)^2
\]  
(6.25)

We solve the coupled amplitude equations (Eqs. 6.18 and 6.19) by making the substitutions \( A_s = a_s e^{i\alpha_s x} \) and \( A_i = a_i e^{i\alpha_i x} \) to obtain:

\[
\frac{\partial a_s}{\partial x} - i \kappa_s a_s^* e^{i(\Delta k_L + 2\alpha_p - \alpha_s - \alpha_i)x} = 0
\]  
(6.26)

\[
\frac{\partial a_i}{\partial x} - i \kappa_i a_i^* e^{i(\Delta k_L + 2\alpha_p - \alpha_s - \alpha_i)x} = 0
\]  
(6.27)
These equations are analogous to the coupled amplitude equations for an optical parametric amplifier, which have the following solution:

\[
a_s(x) = \left[ a_s(0) \left( \cosh gx - \frac{i\Delta k}{2g} \sinh gx \right) + \frac{i\kappa_s}{g} a_s^*(0) \sinh gx \right] \frac{e^{i\Delta k x/2}}{\sqrt{1 + (\Delta k/2)^2}}
\]

\[
a_i(x) = \left[ a_i(0) \left( \cosh gx - \frac{i\Delta k}{2g} \sinh gx \right) + \frac{i\kappa_i}{g} a_i^*(0) \sinh gx \right] \frac{e^{i\Delta k x/2}}{\sqrt{1 + (\Delta k/2)^2}}
\]

where \(\Delta k\) and \(g\) are defined as:

\[
\Delta k = \Delta k_L + 2\alpha_p - \alpha_s - \alpha_i
\]

\[
= 2k_p - k_s - k_i + 2\alpha_p - \alpha_s - \alpha_i
\]

\[
g = \sqrt{\kappa_s\kappa_i^* - (\Delta k/2)^2}
\]

### 6.3 Derivation of the linear dispersion relation

Now we calculate the dispersion relation for the transmission line with the LC resonator shunt. For the considered circuit topology, the ABCD matrix is:

\[
\begin{pmatrix}
1 & -Z_1 \\
-1/Z_2 & 1 + Z_1/Z_2
\end{pmatrix}
\]

where

\[
Z_1 = Z_L || Z_{Cj} = \left( \frac{1}{i\omega L} + i\omega C_j \right)^{-1}
\]

\[
Z_2 = Z_C || Z_{res}
\]

\[
Z_{res} = Z_{Cc} + Z_{Cr} || Z_{Lr} = \frac{1 - (C_c + C_r)L_r\omega^2}{i\omega C_c(1 - C_rL_r\omega^2)}
\]

The wavevector and characteristic impedance in terms of the ABCD matrix elements are:

\[
k = \cos^{-1} \frac{A + D}{2}
\]

\[
Z_{char} = \frac{(A - D) - \sqrt{(A + D + 2)(A + D - 2)}}{2C}
\]

This wave-vector is then used in the coupled wave equations to calculate the gain.

### 6.4 Resonant phase matching proposal

The proposed traveling wave parametric amplifier consists of a Josephson-junction-loaded transmission line (Fig. 7.1a) with a capacitively-coupled parallel LC resonator shunt to allow phase matching. The LC resonator shunt (colored red in Fig. 7.1a,b) creates a stop
Figure 6.1: Resonantly phase-matched traveling wave parametric amplifier. (a) Signal photons are amplified through a nonlinear interaction with a strong pump as they propagate along the 2000 unit cell transmission line with a lattice period of \( a = 10 \mu \text{m} \). (b) In each unit cell a Josephson junction, a nonlinear inductor, is capacitively coupled to an \( LC \) resonator. The circuit parameters are: \( C_j = 329 \text{ fF} \), \( L = 100 \text{ pH} \), \( C = 39 \text{ fF} \), \( C_c = 10 \text{ fF} \), \( C_r = 7.036 \text{ pF} \), \( L_r = 100 \text{ pH} \), \( I_0 = 3.29 \mu \text{A} \). (c) The \( LC \) circuit opens a stop band (red) in the dispersion relation of the TWPA (black dashed) whose frequency depends on the circuit parameters. In the inset, we plot the pump frequency to phase-match a pump current of \( 0.3I_0 \) (blue), \( 0.5I_0 \) (purple), and \( 0.7I_0 \) (green), where \( I_0 \) is the junction critical current.

We now calculate the value of the phase mismatch and the expected device performance when phase matching is achieved. We use a first principles model for the nonlinear dynamics in the Josephson junction transmission line\[143, 144\] which has been validated by experiments. By making the ansatz that the solutions are traveling waves, taking the slowly varying envelope approximation, and neglecting pump depletion, we obtain a set of coupled wave equations which describe the energy exchange between the pump, signal, and idler in the undepleted pump approximation:

\[
\frac{\partial a_s}{\partial x} - i\kappa_s a_s^* e^{i(\Delta k L + 2\alpha_p - \alpha_s - \alpha_i)} = 0
\]

(6.37)

\[
\frac{\partial a_i}{\partial x} - i\kappa_i a_i^* e^{i(\Delta k L + 2\alpha_p - \alpha_s - \alpha_i)} = 0
\]

(6.38)
where $a_s$ and $a_i$ are the signal and idler amplitudes, $\Delta k_L = 2k_p - k_s - k_i$ is the phase mismatch in the low pump power limit, and the coupling factors $\alpha_p$, $\alpha_s$, and $\alpha_i$ represent the change in the wave vector of the pump, signal, and idler due to self and cross phase modulation induced by the pump. The coupling factors depend on the circuit parameters and scale quadratically with the pump current. Maximum parametric gain is achieved when the exponential terms are constant: the phase mismatch, $\Delta k = \Delta k_L + 2\alpha_p - \alpha_s - \alpha_i$, must then be zero. The coupled wave equations (6.37), (6.38) are similar to the coupled amplitude equations for an optical parametric amplifier[5] and have the solution:

$$a_s(x) = a_s(0) \left( \cosh gx - \frac{i\Delta k}{2g} \sinh gx \right) e^{i\Delta k x / 2}$$  (6.39)

with the gain coefficient $g = \sqrt{\kappa_s \kappa_i^2 - (\Delta k/2)^2}$. For zero initial idler amplitude and perfect phase matching, this leads to exponential gain, $a_s(x) \approx a_s(0) e^{gx/2}$. For poor phase matching $g$ is imaginary and the gain scales quadratically with length rather than exponentially.

Without resonant phase matching, the parametric amplification is phase matched at zero pump power, but rapidly loses phase matching as the pump power increases. Neglecting dispersion and frequency dependent impedances, the exact expression for the phase mismatch can be simplified to yield $\Delta k \approx 2k_p - k_s - k_i - 2k_p \kappa$, where $\kappa = \frac{\alpha^2 k_p^2 |Z_{char}|^2}{16L^2 \omega_p^2 (I_p/I_0)^2}$. The nonlinear process creates a pump power dependent phase mismatch which can be compensated by increasing the pump wave vector.

### 6.4.1 Gain and bandwidth

In Fig. 6.2, we show the increase in gain due to resonant phase matching for the device described in Fig 7.1. Resonant phase matching increases the gain by more than one order of magnitude from 10 dB to 21 dB (Fig. 6.2a) for a pump current of half the junction critical current and a pump frequency, 5.97 GHz, on the lower frequency tail of the resonance as shown in the inset of Fig. 7.1c. The increase in the pump wave vector due to the resonance compensates the phase mismatch from cross and self phase modulation (Fig. 6.2b, black dashed) leading to perfect phase matching near the pump frequency (Fig. 6.2b, purple). For higher pump currents, the benefits are even more pronounced: the RPM TWPA achieves 50 dB of gain (compared to 15 dB for the TWPA) with a pump current of 0.7$I_0$ (Fig. 6.2c). Achieving 50 dB of gain over a 3 GHz bandwidth would require a larger junction critical current than used here to prevent gain saturation by vacuum photons. By varying the pump frequency relative to the resonance, the parametric amplification can be phase matched for arbitrary pump currents (Fig. 6.2d). Due to this ability to tune the pump phase mismatch over the entire range of possible wavevectors, this technique is highly flexible.

### 6.4.2 Scaling relations

We now examine the scaling relations for the gain in order to obtain the optimum gain through engineering the linear and nonlinear properties of the transmission line. Simplifying the expression for the gain by assuming perfect phase matching and neglecting the effects of the resonant element and the junction resonance on the dispersion we find that the
Figure 6.2: Gain of the resonantly phase matched traveling wave parametric amplifier (RPM TWPA). (a) The gain as a function of signal frequency in dB with RPM (purple) and without (black dashed) for a pump current of $0.5I_0$ and a pump frequency of 5.97 GHz. (b) The phase mismatch with (purple) and without (black dashed) RPM. (c) The peak gain as a function of pump current without RPM (black dashed) and with RPM for three different pump frequencies, which phase match the parametric amplification for pump currents of $0.3I_0$ (red), $0.5I_0$ (purple), and $0.7I_0$ (green). (d) The phase mismatch as a function of pump current. The dots mark the pump current where the parametric amplification is perfectly phase matched.
Figure 6.3: Effect of pump depletion on dynamic range. (a) The gain as a function of input signal current (normalized to the pump current) for a small signal gain of 10, 15, and 20 dB obtained with a pump current of $0.5I_0$ and device lengths of 1150, 1530, and 1900 unit cells. The approximation for the gain depletion (dashed lines) from Eq. 6.40 is in excellent agreement with the result obtained by solving the full nonlinear dynamics (solid lines). (b) $P_{1dB}$, the input signal power where the gain decreases by 1 dB, as a function of junction critical current $I_0$ with the pump current fixed at $0.5I_0$. The black dashed line corresponds to the device considered in this article which has a gain compression point of $P_{1dB} = -87,-93$, and -98 dBm for a gain of 10, 15, and 20 dB for a junction critical current of $I_0 = 3.29$ µA.

The exponential gain coefficient is directly proportional to the wave vector $g \propto k_p I_p^2/I_0^2$. Thus, for a fixed pump strength relative to the junction critical current, the gain coefficient is proportional to the electrical length. In other words, a larger wave vector and thus slower light leads to a larger effective nonlinearity due to the higher energy density; this effect is well known in photonic crystals[123]. For convenient integration with commercial electronics the characteristic impedance is designed to be $Z_{char} \approx \sqrt{L/(C + C_c)} \approx 50\Omega$ which fixes the ratio of the inductance and capacitance. The wave vector is proportional to the product of the capacitance and inductance $k \approx \omega/a\sqrt{L(C + C_c)}$. Increasing both the capacitance and inductance or decreasing the unit cell size are effective strategies for increasing the gain per unit length while maintaining impedance matching for a 50 ohm load. The current design represents a trade-off between unit cell size and component values which is convenient to fabricate.

### 6.4.3 Dynamic range

Next we consider the dynamic range of the amplifier. The upper limit of the dynamic range is due to pump depletion: the pump transfers energy to the signal and idler which reduces the parametric gain. To investigate this regime, we solve for the coupled wave equations without the undepleted pump approximation, resulting in four coupled nonlinear differential equations which are solved by transforming them to real differential equations and expressing them as a Jacobi elliptic integral[27]. The gain as a function of input signal power calculated including pump depletion (solid lines in Fig. 6.3) is in excellent agreement with the approximate yet general solution for pump depletion (dashed lines in Fig. 6.3) in
a four photon parametric amplifier [63]:

\[ G = \frac{G_0}{1 + 2G_0I_s^2/I_p^2} \]  

(6.40)

where \( G_0 \) is the small signal gain in linear units and \( I_s \) and \( I_p \) are the input signal and pump currents. From Eq. 6.40, the gain compression point is approximately \( P_{1dB} = P_p/(2G_0) \). Thus, the threshold for gain saturation is independent of the specific device configuration and depends only on the small signal gain and the pump power. The gain as a function of input signal current is plotted for three values of the small signal gain in Fig. 6.3a. The signal current at which the gain drops by 1 dB is marked on the curves of Fig. 6.3a. With a pump current of 0.5\( I_0 \) the signal power where the gain decreases by 1 dB is \(-87, -93, \) and \(-98 \) dBm for a small signal gain of 10, 15, and 20 dB, respectively. These gain compression points are consistent with the approximate relation with the pump power of -69 dBm. The dynamic range of the TWPA is significantly higher than a cavity based Josephson parametric amplifier with the same junction critical current since the lack of a cavity enables a higher pump current before the Josephson junction is saturated.

To further increase the threshold for gain saturation, the junction critical current can be scaled up, as seen in Fig. 6.3b. However, increasing the junction critical current decreases the inductance which reduces the wave vector; leading to a weaker nonlinearity. One potential solution is to use N Josephson junctions each with a critical current \( I_0N \) in series as a superinductor [80]. The gain compression point then scales as \( N^2 \) leading to a larger dynamic range at the expense of more complex fabrication.

6.4.4 Advantages over periodic loading

Resonant phase matching has an important advantage over dispersion engineering through periodic loading which has been used to phase match TWPAs based on the weaker nonlinear kinetic inductance [54, 13]. A disadvantage of periodic loading is the potential phase matching of backward parametric amplification. Dispersion engineering through periodic loading opens a photonic band gap (Fig. 6.4) near the pump frequency and through band bending changes the pump wave propagation constant to phase match forward parametric amplification. Periodic loading creates an effective momentum inversely proportional to the periodicity of the loading \( G = 2\pi/\Lambda \) where \( \Lambda \) is the periodicity of the loading and \( G \) is the reciprocal lattice vector. The periodicity is chosen so that the stop band is at \( G/2 \approx k_p \). In such a periodic system the phase matching relation needs only to be satisfied up to an integer multiple of the reciprocal lattice vector [12]. As can be seen from the phase matching relation, the effective momentum from the lattice phase-matches the parametric amplification process for a backward propagating signal \( \Delta k_{L,b} = 2k_p + k_s + k_i + nG \approx 0 \) for \( n = -2 \). Under this condition, any backward propagating photons present in the system will be amplified, leading to gain ripples and a reduced threshold for parametric oscillations. Due to imperfect impedance matching operating the band of the amplifier, a weak standing wave condition will be set up in the nonlinear transmission line due to the return loss at the output and input. If the return loss in dB is \( R \), then the magnitude of the standing signal will be of order \( 2R \). However, the signal experiences some gain (in dB) in the forward and reverse directions, \( G_f \) and \( G_r \). If \( G_f + G_r + 2R \) approaches unity, the
device becomes a parametric oscillator. The proposed resonant phase matching technique phase matches only the forward parametric amplification process, so the maximum gain before the onset of parametric oscillations may be higher than in a device utilizing periodic loading.

6.5 Conclusions

In conclusion, we have proposed a traveling wave parametric amplifier which is phase matched by sub-wavelength resonant elements in order to achieve 20 dB of gain, 3 GHz of bandwidth, and a saturation power ($P_{1dB}$) of -98 dBm. Experiments based on this scheme have achieved these performance parameters[76]. This device will be well suited to multiplexed readout of quantum bits and astronomical detectors. Applying metamaterial design techniques to nonlinear superconducting systems may yield a number of useful devices for circuit quantum electrodynamics such as backward parametric amplifiers or mirror-less optical parametric oscillators[99].
Chapter 7

Strong field dynamics and quantum noise of Josephson traveling wave parametric amplifiers (JTWPAs)

Josephson traveling wave parametric amplifiers (JTWPAs) have demonstrated high gain over a broad bandwidth with near quantum limited noise performance\cite{76} once phase matching is achieved\cite{96}. As JTWPAs promise to become workhorse amplifiers for single photon level microwave measurements, it is important to understand the non-idealties in the amplifier behavior. In particular, the rapid drop in gain for pump currents near the junction critical current and the non-unity intrinsic quantum efficiency observed in experiments\cite{76} should be fully understood and if possible mitigated. Existing theoretical treatments of the JTWA based on a perturbative analysis of the first nonlinear order\cite{143, 144, 96, 26} have proven highly useful in designing traveling wave parametric amplifiers; however, to understand the non-idealities one must consider the strong field dynamics without assuming a weak nonlinearity or a slowly varying envelope.

In this chapter we study the nonlinear wave equation for the Josephson traveling wave parametric amplifier for a strong pump. We find the traveling wave solution and propose a snoidal drive scheme to potentially increase the blow-up threshold for large pump powers. Furthermore, we investigate the generation of higher order signal and idler sidebands as a source of quantum noise in this device.

The JTWA is described by the nonlinear wave equation derived in the previous chapter:
\begin{equation}
\frac{aE_J}{\phi_0^2} \cos \left( \frac{a}{\phi_0} \phi_x \right) \phi_{xx} - \frac{C}{a} \phi_{tt} + C_J a \phi_{xxtt} = 0
\end{equation}
where $C_J$ is the junction capacitance, $E_J = \phi_0 I_0$ is the Josephson energy, $I_0$ is the junction critical current, $C$ is the capacitance to ground, $a$ is the unit cell size, and $\phi_0 = \hbar/(2e)$ is the reduced flux quantum.

We then define a normalized time $\tilde{t} = t/\sqrt{L_J C}$, distance $\tilde{x} = x/a$, and magnetic flux $\tilde{\phi} = \phi/\phi_0$ where $L_J = \phi_0/I_0$ is the Josephson inductance. Dropping the tildes, we obtain the exact nonlinear wave equation with a single parameter $\beta = C_J/C$ characterizing...
Figure 7.1: (a) Circuit diagram of the Josephson traveling wave parametric amplifier (JTWPA). (b) Wavevector as a function of frequency, scaled by $\sqrt{\beta}$. At low frequencies the dispersion is linear with increasing curvature as one nears the plasma frequency at $\omega = 1/\sqrt{\beta}$. The location of an experiment on this curve determines the pump dynamics. In the RPM theory paper (Ref. 2) $\omega \sqrt{\beta} \approx 0.2$ and in the experiment (Ref. 1) $\omega \sqrt{\beta} \approx 0.09$. (c) Wavevector vs pump amplitude. Due to the Josephson nonlinearity, the wavevector increases for a strong pump. At a certain pump amplitude ($A \approx 20$ for $\omega = 0.08$ and $\beta = 1.205$) the wave-vector becomes complex and waves decay exponentially along the transmission line. The current passing through the Josephson junction reaches the critical current at $A \cdot k = \pi/2$ then decreases with increasing amplitude and wavevector.
the dispersion due to the junction resonance:

$$\cos(\phi_x)\phi_{xx} - \phi_{tt} + \beta\phi_{xxtt} = 0$$  \hspace{1cm} (7.2)$$

One important feature of this equation is the single parameter, $\beta$, that governs the dynamics in the JTWPA. In the resonant phase matching theory paper[96] we considered $\beta = 6.7$ (a $2\pi \cdot 28$ GHz plasma frequency), while in the experimental demonstration[76] we decreased the junction capacitance and decreased $\beta$ to $1.205$ ($2\pi \cdot 80$ GHz plasma frequency) in order to increase the bandwidth. In both cases, a strong microwave tone near 7 GHz was used to pump the amplifier. In normalized units, 7 GHz is $\omega = 0.08$.

For a small amplitude wave $|\phi_x| \ll 1$, the wave equation is approximately linear and a sinusoidal wave of the form $\phi(x,t) = A \cos(kx - \omega t)$ propagates with a dispersion relation:

$$\omega^2 - k^2 + \beta k^2 \omega^2 = 0$$  \hspace{1cm} (7.3)$$

which has the junction plasma frequency at $\omega = 1/\sqrt{\beta}$ as seen in Fig. 7.1b. For large amplitude waves, neglecting higher harmonics, we obtain the dispersion relation (see Appendix 2 for derivation):

$$\omega^2 - k^2 \left[ J_0(\bar{A}k) + J_2(\bar{A}k) \right] + \beta k^2 \omega^2 = 0$$  \hspace{1cm} (7.4)$$

The zeros of the dispersion relation give the wavevector as a function of pump amplitude, frequency, and dispersion ($\beta$).

We now solve the nonlinear wave equation (Eq. 7.2) for a sinusoidal pump. We set the initial conditions to generate a sinusoidal wave $\phi(x,t) = A \cos(kx - \omega t)$ using the wavevector from Eq. 7.4. We use the finite element method to solve for the pump flux as a function of time in a wavelength sized section of the nonlinear transmission line using periodic boundary conditions: $\phi(0,t) = \phi(2\pi/k,t)$.

The features we observe are the generation of higher harmonics for moderate pump amplitudes and a blow-up of the waveform at a certain pump amplitude (Fig. 7.2) which depends weakly on the dispersion (Fig. 7.3). An analytic proof of the sufficient conditions for blow-up may be possible using the methods of Ref. [132].

### 7.1 Traveling wave solution

To find an analytic solution for wave propagation including higher harmonics, we look for traveling wave solutions of the nonlinear wave equation (Eq. 7.2). We first transform to a frame moving at velocity $v$ by substituting $z = x - vt$ into the nonlinear wave equation, where $v$ is the wave velocity. By integrating the expression and defining a new variable $f = \phi'$, we obtain the ODE which traveling wave solutions must satisfy:

$$\sin(f) - v^2 f + \beta v^2 f'' = 0$$  \hspace{1cm} (7.5)$$

where we set the integration constant equal to zero as we are looking for solutions with zero average flux. The flux is obtained by integrating $f$.

From the phase space diagram, Fig. 7.4a, the JTWPA has closed orbits for small initial amplitudes with saddle points at $v^2 f = \sin f$, for $|v| < 1$. We consider the case...
Figure 7.2: Solution of the nonlinear wave equation in a wavelength sized unit cell in the time domain for the initial conditions of a sinusoidal wave. (a) Propagation and distortion due to harmonic generation (b) Blow-up occurs for this amplitude after the wave propagates \( \approx 200 \) unit cells (c). Amplitude of the first, third, and fifth pump harmonics as a function of pump amplitude and time.

Figure 7.3: (a)-(d) Solution of the nonlinear wave equation for a sinusoidal wave as a function of pump amplitude and time for different amounts of dispersion. (e)-(h) Values of the nonlinear term in the wave equation, \( \cos \phi_x \), as a function of pump amplitude and time for \( \beta=1.205, 2, 6, \) and 10.
\[ \phi = f, \quad \phi' = f'' \]

\[ \kappa = \phi - \phi' \]

| \( \phi' / \phi'' \) is constant along the trajectory. However, as the amplitude increases, \( \phi' / \phi'' \) varies with time (red, blue, and black curves in Fig. 7.4a,b,c), which is due to the traveling wave solution containing higher harmonics. For initial amplitudes larger than the critical point, no traveling wave solutions exist and the wave-form will blow up.

The phase space diagram of the JTWPA is similar to the phase space diagram of the sine-Gordon equation, which governs fluxon dynamics in a long Josephson junction or a series array of Josephson junctions\[60, 117, 83, 43\]; however, for the JTWPA, only a single well exists.

Now we look for an analytic solution to the traveling wave equation. First we consider the approximate PDE. We Taylor expand the cosine term to the first nonlinear order:

\[ \phi_{xx} - \phi_{tt} - \frac{1}{2} \phi_{x}^{2} \phi_{xx} + \beta \phi_{xxx} = 0 \] (7.6)

Applying the Jacobi elliptic function expansion method\[71, 146\], we find traveling wave solutions in the form of snoidal waves:

\[ f(z) = B \text{sn}(k_0 z, m) \] (7.7)

where \( k_0 \) is the wave-vector only in the weak pump limit and \( m \) is the parameter, a real number between zero and one. For small values of the parameter, the Jacobi elliptic function approaches a sine function. Integrating \( f(z) \) yields the flux:

\[ \phi(x, t) = \frac{B \log [\text{nd}(k_0 (x - vt), m) + \sqrt{m} \text{sd}(k_0 (x - vt), m)]}{k \sqrt{m}} \] (7.8)
Figure 7.5: Solution of the nonlinear wave equation in a wavelength sized unit cell in the time domain for the initial conditions of a snoidal wave. (a) Propagation without distortion due to the fixed amplitude and phase relationship between the pump harmonics (b) Amplitude of the first, third, and fifth pump harmonics as a function of pump amplitude and time. As a note, for a fair comparison with a sinusoidal drive, the pump amplitude on the y axis of panel (b) is that of the first harmonic of the traveling wave (at ω). The peak amplitude of the snoidal wave will be larger due to the constructive interference of the fundamental and higher harmonic components.

We substitute this solution into Eq. 7.6 and obtain the dispersion relations from the coefficients of each power of the Jacobi elliptic functions:

\[
v = \sqrt{\frac{1 - \frac{B^2}{12}}{1 + k_0^2 \beta}} \tag{7.9}
\]

\[
m = \frac{B^2}{12 \beta k_0^2} \frac{1 + k_0^2 \beta}{1 - \frac{B^2}{12}} = \frac{B^2}{12 \beta k_0^2 v^2} \tag{7.10}
\]

The frequency is defined through the periodicity of the Jacobi elliptic functions, \(\text{sn}(k_0 z, m) = \text{sn}(k_0 z + 4K[m], m)\), where \(K[m]\) is the complete elliptic integral of the first kind:

\[
2\pi k_0 v / \omega = 4K[m] \tag{7.11}
\]

Numerical solutions of Eqs. 7.9, 7.10, and 7.11 yield the parameters specifying the traveling wave, \(v(B, \omega, \beta), m(B, \omega, \beta)\) and \(k_0(B, \omega, \beta)\), as a function of frequency and amplitude. The wave-vector for a strong pump is \(k(B, \omega, \beta) = \pi k_0/(2K[m])\) rather than \(k_0\) due to the dependence of the period of the Jacobi elliptic function on the parameter \(m\).

Now we consider whether the method above can solve the exact nonlinear wave equation. We substitute the snoidal wave ansatz into the exact nonlinear wave equations. We then obtain two equations which approximately define the velocity and parameter of the Jacobi elliptic function. We obtain the first by setting \(t = 0, x = 4K[m]/k_0\) and the
second by taking the time derivative and setting $t = x = 0$:

$$
v = \sqrt{\frac{(B + \sin B)/(2B)}{1 + k_0^2 \beta}}
\tag{7.12}
$$

$$
m = \frac{B - \sin B}{2k_0^2 \beta v^2 B}
\tag{7.13}
$$

We then numerically solve the exact nonlinear wave equation using the snoidal wave as the initial condition. We find that the snoidal wave solution with the approximate coefficients does generate a traveling wave solution to the exact nonlinear wave equation, as shown in Fig. 7.5. The snoidal wave solution enables a higher pump amplitude before blow-up occurs, as shown in Fig. 7.6.

We now consider the feasibility of generating snoidal waves in an experiment. At the GHz frequencies of typical circuit QED experiments, arbitrary waveform generation is challenging. The simplest method is to combine several harmonics of a sinusoidal pump with the proper phases and amplitudes. Performing a Fourier expansion of the flux (Eq. 7.8), we obtain the traveling wave drive as a combination of harmonic drives:

$$
\phi(x, t) = \frac{-4B}{k_0 \sqrt{m}} \sum_{n=0}^{\infty} \frac{1}{(2n + 1)} \frac{q^{n+1/2}}{1 - q^{2n+1}}
\times \cos \left[ \frac{\pi(2n + 1)}{2K[m]} k_0(x - vt) \right]
\tag{7.14}
$$

where $q = \exp (-\pi K [1 - m]/K[m])$. The number of harmonics needed to approximate the solution depends on the pump strength. For weak pumps the parameter $m$ approaches zero and the solution is approximately sinusoidal. As the pump strength increases, the parameter $m$ approaches one and the number of harmonics required increases. For example, in Fig. 7.7, we plot the traveling wave solution for a pump power near breakdown ($m = 0.999950$) along with harmonic approximations containing the first harmonic through the third, first
Figure 7.7: Feasibility of traveling wave pump. (a) The traveling wave solution (black dashed) and the harmonic approximations consisting of the first and up to the third harmonic (blue), fifth (green), and seventh (red) harmonics. (b) Differences between the traveling wave solution and the harmonic approximations through fifth, and first through seventh pump harmonics. Including only the first and third pump harmonics, the traveling wave drive can be approximated with an error less than five percent.

7.2 Multi-mode parametric amplification

To solve for the parametric amplification process, we linearize the equations of motion about the pump solution. We substitute in \( \phi = \phi + \epsilon \psi \) into Eq. 7.6, assuming \( \phi \) is a solution to the pde, and keep terms of order \( \epsilon \):

\[
\partial_x [\cos (\phi_x) \psi_x] - \psi_{tt} + \beta \psi_{xxtt} = 0 \tag{7.15}
\]

Fourier transforming the above equation, we obtain:

\[
\partial_x [F[\cos (\phi_x)] \ast \psi_x] + \omega^2 \psi - \beta \omega^2 \psi_{xx} = 0 \tag{7.16}
\]

We can then express the cosine of the derivative of the pump flux as a series of harmonics:

\[
F[\cos (\phi_x)] = \sum_{m=-\infty}^{\infty} A_m(x)e^{i2mk_p x} \delta(\omega - 2m\omega_p) \tag{7.17}
\]

Inserting this expression into the Fourier domain wave equation and performing the convolution, we obtain:

\[
\partial_x \left[ \sum_{m=-\infty}^{\infty} A_m(x)e^{i2mk_p x} \psi_x(\omega - 2m\omega_p) \right] - \beta \omega^2 \psi_{xx}(\omega) + \omega^2 \psi(\omega) = 0
\]

To solve the above equations, we consider \( N + 1 \) modes and write this in matrix form where \( \mathbf{u} = [\psi_0, \cdots, \psi_N, \psi'_0, \cdots, \psi'_N]^T \):

\[
\begin{bmatrix}
\mathbf{\Omega} & -\beta \omega^2
\end{bmatrix}
\begin{bmatrix}
\mathbf{\Omega}'
\end{bmatrix} = \begin{bmatrix}
\mathbf{0}
\end{bmatrix}
\]

\[
\begin{bmatrix}
\mathbf{\Omega}'
\mathbf{0}
\end{bmatrix}
\begin{bmatrix}
\mathbf{\Omega} - \beta \omega^2
\end{bmatrix}
\begin{bmatrix}
\mathbf{u}'
\end{bmatrix} = \begin{bmatrix}
\mathbf{0}
\end{bmatrix}
\]

\[
\begin{bmatrix}
\mathbf{I}
0
\end{bmatrix}
\begin{bmatrix}
\mathbf{u}'
\end{bmatrix} = \begin{bmatrix}
\mathbf{0}
\end{bmatrix}
\]

\[
\begin{bmatrix}
\mathbf{0}
\mathbf{I}
\end{bmatrix}
\begin{bmatrix}
\mathbf{u}
\end{bmatrix} = \begin{bmatrix}
\mathbf{0}
\end{bmatrix}
\]

\[
\begin{bmatrix}
\mathbf{u}'
\end{bmatrix} = \begin{bmatrix}
\mathbf{0}
\end{bmatrix}
\]

\[
\begin{bmatrix}
\mathbf{u}
\end{bmatrix} = \begin{bmatrix}
\mathbf{0}
\end{bmatrix}
\]
where

\[ \omega = \begin{bmatrix} \omega_0 & 0 & 0 \\ 0 & \ddots & 0 \\ 0 & 0 & \omega_N \end{bmatrix} \quad \Omega = \begin{bmatrix} \Omega_0 & \cdots & \Omega_N \\ \vdots & \ddots & \vdots \\ \Omega_{-N} & \cdots & \Omega_0 \end{bmatrix} \] (7.20)

and \( \Omega_m = A_m(x)e^{2imk_p x} \). We use the boundary conditions \( \psi(\omega_s) = a_s \) and \( \psi_x(\omega_s) = a_{s,0} \omega_s/\sqrt{1 - \beta \omega_s^2 - \Omega_0(0)} \). For the idler, we set the initial values of the derivative of the flux, by setting the flux to zero and integrating. The initial values of modes not directly coupled to the signal mode can be safely set to zero with negligible error. The Toeplitz matrix \( \Omega_n \), resulting from the convolution in Eq. 7.17, couples the pump and the signal, idler, and sidebands. As an example, a sinusoidal pump has \( A_0 = a_p^2/2 \), and \( A_{\pm 1} = -a_p^2/4 \). The \( A_0 \) term leads to self phase modulation while \( A_{\pm 1} \) induces nearest neighbor coupling between the modes at \( \omega_n \) and \( \omega_n \pm 2\omega_p \) as seen in Fig. 7.8. Higher pump harmonics couple modes with larger frequency separations, and can increase or decrease the gain, depending on the relative phase of the higher harmonics with the pump.

We note that the slowly varying envelope approximation (SVEA) can be used to reduce the order of the ODE as in[26]; however, in a JTWPA the rapid growth of the signal and idler can invalidate the condition required to neglect the higher order derivatives in the SVEA, \( |k(\omega)\psi(\omega)/\psi_x(\omega)| \gg 1 \). In a typical JTWPA with a gain of \( \approx 30 \) dB the slowly varying approximation is not well justified with \( |k(\omega)\psi(\omega)/\psi_x(\omega)| \approx 15 \) leading to errors in the gain calculation of several dB.

A plot of the power in the signal, idler, and higher order sidebands is shown in Figure 7.8. One observes that the power in the sidebands is only several dB lower than the power in the signal and idler. This implies the sidebands could be a source of noise, which we will investigate in the next section.

### 7.3 Quantum noise

The quantum noise properties of JTWPA s are critically important for their role as low noise amplifiers for circuit QED, opto-mechanics, and dark matter searches as well as their potential as bright sources of non classical photon states. Here we derive the noise figure for the signal, idler, and sidebands and find that coupling to higher order sidebands imposes an upper limit for the quantum efficiency of traveling wave parametric amplifiers. Importantly, this quantum efficiency can be made near unity by engineering the phase mismatch of the pump and the dispersion of the transmission line (\( \beta \)).

Transfer of energy between the signal and idler and their higher order sidebands will lead to a drop in the quantum efficiency of the device because the original signal will be mixed with the vacuum fluctuations at all of the sideband frequencies. In addition, the information contained in the signal will be conveyed into the higher order sidebands, which are typically not observed, leading to a drop in the quantum efficiency.

As shown in Ref. [82], the noise figure for parametric amplifier with an arbitrary number of modes in the limit where the mean photon number of the coherent state is larger
than the variance is given by:

$$F_j \approx 1 + \sum_{k \neq j} \frac{|\mu_{jk}|^2}{|\mu_{ji}|^2}$$  \hspace{1cm} (7.21)

where $\mu_{jk}$ are elements of the classical transfer matrix. The signal noise-figure $F_j$ is defined as the ratio of the input and output signal to noise ratios for a given mode.

An ideal parametric amplifier has unity quantum efficiency. In the experiments for the JTWPA manuscript, we observed an intrinsic quantum efficiency of 0.85 after neglecting the known noise contributions such as insertion loss, distributed loss, and following amplifier noise. For the dispersion used in the TWPA experiments ($\beta = 1.205$), theory predicts an intrinsic quantum efficiency of 0.80 at 20 dB gain which is comparable to the measured value of 0.85. The small discrepancy between the measured and predicted values may result from not including the effects of attenuation on the signal, idler, and their respective sidebands. As seen in Figure 7.9, increasing the dispersion (by increasing the junction capacitance) to $\beta = 5$, will increase the predicted intrinsic quantum efficiency to approximately 97 percent.

In conclusion, we have derived a traveling wave solution for the Josephson traveling wave parametric amplifier which will propagate without distortion. In addition, we find that the higher order signal and idler sidebands present a potential quantum noise source, which can be mitigated by engineering the dispersion relation.
Figure 7.9: (a). gain as a function of pump wave-vector and pump amplitude (b). noise figure as a function of pump wave-vector and pump amplitude. (c) noise figure as a function of gain for several values of the dispersion. (d) quantum efficiency as a function of gain for several values of the dispersion.
Chapter 8

Conclusion

In this chapter, we will summarize the results presented in this thesis and discuss potentially promising future directions. In this thesis, we have explored nonlinear light matter interactions in metamaterials, systems in which the electromagnetic properties have been tailored on a sub-wavelength scale. Nonlinear metamaterials is a young but growing field, where advances will impact diverse fields such as medical diagnostics, telecommunications, and quantum computing.

8.1 Nonlinear optics in zero and negative index metamaterials

In Chapter 5, we presented the first experimental results on nonlinear propagation in a bulk zero or negative index metamaterial. There are a number of interesting future directions and predictions for nonlinear optics in zero and negative index materials yet to be experimentally tested such as novel behavior for parametric amplification and entangled photon generation[100, 98, 99]. In our experiments, the optical loss due to the metal of the fishnet metamaterial presented a challenge for demonstrating parametric amplification and entangled photon generation. All dielectric zero-index metamaterials[90, 68] have significantly reduced optical attenuation and may be promising platforms for further exploring the nonlinear properties of zero index metamaterials. As metamaterial fabrication techniques and designs continue to improve, we expect nonlinear optics in zero and negative index metamaterials to advance.

8.2 Josephson traveling wave parametric amplifier

In Chapter 6, we detailed a phase matching scheme for Josephson Traveling Wave Parametric Amplifiers (JTWPAs) called “resonant phase matching” and in Chapter 7 we further explore the nonlinear behavior beyond the perturbative regime. Although amplifiers using this phase matching scheme[96] are the state of the art amplifier for single-photon level microwave measurements[76]: there are many routes to further improving the performance. Junction arrays can be used to increase the dynamic range by maintaining the linear
inductance of a single junction yet allowing a much larger drive current which enables an increased dynamic range, as demonstrated in Josephson parametric amplifiers\cite{42, 38}. The results in Chapter 7 imply that the quantum efficiency drop due to generating higher order signal and idler modes can be mitigated by increasing the dispersion. Increasing the dispersion by decreasing the junction plasma frequency should increase the quantum efficiency at the expense of bandwidth; however, it should be studied whether this trade-off is fundamental or whether it can be avoided by more complicated engineering of the dispersion relation.
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