Review of Nonlinear Optics in Metamaterials

Yuanjiang Xiang, Xiaoyu Dai, Shuangchun Wen, and Dianyuan Fan
Research Center of Laser Science and Engineering and School of Computer and Communication
Hunan University, Changsha 410082, China

Abstract— Recent researches on metamaterials (MMs) demonstrated that a nonlinear MM exhibits a rich spatiotemporal dynamics where both linear and nonlinear effective properties can be tailored simply. This provides opportunity to explore the new nonlinear optical phenomena in MMs. This contribution reviews the recent advances on nonlinear interaction between electromagnetic wave with MMs, from the fundamental physical models to unique phenomena and conceived novel devices. The physical origins for the typical nonlinear phenomena, such as second-order harmonic generation (SHG), optical parametric amplification (OPA), four-wave mixing (FWM), optical soliton, self-phase modulation (SPM), and self-focusing in MMs are explained. Several proposals for applications of nonlinear MMs in manipulating light are demonstrated.

1. INTRODUCTION

Metamaterials (MMs) are artificial structures that can be pre-designed to show specific electromagnetic properties not commonly found in nature [1–6]. MMs include positive-index regime, absorption regime and negative-index regime [3, 4]. One of the most exciting opportunities for MMs is the development of negative-index metamaterials (NIMs) [1]. NIMs have a number of peculiar properties: reversed Snell refraction, reversed Doppler Effect, reversed radiation tension, negative Cerenkov radiation [1], reversed Goos-Hänchen shift [2] and so on. NIMs have opened new doors in optics and have truly excited the imagination of researchers worldwide. Most of the properties of NIMs have been studied only for linear waves, such as negative refraction and superlensing [7]. However, it has been already noticed that the NIMs may possess quite complicated nonlinear response. Thanks to rapidly developing nanofabrication and sub-wavelength imaging techniques, the optical NIMs can now be fabricated. The possibility of nonlinear electromagnetic responses, including cubic or quadratic nonlinear responses, in MMs is also demonstrated by the inclusion of nonlinear elements within the MMs, for instance by embedding the split-ring resonators (SRRs) in a Kerr-type dielectric [8], or by inserting certain nonlinear elements (e.g., diodes) in the split-ring resonators' paths [9]. The accessibility of linear and nonlinear MMs exhibiting negative electric and magnetic properties in the infrared and optical frequencies [10–12] have truly excited the imagination of researchers worldwide and stimulated intense investigation on the nonlinear optics of MMs, including second-order nonlinear optical phenomena such as second-harmonic generation and optical parametric amplification [12–18], as well as the 3rd-order nonlinear interaction of ultrashort electromagnetic pulse with MMs [13, 19–24]. It is known that, optical magnetization, which is normally ignored in linear and nonlinear optics of the ordinary media, plays a crucial role in MMs. Several authors have shown that it is the dispersive magnetic permeability that significantly leads to the difference between the propagation models for ultrashort pulses in MMs and in ordinary media. For propagation of ultrashort pulses in MMs with a nonlinear electric polarization, it is demonstrated that the linear dispersive magnetic permeability is incorporated into the nonlinear polarization, resulting in a controllable self-steepening (SS) effect and a series higher-order dispersive nonlinear terms in the propagation models [20–23]. The role of the controllable SS effect in MI has been identified [20, 21].

As the wavelength for which we observe a negative refractive index in MMs continues to push into the optical regime, the study of nonlinear effects will become very important, particularly from an applications point of view. In this paper, we report on the recent progress on investigation of some typical nonlinear optical phenomena in MMs, including third-order nonlinear optical phenomena such as soliton propagation, self-focusing, and spatiotemporal instability. The controllability and the novel properties of the nonlinear phenomena in MMs are demonstrated. Here we consider nonlinear optical processes in a MMs and show that they also exhibit unusual properties with respect to energy conversion and propagation. This will further extends the conventional area of optics, particularly nonlinear optics, and leads to completely new electronic and optical devices to manipulate light waves.

2. THE SECOND-ORDER NONLINEAR OPTICAL PHENOMENA IN MMS

In 1961, Franken and his collaborators experimentally discovered second-harmonic generation (SHG) [25], since then, SHG has become one of the most investigated and discussed nonlinear optical processes. Recently, the study of second order processes in MMs has been addressed by Lapine through inserting diodes in the split-ring resonators' paths [9], they have shown that for the case of small amplitudes of the interacting waves, when the diodes are driven by relatively low voltage, the nonlinear response of the MM is described by a quadratic magnetic susceptibility. Subsequently, they analyzed the three-wave coupling processes with a strong pump wave and two weak signals [26]. However, for the quadratic nonlinear, we will more concerns with SHG, optical parametric amplification (OPA), four-wave mixing (FWM).

2.1. Second-order Harmonic Generation in MMs

The first analysis of SHG from a semi-infinite NIM was briefly presented by Agranovich et al. [13], who considered that SHG in transmission is badly phase mismatched. Detailed theoretical studies have been made on second harmonic generation [14–17] where one of the important results was the demonstration of the possibility of exact phase matching when the fundamental and second harmonic waves are counter-propagating. Shadrivov et al. demonstrate that the original paper by Agranovich et al. missed an important additional phase-matching condition, quite specific for the harmonic generation by the backward waves [16]. They demonstrate that exact phase matching between a backward-propagating wave of the fundamental frequency (FF) and the forward propagating wave at the second harmonics (SH) is indeed possible. This novel phase-matched process allows the creation of an effective "quadratic mirror" that reflects the SH component generated by an incident FF wave.

Shalaev et al. have also done a lot of work in the second-order nonlinear optical processes. They investigate not only second-harmonic generation and Manley-Rowe relations but also the parametric amplification in NIMs [14]. In the paper, they propose the possibility of a left-handed nonlinear-optical mirror, which converts the incoming radiation into a reflected beam at the doubled frequency with efficiency that can approach 100% for lossless and phase-matched medium considered.

Moreover, Scalora et al. studied pulsed SHG in MMs under the conditions of significant absorption [27]. By tuning the pump in the negative index range, a second harmonic signal is generated in the positive index region, such that the respective indices of refraction have the same magnitudes but opposite signs. This insures that a forward-propagating pump is exactly phase matched to the backward-propagating second harmonic signal. Using peak intensities of $\sim 500\,\mathrm{MW/cm^2}$, assuming $\chi^{(2)} \sim 80\,\mathrm{pm/V}$, they predicted conversion efficiencies of 12% and 0.2% for attenuation lengths of 50 and $5\,\mu\mathrm{m}$, respectively. Following this paper, Ceglia et al. studied SHG in a NIM cavity [28], the nonlinear process is made efficient by local phase-matching conditions between a forward-propagating pump and a backward-propagating second harmonic signal. By simultaneously exciting the cavity with counterpropagating pulses, and by varying their relative phase different, one is able to enhance or inhibit linear absorption and the second-harmonic conversion efficiency.

The theoretical research has been followed by a recent experimental demonstration of second harmonic generation in a magnetic MM [15, 18]. They have observed SHG from MMs composed of split-ring resonators excited at 1.5-micrometer wavelength and much larger signals are detected when magnetic-dipole resonances are excited, as compared with purely electric-dipole resonances. The experiments are consistent with the calculations based in the magnetic component of the Lorentz force exerted on metal electrons, where an intrinsic SHG mechanism that plays no role in nature materials, but this mechanism becomes relevant in this experiment as a result of the enhancement and the orientation of the local magnetic fields associated with the magnetic-dipole resonances of the split-ring resonances.

Recently, Roppo et al. analyzed pulsed SHG in ordinary and NIMs under phase mismatched conditions [29], and found that a portion of the generated second-harmonic signal is phase locked, trapped and dragged along by the pump pulse. In the case NIMs, it turns out that the trapped pulse and the pulse back-reflected at the interface constitute a set of twin pulses having the same negative wave vector but propagating in opposite directions as a result of the trapping mechanism, this work thus extends previous investigations done in ordinary materials, and bridges the gap with MMs by revealing exciting new dynamical characteristics hitherto unknown.

2.2. Optical Parametric Amplification in MMs

In particularity, Shalaev et al. propose a new approach to compensate losses in NIMs [14]. They demonstrate that the amplification of the left-handed wave can be turned into a cavity-less oscillation when the denominator tends to zero. Further, they have also shown the feasibility of compensating losses in NIMs by OPA. In this process, the wave-vectors of all three coupled waves are co-directed, whereas the energy flow for the signal wave is counter-directed with respect to those for the pump and the idler waves. As seen in Fig. 2, the process is characterized by properties that are in strict contrast with those known for conventional nonlinear-optical crystals. Such extraordinary features allow one to realize optical parametric oscillations (OPOs) without a cavity at frequencies where the refractive index is negative. It is shown that the OPA and OPO in NIMs enable the generation of pairs of entangled counter-propagating right- and left-handed photons inside the NIM slabs.

3. THE THIRD-ORDER NONLINEAR OPTICAL PHENOMENA IN MMS

Owing to their unconventional characteristics, MMs with third-order nonlinearity have attracted a lot of recent interest in the properties of wave propagation in such material, which are most investigated in two aspects, the behavior of waves propagating in the bulk homogeneous MMs or across the interface between the conventional medium and the MMs such as the nonlinear MMs slab or the periodic structure composed of MMs and conventional right handed material.

3.1. The Propagation Properties for Electromagnetic Wave across the Interface of MMs

When the electromagnetic wave propagates through the interface of MMs, some novel phenomena will happen. Such as surface waves, gap soliton and optical bistability. Surface wave propagate along the interface and decay in the transverse direction, which is particularly important for the lensing effect since the amplification of evanescent modes is responsible for the subwavelength resolution. Darmanyan et al. studied the properties of nonlinear TE-polarized surface modes at the interface between different conventional and NIM and between two NIM [30], the constraints for the mode existence are identified and the energy flow associated with the surface modes was calculated.

Moreover, Hegde et al. have studied a periodic structure which is consisting of alternating layers of positive-index and negative-index materials [31]. They found that a novel band gap where the average refractive index is zero. The zero-n gap differs from the usual Bragg gap in which it is invariant to scale length and relatively insensitive to disorder and input angle [32, 33]. In the presence of Kerr nonlinearity, this zero-n gap can switch from low transmission to a perfectly transmitting state, forming a nonlinear resonance or gap soliton in the process. Furthermore, the phenomena such as hysteresis and bistability have been predicted. Optical bistability is a class of optical phenomena in which a system can exhibit two steady transmission states for the same input intensity, which has attracted interesting for the potential application of the all optical switching.

Recently, Kochaert et al. have studied a ring cavity filled with a slab of a right-handed material and s slab of left-handed materials, both layers are assumed to be nonlinear Kerr media [34, 35]. By constructing a mean-field model, they show that the sign of diffraction can be made either positive or negative in this resonator, depending on the thickness of the layers. And they also demonstrated that the dynamical behavior of the modulation instability is strongly affected by the sign of the diffraction coefficient.

3.2. The Propagation Properties for Ultrashort Pulses in MMs

There have some investigations of pulse propagation in MMs, especially in NIMs [17–22, 36]. Another important theoretical contribution has been the derivation of a generalized nonlinear Schr?dinger equation that can be employed to study pulse propagation and solitary waves in negative index media [19]. It has been demonstrated that the nonlinear MM exhibits a rich spatiotemporal dynamics where both linear and nonlinear effective properties can be tailored by simply engineering the MM [20, 21]. It is known that the most important difference between an ordinary medium and a MM is that the former has a constant permeability, while the latter has a dispersive permeability [37]. Therefore, it can be convinced that the most important difference between the propagation of ultrashort pulses in these two kinds of materials should result mainly from the dispersive permeability. To disclose the features of ultrashort pulse propagation resulting from the unique properties of MMs, it is apparent that a careful re-examination of the propagation of ultrashort pulses is needed. In addition, it is believed that deep understanding of the nonlinear interaction of ultrashort pulses with MMs will lead to new devices with previously inconceivable properties. We consider pulse propagation in the latter regime, and thus can assume that the pulse is propagating in uniform, bulk material, in which there are no free charges and in which no free currents flow. In addition, we assume the MM has a nonlinear electric polarization and a nonlinear magnetization. The coupled nonlinear Schrödinger equations (NLSEs) for the envelopes of the electric and magnetic fields will be derived by [22]:

$$\begin{aligned}\frac{\partial E}{\partial \xi} &= i\hat{D}E + \frac{i}{2\beta_0\hat{S}_1}\nabla_\perp^2 E + \frac{i\mu_0\varepsilon_0\chi_E^{(3)}\omega_0^2}{2\beta_0}\frac{\hat{S}_2^2}{\hat{S}_1}\hat{G}\left(|E|^2 E\right) + \frac{i\omega_0\mu_0\chi_H^{(3)}}{2}\hat{S}_2\left(|H|^2 H\right)\\ \frac{\partial H}{\partial \xi} &= i\hat{D}H + \frac{i}{2\beta_0\hat{S}_1}\nabla_\perp^2 H + \frac{i\varepsilon_0\mu_0\chi_H^{(3)}\omega_0^2}{2\beta_0}\frac{\hat{S}_2^2}{\hat{S}_1}\hat{Q}\left(|H|^2 H\right) + \frac{i\omega_0\varepsilon_0\chi_E^{(3)}}{2}\hat{S}_2\left(|E|^2 E\right)\end{aligned} \tag{1}$$

For the purpose of calculations, it is convenient to rewrite Eq. (1) in normalized units. To this end we define the mth-order dispersion length, $L_{dm} = \tau_p^m/\beta_m$, where τ_p is the pulse width $(1 = e)$, the nonlinear polarization length, $L_{Pnl} = 2\beta_0/(\mu_0\varepsilon_0\nu_0\chi_p^{(3)}|E_0|^2\omega_0^2)$, the nonlinear magnetization length, $L_{Mnl} = 2\beta_0/(\varepsilon_0\mu_0\vartheta_0\chi_M^{(3)}|H_0|^2\omega_0^2)$, and a characteristic length, $L_\perp = \sqrt{|L_{d2}/\beta_0|}$, and take the transformations, $U = E/E_0$, $V = H/H_0$, $T = \tau/\tau_p$, $X = x/L_\perp$, $Y = y/L_\perp$, $Z = \xi/|L_{d2}|$, where E_0 and H_0 are the initial amplitudes of E and H. Eq. (1) is thus transformed to the following form:

$$\begin{aligned}\frac{\partial U}{\partial Z} &= i\tilde{D}U + \frac{i\mathrm{sgn}(\beta_0)}{2\tilde{S}_1}\nabla_T^2 U + iN_e\frac{\tilde{S}_2^2}{\tilde{S}_1}\left(1+\tilde{G}\right)\left(|U|^2 U\right) + i\frac{N_E n}{\nu_0\chi}\tilde{S}_2\left(|V|^2 V\right)\\ \frac{\partial V}{\partial Z} &= i\tilde{D}V + \frac{i\mathrm{sgn}(\beta_0)}{2\tilde{S}_1}\nabla_T^2 V + iN_H\frac{\tilde{S}_2^2}{\tilde{S}_1}\left(1+\tilde{Q}\right)\left(|V|^2 V\right) + i\frac{N_h n\chi}{\vartheta_0}\tilde{S}_2\left(|U|^2 U\right)\end{aligned} \tag{2}$$

where $\nabla_T^2 = \partial^2/\partial X^2 + \partial^2/\partial Y^2$, $N_E = |L_{d2}|/L_{Pnl}$, $N_H = |L_{d2}|/L_{Mnl}$, $\chi = \sqrt{\varepsilon_0}\chi_p^{(3)}|E_0|^2 E_0/(\sqrt{\mu_0}\chi_M^{(3)}$ $|H_0|^2 H_0)$, the linear dispersive operators

$$\tilde{D} = \sum_{m=2}^{\infty}\frac{i^m b_m}{m!}\frac{\partial^m}{\partial T^m}, \quad \tilde{G} = \sum_{m=1}^{\infty}\frac{i^m g_m}{m!}\frac{\partial^m}{\partial T^m}, \quad \tilde{Q} = \sum_{m=1}^{\infty}\frac{i^m q_m}{m!}\frac{\partial^m}{\partial T^m}, \tag{3}$$

where $L_{dm} = \tau_p^m/\beta_m$, $b_m = |L_{d2}|/L_{dm}$, $g_m = \nu_m/\left(\nu_0\tau_p^m\right)$, $q_m = \vartheta_m/\left(\vartheta_0\tau_p^m\right)$, and the SS operators

$$\tilde{S}_1 = 1 + ivs\frac{\partial}{\partial T}, \quad \tilde{S}_2 = 1 + is\frac{\partial}{\partial T}, \tag{4}$$

where $v = \omega_0\beta_1/\beta_0 = v_p/v_g$, $s = 1/\omega_0\tau_p$.

The terms on the right-hand side of each equation of the set (2) represent linear dispersion, diffraction, nonlinearity and cross-phase modulation, respectively. The prefactor of the diffraction term denotes the effect of space-time focusing, and the prefactor of the nonlinear term includes the effects of SS, resulted from the SVEA as well as the dispersive permeability (the equation for electric field) and permittivity (the equation for magnetic field), and higher-order dispersive nonlinear terms resulted from the linear dispersive property of MM. From the expressions for b_m, s, g_m and q_m, it is obvious that the higher-order linear and nonlinear dispersion terms become more important as the pulse width decreases. For ordinary dielectrics, $M_{nl} = 0$ and $\mu_r = 1$, and thus $\tilde{G} = 0$. In this case, the propagation equation for electric field, i.e., the first equation in Eq. (2) is identical to the few-cycle pulse propagation equation of Ref. [38]. If we further make use of the approximation [19], this equation is reduced to the propagation equation obtained by Brabec and Krausz [39].

It can be seen from the Fig. 1 that the MI gain band shrinks as the self-steepening coefficient increases and disappears after the self-steepening coefficient arrives at a critical value [20, 21]. As stated before, the self-steepening effect can be engineered, thus the MI can be manipulated. These results illustrate not only the unusual nonlinear effects that can be seen in NIMs, but also the new ways of manipulating solitons.

Due to the direction of wave vector is opposite to the Poyntiny vector in NIM, which will induce anomalous spatial and spatiatemporal MI. There is large different between the spatial MI of NIM and the conditional material, such as, spatial MI only occurs in defocusing NIM, contrary to that

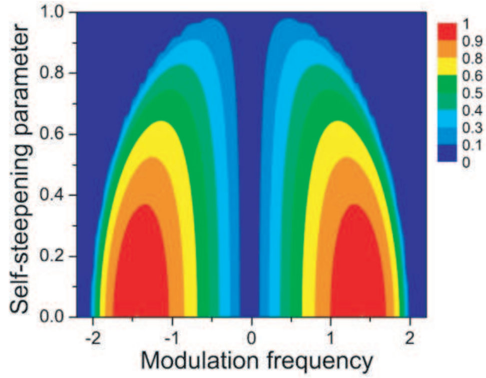

Figure 1: Modulation instability gain spectrum for different SS parameter S1 in the abnormal dispersion regime of negative-index material (From [21]).

in ordinary material, in which spatial MI only occurs in the focusing regime. This opposition is due to the fact that negative refraction reverses the sign of the diffraction term, with the nonlinearity coefficient unchanged. The special spatial MI will result in anomalous spatiotemporal in NIM. Fig. 2 is the spatiotemporal under three conditions. Comparing the results with those in ordinary nonlinear dispersive material [20, 21], we find an interesting fact that the spatiotemporal MI in NIM for a definite combination of dispersion and nonlinearity is just that in ordinary material for a combination of opposite dispersion and opposite nonlinearity. For example, the gain spectra in the three cases, corresponding to Figs. 2(a)–2(c), are, respectively, the same as those in ordinary material for the cases of (i) defocusing nonlinearity and normal dispersion, (ii) focusing nonlinearity and anomalous dispersion, and (iii) focusing nonlinearity and normal dispersion. The physical origin of the fact is that the negative refraction reverses the phase velocity, and thus reverses the diffraction term. This leads to the diffraction and normal dispersion being equivalent when acting with nonlinearity in NIM, while in ordinary material, diffraction is equivalent with anomalous dispersion. In addition, spatiotemporal MI shows how diffraction and dispersion act together to couple space and time. It occurs due to the simultaneous presence of temporal MI and spatial MI in a nonlinear medium, unlike independently occurring temporal MI and spatial MI. Thus the three cases in which spatiotemporal MI can occur can also be obtained by combining the former two kinds of MI, which provide more chance for the form of spatiotemporal soliton.

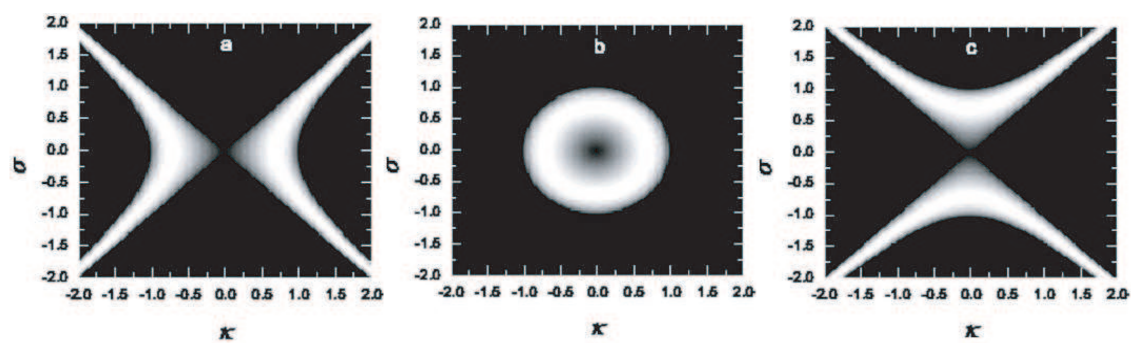

Figure 2: Spatiotemporal MI gain in the (κ, σ) plane for the cases of (a) focusing material with anomalous dispersion, (b) defocusing material with normal dispersion, and (c) defocusing material with anomalous dispersion (From [20]).

The dispersive permeability not only will result in the additional SS, but also will result in a series higher-order dispersive nonlinear terms. Wen et al. focus their attention on the additional MI phenomena inducing by the 2nd-order nonlinear dispersion effect in MI and show that a new kind of MI can occur in the normal group-velocity dispersion (GVD) regime and at the zero GVD point of the MMs, which is resulting from the additional 2nd-order nonlinear dispersion term induced by the dispersive magnetic permeability [40]. This provides a method for generating a train of ultrashort pulse in normal GVD regime, and there is significant value in the practical application.

The influence of the 2nd-order dispersion on MI in the forenamed three cases is plotted in Fig. 3.

In the case (a), the 2nd-order nonlinear dispersion parameter can be positive or zero or negative. An abnormal phenomenon is that the coherent MI occurs when $s_2 < 0$, as Fig. 3(a) shows. Here the negative 2nd-order nonlinear dispersion acts as the role of the anomalous GVD, and thus makes the otherwise impossible MI possible. In addition, we see from Fig. 3(a) that the partial coherence tends to suppress the MI induced by the 2nd-order nonlinear dispersion.

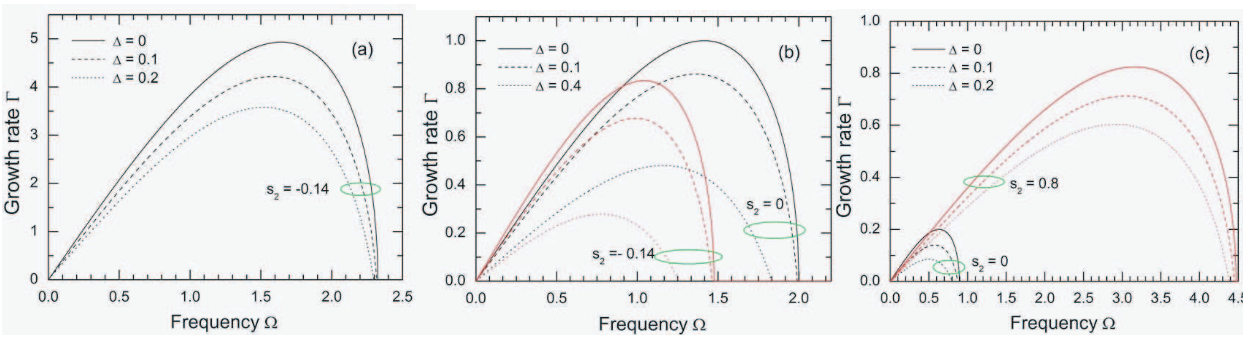

Figure 3: Influence of the 2nd-order nonlinear dispersion on MI in metamaterial. (a) MI gain spectrum in the negative-index region with normal dispersion, (b) MI gain spectrum in the negative-index region with anomalous dispersion, and (c) MI gain spectrum in the positive-index region with anomalous dispersion (From [40]).

In the case (b), the 2nd-order nonlinear dispersion parameter is negative. The influence of 2nd-order nonlinear dispersion on MI in both full and partial coherence is demonstrated in Fig. 3(b). It is shown that, both the 2nd-order nonlinear dispersion and the partial coherence tend to suppress MI in this case. In the case (c), the 2nd-order nonlinear dispersion parameter is positive. We see that the partial coherence tend to suppress MI, while the 2nd-order nonlinear dispersion enhance MI in this case.

To demonstrate this analytical prediction they get the numerical result through the using the standard split-step Fourier method for $\omega_{pm}/\omega_{pe} = 0.8$ in the case of focusing nonlinearity and no linear dispersion [55]. The initial field distribution is a cosinoidally modulated plane wave, $u(T, 0) = u_0[1 + a_0 \cos(\Omega T)]$, where $u_0 = 10$ is the initial amplitude of background wave and a_0 is the initial amplitude of modulation wave which is set to be 0.05, Ω is chosen such that it is the fastest growing frequency for the corresponding case. The numerical results are shown in Fig. 4, and Fig. 4(b) is plotted under the condition of neglecting the SS effect. As shown in Fig. 4(b), the cosinoidally modulated plane wave evolves into a train of pulses with much higher amplitude than the initial modulation as the propagation distance increases. Compared with Fig. 4(b), Fig. 4(a) shows that the center of modulated wave or generated pulse moves toward the leading side and the modulation growth rate is slowed down, which is due to the role of negative SS. These numerical results further demonstrate that MI can appear in the case of no linear dispersion.

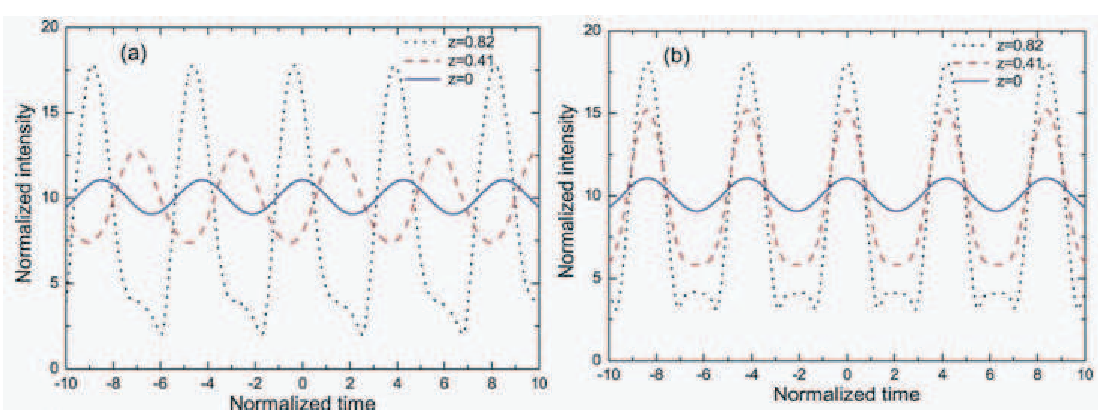

Figure 4: Temporal distributions of the field intensity of cosinoidally modulated plane wave at different propagation distances for different parameters at the zero GVD point (a) $s_1 = -0.3122$, $s_2 = -0.1424$; (b) $s_1 = 0$, $s_2 = -0.1424$ (From [55]).

4. APPLICATIONS OF NONLINEAR MMS

Nonlinear media with negative index of refraction have a variety of potential application. From a nonlinear applications perspective, nonlinear MMs in the optical regime are very attractive owing to the availability of high intensity light sources. There has been tremendous progress in the area of NIMs in the optical regime with a variety of structures and negative index wavelengths. Nonlinearities of MMs stated above suggest their novel application such as frequency conversion, tunable transmission, nonlinear beam focusing, seconding-harmonic imaging, soliton propagation, spatiotemporal soliton or light bullet. Furthermore, for the unconventional nonlinear properties of MMs, there are still some novel applications.

4.1. Opaque Nonlinear Left-handed Lenses

Pendry et al. found that the flat slab of NIMs can focus electromagnetic waves, and they argued that a slab of lossless NIM with $\varepsilon = \mu = -1$ should behave like a perfect lens enabling to obtain an ideal image of a point source through the amplification of the evanescent components of the field [7]. While recent experimental demonstrations confirmed the main features of negative refraction of the NIMs, near-perfect imaging by a flat lens and near-field focusing are severely constrained because of strong dissipation and dispersion of MMs [41, 42]. Nevertheless, numerical studies indicate that nearly-perfect imaging should be expected even under realistic conditions when both dispersion and losses are taken into account [43, 44]. Most of the properties of NIMs have been studied only for linear waves. However, Zharov et al. have studied the second-harmonic generation in a slab of NIM with quadratic nonlinear response [45]. They demonstrated that such a slab can act as a nonlinear lens and it can form an image of the second-harmonic field of the source being opaque at the fundamental frequency, with the resolution that can be made better than the radiation wavelength.

They considered a slab of NIMs and assume that each SRR includes a nonlinear element (a diode inserted into the SRR slot). As a result, the amplitudes of the magnetic momenta induced in the unit cell by the magnetic field $\mathbf{H}$ and $-\mathbf{H}$ are different, and the response of such an effective quadratic nonlinear medium should include the second-harmonic field. They choose the material parameters, making the refractive index of second-harmonic wave (2ω) is -1 and the refractive index of fundamental frequency ω is imaginary number, and the fundamental waves do not penetrate into the slab. However, an effective nonlinear quadratic response of the MM allows the process of the second-harmonic generation. Using the so-called undepleted pump approximation, they show that the second-harmonic field can propagate through the slab creating an image of the source behind the slab.

4.2. Subwavelength Discrete Solitons

Yongmin Liu et al. studied the metal-dielectric multilayers (MDMLs) [46], this kind MMs is periodic structures consisting of nanoscaled metallic and dielectric slabs [47]. They show that the MDML with nonlinear (Kerr-type) dielectric can exhibit self-focusing of light, and form subwavelength discrete solitons. The formation of such solitons is a result of the threefold interplay between periodicity, nonlinearity, and surface plasmons tunneling, leading to new and intriguing phenomena that are not found in nonlinear DWGAs. They have addressed the issue of the intrinsic loss in such structures, calculated the gain required to compensate such losses, and suggested a feasible configuration in which subwavelength discrete solitons could be experimentally observed.

4.3. Optical Bistability and Optical Switch

NIMs posses simultaneously negative dielectric permittivity and magnetic permeability, which will result in the refract light in the opposite way with respect to what in an ordinary material. According this special property, Giuseppe D'Aguanno study a Negative Index Fabry-Perot Etalon, and show the presence of bright and dark gap soliton is supported in a single slab of material [48]. It is surprising is that in a single slab of frequency dispersive NIM together with a cubic nonlinearity appears to support both bright and dark gap soliton. Following this work, they predicted the existence of gap solitons in a nonlinear, quadratic Fabry-Perot cavity [49], they found that an intense, fundamental pump pulse is able to shift the band edge of negative index cavity, and make it possible for weak second harmonic pulse initially tuned inside the gap to be transmitted, giving rise to a gap soliton, the process is due to cascading, which causes pulse compression due to self-phase modulation. Moreover, Litchinister et al. investigated nonlinear transmission in a layered structure consisting of a slab of positive index material with Kerr-type nonlinearity and a subwavelwngth layer of linear negative index material sandwiched between semi-infinite linear dielectrics [50], they found that a thin layer of NIM leads to significant changes in the hysteresis width when the nonlinear slab is illuminated at an angle near that of total internal reflection, and the unidirectional diodelike transmission with enhanced operational range is demonstrated. These results suggest that NIMs could find further application in all optical switching devices and all-optical buffering, for example.

Nonlinear optical couplers have also attracted interesting for the potential application of the all optical switching, power limiting, and so on. Natalia M. Litchinitser describes a novel nonlinear optical coupler structure that utilizes a NIM [3, 12] in one of the channels and a conventional positive index material (PIM) in another channel. They found such a nonlinear coupler (NLC) can be bistable, which is resulted from that the backward coupling between the modes propagating in the PIM and NIM channels enabled by the basic property of NIMs, oppositely directed phase velocity, and the Poynting vector, results in optical bistability in PIM-NIM NLC and gap soliton formation. These effects have no analogies in conventional PIM-PIM couplers composed of uniform (homogeneous) waveguides with no feedback mechanism [51].

4.4. Compensating Losses by Four-wave Mixing

The most detrimental obstacle toward applications of NIMs is strong absorption that is inherent to this class of materials. The possibility to overcome such obstacles based on three wave optical parametric amplification (OPA) in NIMs was shown in [14, 52]. In Section 2.2, it has demonstrated the feasibility of compensating losses in NIMs with second-order nonlinearity by optical parametric amplification. For the MMs with Kerr-nonlinearity, Popov et al. have proposed a approach to compensate losses, which basic idea is that a slab of NIM is doped by four-level nonlinear centers [53]. Chowdhury et al. have shown that second-order and third-order nonlinear susceptibilities and wave-mixing properties of NIM are important [54], especially in this material the three- and four-wave mixing can be naturally phase matched. In the approach, the frequency ω_4 falls in the NI domain, whereas all the other frequencies are in the positive index domain. They demonstrated that the losses of NIMs for strongly absorbing can be compensated by embedded optical nonlinearities. Further, they have studied the resonant FWM-based OPA in such composite NIM at the frequency of the signal and a positive index for all other coupled waves. In particularly, the strong nonlinear optical response of the composite can be adjusted independently, which is because nonlinear optical response is determined by the embedded four-level nonlinear centers.

5. CONCLUSIONS

In this paper, we briefly review some typical nonlinear phenomena of electromagnetic pulse propagate in the nonlinear MMs, such as second-order harmonic generation (SHG), optical parametric amplification (OPA), four-wave mixing (FWM), optical soliton, self-phase modulation (SPM), and self-focusing and so on. Some unique properties of nonlinear MMs have been stated. Furthermore, some potential applications for the nonlinear MMs are also addressed and reviewed. With the rapidly development of nanofabrication and sub-wavelength imaging techniques, the optical MMs will be fabricated more easy, and the nonlinear MMs will get great development. Although further material developments are still needed especially to minimize the losses present in currently available MMs, the novel and controllable properties of nonlinear MMS are very promising and point toward the possibility to achieve various applications in the future optical communication technology.

ACKNOWLEDGMENT

This work is supported by the National Natural Science Foundation of China (Grant Nos. 10674045 and 10576012), the National High Technology Research and Development Program for Inertial Confinement Fusion of China, the Program for New Century Excellent Talents in University and the Specialized Research Fund for the Doctoral Program of Higher Education of China (Grant No. 20040532005).

REFERENCES

1. Veselago, V. G., "The electrodynamics of substances with simultaneously negative values of e and μ," *Sov. Phys. Usp.*, Vol. 10, No. 4, 509–514, 1968.
2. Berman, P. R., "Goos-Hänchen shift in negatively refractive media," *Phys. Rev. E*, Vol. 66, No. 6, 067603, 2002.

3. Ziolkowski, R. W., "Superluminal transmission of information through an electromagnetic metamaterial," *Phys. Rev. E*, Vol. 63, No. 4, 046604, 2001.
4. Ziolkowski, R. W. and A. D. Kipple, "Causality and double-negative metamaterials," *Phys. Rev. E*, Vol. 68, No. 2, 026615, 2003.
5. Smith, D. R., W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, "Composite medium with simultaneously negative permeability and permittivity," *Phys. Rev. Lett.*, Vol. 84, No. 18, 4184–4187, 2000.
6. Smith, D. R. and N. Kroll, "Negative refractive index in left-handed materials," *Phys. Rev. Lett.*, Vol. 85, 2933–2936, 2000.
7. Pendry, J. B., "Negative refraction makes a perfect lens," *Phys. Rev. Lett.*, Vol. 85, 3966, 2000.
8. Zharov, A. A., I. V. Shadrivov, and Y. S. Kivshar, "Nonlinear properties of left-handed metamaterials," *Phys. Rev. Lett.*, Vol. 91, 037401, 2003.
9. Lapine, M., M. Gorkunov, and K. H. Ringhofer, "Nonlinearity of a metamaterial arising from diode insertions into resonant conductive elements," *Phys. Rev. E*, Vol. 67, 065601, 2003.
10. Shalaev, V. M., W. Cai, U. K. Chettiar, H. K. Yuan, A. K. Sarychev, V. P. Drachev, and A. V. Kildishev, "Negative index of refraction in optical metamaterials," *Opt. Lett.*, Vol. 30, 3356–3358, 2005.
11. Dolling, G., M. Wegener, C. M. Soukoulis, and S. Linden, "Negative-index metamaterial at 780 nm wavelength," *Opt. Lett.*, Vol. 32, 53–55, 2007.
12. Shalaev, V. M., "Optical negative-index metamaterials," *Nature Photonics*, Vol. 1, 41–48, 2007.
13. Agranovich, V. M., Y. R. Shen, R. H. Baughman, and A. A. Zakhidov, "Linear and nonlinear wave propagation in negative refraction metamaterials," *Phys. Rev. B*, Vol. 69, 165112, 2004.
14. Popov, A. K. and V. M. Shalaev, "Negative-index metamaterials: Second-harmonic generation, Manley-Rowe relations and parametric amplification," *Appl. Phys. B*, Vol. 84, 131–137, 2006.
15. Klein, M. W., C. Enkrich, M. Wegener, and S. Linden, "Second-harmonic generation from magnetic metamaterials," *Science*, Vol. 313, 502–504, 2006.
16. Shadrivov, I. V., A. A. Zharov, and Y. S. Kivshar, "Second-harmonic generation in nonlinear left-handed metamaterials," *J. Opt. Soc. Am. B*, Vol. 23, 529–534, 2006.
17. De Ceglia, D., A. D'Orazio, M. De Sario, V. Petruzzelli, F. Prudenzano, M. Centini, M. G. Cappendu, M. J. Bloemer, and M. Scalora, "Enhancement and inhibition of second-harmonic generation and absorption in a negative index cavity," *Opt. Lett.*, Vol. 32, 265–267, 2007.
18. Klein, M. W., M. Wegener, N. Feth, and S. Linden, "Experiments on second- and third-harmonic generation from magnetic metamaterials," *Opt. Express*, Vol. 15, 5238–5247, 2007.
19. Scalora, M., M. S. Syrchin, N. Akozbek, E. Y. Poliakov, G. D'Aguanno, N. Mattiucci, M. J. Bloemer, and A. M. Zheltikov, "Generalized nonlinear Schrödinger equation for dispersive susceptibility and permeability: Application to negative index materials," *Phys. Rev. Lett.*, Vol. 95, 013902, 2005.
20. Wen, S., Y. Wang, W. Su, Y. Xiang, X. Fu, and D. Fan, "Modulation instability in nonlinear negative-index material," *Phys. Rev. E*, Vol. 73, No. 3, 036617-1-6, Copyright @ 2006, Optical Society of America, March 24, 2006.
21. Wen, S., Y. Xiang, W. Su, Y. Hu, X. Fu, and D. Fan, "Role of the anomalous self-steepening effect in modulation instability in negative-index material," *Opt. Express*, Vol. 14, No. 4, 1568–1575, Copyright @ 2006, Optical Society of America, February 22, 2006.
22. Wen, S., Y. Xiang, X. Dai, Z. Tang, W. Su, and D. Fan, "Theoretical models for ultrashort electromagnetic pulse propagation in nonlinear metamaterials," *Phys. Rev. A*, Vol. 75, 033815, 2007.
23. Lazarides, N. and G. P. Tsironis, "Coupled nonlinear Schröinger field equations for electromagnetic wave propagation in nonlinear left-handed materials," *Phys. Rev. E*, Vol. 71, 036614, 2005.
24. Kourakis, I. and P. K. Shukla, "Nonlinear propagation of electromagnetic waves in negative-refraction-index composite materials," *Phys. Rev. E*, Vol. 72, 016626, 2005.
25. Franken, P. A., A. E. Hill, C. W. Peters, and G. Weinreich, "Generation of optical harmonics," *Phys. Rev. Lett.*, Vol. 7, 118, 1961.
26. Lapine, M. and M. Gorkunov, "Three-wave coupling of microwaves in metamaterial with nonlinear resonant conductive elements," *Phys. Rev. E*, Vol. 70, 066601, 2004.

27. Scalora, M., G. D'Aguanno, M. Bloemer, et al., "Dynamics of short pulses and phase matched second harmonic generation in negative index materials," *Opt. Express*, Vol. 14, 4746–4756, 2006.
28. De Ceglia, D., A. D'Orazio, M. D. Sario, et al., "Enhancement and inhibition of second-harmonic generation and ansorption in a negative index cavity," *Opt. Lett.*, Vol. 32, 265–267, 2007.
29. Roppo, V., M. Centini, C. Sibilia, et al., "Role of phase matching in pulsed second-harmonic generation: Walk-off and phase-locked twin pulses in negative-index media," *Phys. Rev. A*, Vol. 76, 033829, 2007.
30. Darmanyan, S. A., M. Neviere, and A. A. Zakhidov, "Nonlinear surface waves at the interfaces of left-handed electromagnetic media," *Phys. Rev. E*, Vol. 72, 036615, 2005.
31. Hegde, R. S. and H. G. Winful, "Zero-n gap soliton," *Opt. Lett.*, Vol. 30, 1852–1854, 2005.
32. Li, J., L. Zhou, C. T. Chan, and P. Sheng, "Photonic band gap from a stack of positive and negative index materials," *Phys. Rev. Lett.*, Vol. 90, 083901, 2003.
33. Wu, L., S. He, and L. Chen, "Band structure for a one-dimensional photonic crystal containing left-handed materials," *Phys. Rev. B*, Vol. 67, 235103, 2003.
34. Kockaert, P., P. Tassin, G. V. D'Sande, et al., "Negative diffraction pattern dynamics in non-linear cavities with left-handed materials," *Phys. Rev. A*, Vol. 74, 033822, 2006.
35. Tassin, P., L. Gelens, J. Danckaert, et al., "Dissipative structures in left-handed material cavity optics," *Chaos*, Vol. 17, 037116, 2007.
36. Kourakis, I., N. Lazarides, and G. P. Tsironis, "Self-focusing and envelope pulse generation in nonlinear magnetic metamaterials," *Phys. Rev. E*, Vol. 75, 067601, 2007.
37. Pendry, J. B. and D. R. Smith, "Reversing light: Negative refraction," *Phys. Today*, Vol. 57, No. 6, 37–44, 2004.
38. Kinsler, P. and G. H. C. New, "Few cycle pulse propagation," *Phys. Rev. A*, Vol. 67, 023813, 2003.
39. Brabec, T. and F. Krausz, "Nonlinear optical pulse propagation in the single-cycle regime," *Phys. Rev. Lett.*, Vol. 78, 3282–3285, 1997.
40. Zhuo, H., S. Wen, X. Dai, Y. Hu, and Z. Tang, "Influence of nonlinear dispersion on modulation instability in metamaterials," *Appl. Phys. B*, Vol. 87, No. 4, 635–641, Copyright @ 2007, Springer, May 17, 2007.
41. Shelby, R. A., D. R. Smith, and S. Schultz, "Experimental verification of a negative index of refraction," *Science*, Vol. 292, 77, 2001.
42. Parazzoli, C. G., R. B. Greegor, K. Li, B. E. C. Koltenbah, and M. Tanielian, "Wave-vector-dependent exciton exchange interaction," *Phys. Rev. Lett.*, Vol. 90, 107401, 2003.
43. Cummer, S. A., "Simulated causal subwavelength focusing by a negative refractive index slab," *Appl. Phys. Lett.*, Vol. 82, 1503, 2003.
44. Feise, M. and Y. S. Kivshar, "Sub-wavelength imaging with a left-handed material flat lens," *Phys. Lett. A*, Vol. 334, 326, 2005.
45. Zharov, A. A. and N. A. Zharova, "Subwavelength imaging with opaque nonlinear left-handed lenses," *Appl. Phys. Lett.*, Vol. 87, 091104, 2005.
46. Liu, Y. M. and G. Bartal, "Subwavelength discrete solitons in nonlinear metamaterials," *Phys. Rev. Lett.*, Vol. 99, 153901, 2007.
47. Economou, E. N., "Surface plasmons in thin films," *Phys. Rev.*, Vol. 182, 539, 1969.
48. D'Aguanno, G., N. Mattiucci, M. Scalora, and M. J. Bloemer, "Bright and dark gap solitons in a negative index Fabry-Pérot etalon," *Phys. Rev. Lett.*, Vol. 93, 213902, 2004.
49. Scalora, M., D. D'Ceglia, G. D'Aguanno, et al., "Gap solitons in a nonlinear quadratic negative-index cavity," *Phys. Rev. E*, Vol. 75, 066606, 2007.
50. Litchinister, N. M., I. R. Gabitov, A. I. Maimistov, and V. M. Shalaev, "Effect of an optical negative index thin film on optical bistability," *Opt. Lett.*, Vol. 32, 151–153, 2006.
51. Litchinitser, N. M. and I. R. Gabitov, "Optical bistability in a nonlinear optical coupler with a negative index channel," *Phys. Rev. Lett.*, Vol. 99, 113902, 2007.
52. Popov, A. K. and V. M. Shalaev, "Compensating losses in negative-index metamaterials with optical parametric amplification," *Opt. Lett.*, Vol. 31, 2169, 2006.
53. Popov, A. K., S. A. Myslivets, T. F. George, and V. M. Shalaev, "Four-wave mixing, quantum control, and compensating losses in doped negative-index photonic metamaterials," *Opt. Lett.*, Vol. 32, 3044–3046, 2007.

54. Chowdhury, A. and J. A. Tataronis, "Nonlinear wave mixing and susceptibility properties of negative refractive index materials," *Phys. Rev. E*, Vol. 75, 016603, 2007.
55. Xiang, Y. J., S. C. Wen, X. Y. Dai, Z. X. Tang, W. H. Su, and D. Y. Fan, "Modulation instability induced by nonlinear dispersion in the nonlinear metamaterials," *J. Opt. Soc. Am. B*, Vol. 24, No. 1, Copyright @ 2006, Optical Society of America, 2008 (to be published).