Effect of longitudinal epsilon-near-zero regime in dynamics of ultrashort laser pulses in hyperbolic metamaterials

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Abstract. Optical properties of hyperbolic metamaterials (HMMs) are in stark contrast to properties of ordinary media that fuels interest to various applications of HMMs in photonics. Special attention is attributed to the epsilon-near zero regime (ENZ) of HMMs that is the spectral point in which real part of the permittivity of the HMM becomes zero. This is accompanied by the effects of field enhancement having far-reaching applications. Here we focus on the experimental and theoretical investigation of the propagation of an ultrashort laser pulse through the silver nanorod-based HMM slab in the spectral range over the ENZ. We revealed pronounced resonant change of the pulse delay in HMMs and the transition between the superluminal and slow pulse propagation at the ENZ spectral point. Observed dynamical phenomena are confirmed theoretically and attributed to unusual case when the spectral half of an ultrashort pulse has elliptical dispersion and another has the hyperbolic one. Special attention is payed to the propagation of chirped laser pulses in the HMMs.

1. Introduction
Taming of light dispersion by metamaterials benefits in a wide range of applications in photonics. Recently hyperbolic metamaterials (HMMs), which are uniaxial media with opposite signs of principle components of the dielectric tensor, have attracted much attention. Hyperbolic isofrequency surface of light in HMMs is topologically different from the conventional elliptic dispersion. It is infinite in k-space and supports high-k optical states making HMMs useful for enhancement of light emission [1] and second harmonic generation [2]. Hyperbolic dispersion is intrinsic to a number of natural materials [3] and artificial structures, for example, an array of metal nanorods in a dielectric matrix and metal/dielectric multilayers [4], “Hyperbolic response” of artificial stuctures can be obtained in visible and near-infrared by tuning the volume fraction of metal in the composite [4]. Special interest is aimed to epsilon-near-zero (ENZ) regime of the HMMs, when real part of the principle component of the dielectric tensor of the metamaterial is zero. In the case of nanorod-based HMM, the extraordinary permittivity ($\varepsilon_e$) tends to zero, while ordinary one ($\varepsilon_o$) remains positive. In the ENZ spectral point the transition from the elliptic to the hyperbolic light dispersion takes place with the variation of the light wavelength.

Recently unusual dispersion properties of the HMMs sparked theoretical research efforts in the investigation of ultrashort laser pulse propagation in the HMMs [5]. The ENZ regime of multilayered HMMs was theoretically considered to realize soliton-like propagation of the femtosecond laser pulses in thin HMM [6]. Modulation of near-zero HMMs permittivity by the
optical pump gives superior possibility for an all-optical modulation of the laser pulses that was predicted in multilayer HMMs [7] and demonstrated in nanorod-based ones [8]. In confined geometry of the HMM waveguide the coupling between forward and backward eigenmodes gives the superluminal and slow pulse propagation that was recently predicted [9]. Experimentally the slow-light effect was revealed in hexagonal boron nitride, which exhibits hyperbolic phonon-polaritons [10]. To the best of our knowledge the effect of ENZ resonance in the pulse dynamics in nanorod-based HMMs has not been considered till now, while impressive enhancement of nonlinear optical effect takes place in this regime under action of femtosecond laser radiation [11]. It makes the investigation of interaction of ultrashort pulses with the HMMs primarily important.

In this work we experimentally and numerically investigate the transmission of femtosecond laser pulses through the silver nanorod-based HMMs slab and demonstrate the pronounced resonant change of the pulse retardation in the metamaterial in the spectral vicinity of the ENZ point. The revealed phenomena are accompanied by superluminal and slow-light effects as well as the transition between these regimes. The theoretical description of the observed effects is proposed, together with the consideration of the propagation of chirped pulses in the HMMs.

2. Experiment
Experimental studies of the pulse dynamics in the HMM were performed for the structure consisted of ordered array of silver nanorods with the diameter of about \( d = 34 \) nm grown in anodic aluminum oxide template (AAO). The HMM thickness was about \( L = 570 \) nm while the thickness of AAO was 50 \( \mu \)m. This composite demonstrates resonant losses for p-polarized light near the wavelength of \( \lambda_{ENZ} = 760 \) nm, which is attributed to the ENZ regime of the considered HMM. It is confirmed by the calculation of the spectra of the principle components of the effective dielectric permittivity tensor of the HMM carried out within the effective medium approach [12]. In the ENZ regime the HMM demonstrates zero value of \( Re(\varepsilon_e) \), corresponding to the longitudinal nanorod direction. Due to the sign change of \( Re(\varepsilon_e) \) at \( \lambda_{ENZ} \), the light with shorter wavelengths experiences the elliptic dispersion and the hyperbolic one for larger wavelengths.

The transmission of ultrashort optical pulses through the HMM was studied when using the femtosecond titanium: sapphire laser generating 80 fs pulses and tunable in the spectral range of \( \lambda = 720 \)−900 nm. The retardation of the laser pulses in the HMM slab was measured using the cross-correlation method. Briefly, the laser beam was divided by a beam-splitter into two arms, the first beam passed through the HMM without focusing, the second one was retarded by a motorized delay-line. Both beams were focused by a spherical mirror to the same area of a thin nonlinear BBO crystal. The dependence of the light power of the second-harmonic beam on the temporal delay gave the intensity cross-correlation function \( C(\tau) = \int I_{HMM}(t)I_{ref}(t-\tau)dt \).

Let’s consider the experimental angular dependence of the retardation of the pulse maximum in the HMM for the case of \( \lambda = \lambda_{ENZ} \) [Fig. 1 (a)]. In this dependence the effect of the pulse delay in thick AAO substrate is subtracted from the total delay in the HMM+AAO structure. For this preliminary measurements of the pulse retardation in the AAO without silver nanorods were carried out. We observed a pronounced variation of the pulse delay in narrow angular ranges around 11° and 22°, where the negative retardation turns into the positive one [Fig. 1(a)]. The negative delay achieves \(-29 \) fs and indicates the superluminal pulse propagation. Positive delay of up to 42 fs corresponds to the pulse retardation. These values of the temporal effects correspond to the effective group index of about \( n_g = -14 \) and 24, respectively. The measured \( C(\tau) \) functions for various angles of incidence (\( \theta \)) are shown in Fig. 1(b) exhibiting superluminal effect, which is enhanced with the increase of \( \theta \) for small angles of incidence. Then the transition to the pronounced pulse retardation takes place near \( \theta = 11^\circ \) and \( \theta = 22^\circ \). We demonstrate that the revealed effects are intrinsic to the ENZ regime of the HMM. This is illustrated in Fig. 1(c)
Figure 1. (a) Measured angular dependence of the pulse delay in the HMM for $\lambda = 760$ nm. (b) Measured cross-correlation functions for $\lambda = 760$ nm and various angles of incidence. (c) Experimental spectral-angular dependence of the pulse delay in the HMM.

where the measured pulse delay in the HMM with respect to the light wavelength and the angle of incidence is shown. As seen, the temporal effect is the most significant at $\lambda = \lambda_{ENZ}$.

3. Theory and discussion
To confirm the experimental findings described above, we carried out simulations of the pulse propagation though the HMM slab surrounded by air. Calculations were performed using the binding of fast Fourier transform and the transfer-matrix method assuming that the incident pulse had duration of 80 fs. We consider the chirped incident pulse $E(t) = (1/\sqrt{V}) \exp \left( -\frac{1}{(2V^2)} \right) \exp \left( \frac{i}{(2V^2)} \right) \exp \left( -i\omega t \right)$, where $V = \sqrt{1 + \beta^2}$, modulation phase $\varphi = \left[ \beta/(2V^2) \right] (t/\tau_0)^2 - (1/2) \arctan(\beta)$ and $\beta$ is the chirp parameter.

The results of the calculation of the intensity profile of the transmitted p-polarized pulse with the central wavelength of $\lambda = \lambda_{ENZ}$ are shown in Fig. 2 for various $\beta$. Numerical simulation for

Figure 2. Calculated temporal dependencies of the normalized pulse intensity for various angles of incidence and chirp parameters: (a) $\beta = -2$, (b) $\beta = 0$, (c) $\beta = 1.5$.

the unchirped pulse ($\beta = 0$) exhibits the temporal features of the pulse in the HMMs similar to the experimental ones [Fig. 2(b)]. It demonstrates the negative pulse delay at the increase of $\theta$ up to $12^\circ$ manifesting the superluminal-like effect with the pulse advance up to 40 fs. At this critical angle the pulse profile has two maxima and the pulse advance transforms to the pulse retardation. The similar feature takes place at the second angle of $\theta = 19^\circ$ as in the experiment.

We assert that chirping of the ultrashort pulse has a significant effect in the pulse advance and its retardation in the HMM at the ENZ spectral point. For the negative $\beta = -2$ that corresponds to higher frequency at the trailing edge of the pulse we obtain a strong pulse advance in a wide range of the angles of incidence [Fig. 2(a)]. In turn, when chirp is positive calculations predict an
enhancement of the pulse retardation [Fig. 2(c)]; it is accompanied by a “jump” from the pulse advance to its retardation at $\theta = 10^\circ$ that does not appear for the negative chirp parameter.

Carried out simulations confirm the observation of the enhancement of superluminal and slow light propagation of ultrashort pulses in the spectral vicinity of the ENZ. The spectral dependence of the pulse advance/delay at inclined light incidence has a resonant form with the spectral width of about 11 nm that is in agreement with the experimental results [Fig. 1(c)].

We assert that disclosed dynamical effects are the result of (i) the beating of the pulse field inside the HMM and (ii) its negative phase velocity. Intrapulse beating originates from (i) two-band spectrum of outgoing pulse caused by enhanced optical losses near $\lambda_{ENZ}$ and (ii) simultaneous elliptic and hyperbolic dispersion for various spectral components of the femtosecond laser pulse when it is tuned to the ENZ. The latter leads to significant phase difference between short- and long-wavelength spectral components of the transmitted pulse that gives superluminal fringe move inducing the effective laser pulse advance. We note that Fabry-Perot interference taken into account in simulations does not affect the revealed phenomena due to large optical losses near the ENZ. For chirped pulses the dominant mechanism of the temporal effects is the pulse reshaping caused by the absorption of leading or trailing edges of the pulse resulted from spectral asymmetry of light attenuation in the HMM near the ENZ.

4. Conclusion
Summing up, we observed superluminal and slow propagation of femtosecond laser pulses in metal nanorod-based HMM and the transition between these effects at the variation of the angle of incidence in the spectral vicinity of the ENZ spectral point of the metamaterial. We assert that these phenomena are inherent to the ENZ and disappear away from the ENZ wavelength. The proposed theoretical description of these effects is based on the intrapulse fringe move caused by the resonant absorption at the ENZ and neighborhood of the elliptic and the hyperbolic dispersion of light near the ENZ. We believe that our findings can be used in applications of the HMMs as subwavelength-thick tunable pulse delay lines. Moreover, the observed dynamical phenomena can have a significant effect in nonlinear optics of HMMs.

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