Magneto-optical properties of plasmonic hyperbolic metamaterials

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Abstract. The results of the experimental studies and numerical simulation of optical and magneto-optical properties of composite structures containing gold nanorods in porous anodic alumina templates and coated by a continuous nickel film are presented. We reveal two features in the Faraday rotation spectra of the structures in the vicinity of the wavelengths of 540 nm and 810 nm, which correspond to the epsilon-near-pole and epsilon-near-zero regions, respectively. Experimentally observed distinct enhancement of magneto-optical effects in the structures in the spectral vicinity of these points are consistent with the simulation results and open up novel opportunities for magnetic-field-assisted light manipulation.

1. Introduction

Hyperbolic metamaterials (HMMs) are artificial optical materials characterized by an uniaxial effective permittivity tensor, with components of the opposite signs for directions which are parallel and perpendicular to the optical axis [1-3]. The simplest configurations of HMMs are a metal-dielectric multilayer stack or an ordered array of metal nanowires in a dielectric host material. In both structures the hyperbolic dispersion law is achievable in the optical range (instead of elliptical dispersion in traditional materials) leading to the observation of a number of unusual optical properties that can find a plenty of applications, from high-resolution imaging and lithography [4,5] to biosensing [6] and enhancement of nonlinear processes [7,8].

Due to the dispersion of the metal incorporated into HMMs these artificial materials possess two distinct properties of the spectra of epsilon tensor components. The first one is a change in the sign of the real part of one of the components of the effective permittivity tensor when passing it through zero (Epsilon Near Zero, ENZ). The second one is the appearance of a pole of effective dielectric constant (Epsilon Near Pole, ENP), which leads to a strong optical dispersion. Both pronounced properties of HMMs are accompanied by the enhancement of the birefringence [9] and give rise to the giant anisotropy of optical properties in the spectral vicinity of these features. As well giant enhancement of the electric field of the pump optical radiation leads to amplification of the nonlinear-optical effects [10] and allows one to expect novel optical phenomena, perspective for modern nanophotonics. One of the effective and convenient techniques for the HMMs fabrication is templated electrodeposition of gold or silver inside the cylindrical channels of anodic aluminum oxide (AAO) matrices. It is also well known that in such arrays of nanorods in dielectric matrix two plasmon resonances (along and perpendicular to the nanorods’ axes) can be excited [11].

Possibilities of the light manipulations can be expanded if the HMMs is supplemented by magnetic materials. Arrays of magnetic nickel nanorods have already been studied earlier in [12,13], plasmon-
assisted enhancement of magneto-optical effects was also observed [14-16], whereas these experiments were performed far from the ENZ and hyperbolic dispersion spectral regions. The first experimental studies concerning the magnetic HMMs of different design were presented last year [17,18]. The enhancement of magneto-optical effects was found in the spectral vicinity of the ENZ point in transversal Kerr geometry in nanocomposite containing segmented Ni/Au nanorods [17] and in Voight configuration in the nanostructure consisting of Ni thin film and Au nanorods in alumina template [18]. Here, complex experimental and numerical studies of optical and magneto-optical effects in arrays of gold nanorods in anodic alumina template supplemented by continuous nickel film are presented, with the special accent on the ENZ and ENP features in continuation of work [18].

2. Samples, experimental, and simulation techniques

Arrays of nanorods were formed by templated electrodeposition of gold into the pores of thick AAO films with partially blocked pores. The detailed description of the synthesis procedure is described in [18]. According to the SEM studies, the volume fraction of metal in the nanocomposite is about 8% which is 2.4 times lower than the porosity of the AAO template. The average diameter and length of Au rods are 40±4 nm and 580±40 nm, respectively. To achieve magneto-optical activity of the sample a continuous nickel film of the thickness of about 15±3 nm was deposited onto one side of the template by magnetron sputtering, so that it contacted the vertically aligned Au nanorods (Fig. 1).

For the measurements of the transmission or reflection spectra of the samples, a halogen lamp was used as the source of a broadband light, the polarizer was set up to make p-polarization of the incident light. In the magneto-optical experiments, a DC magnetic field of 1 kOe formed by two permanent ring magnets was applied in the Faraday geometry (along the light beam) or longitudinal Kerr geometry (in the plane of incidence and parallel to the sample’s surface). Both geometries suppose magnetization-induced polarization plane rotation. The magneto-optical effect in the Faraday geometry was measured through the magnetic contrast of the transmission for the analyzer set at 45° with respect to p-polarization. Magnetic contrast is defined as \( \rho = (T(H)-T(-H))/(T(H)+T(-H)) \), where \( T(H) \) is transmittance of the structure for the opposite directions of the applied magnetic field \( H \). Importantly that the beam passes first through the nickel film and then through the nanorods slab. The measure of the longitudinal magneto-optical Kerr effect is the magnetic contrast of the reflection coefficients (measured in a similar manner) for the opposite directions of magnetization. In this experiment the incident beam was directed from the nanorods’ side.

The calculations were performed within the effective medium theory approximation modified for an anisotropy medium, accordingly to [11]. We accounted distribution of the vectorial electric field within the structure and neglecting the spectral dependencies of the nickel gyration vector.

3. Results

Figure 2 represents the transmission spectra of the sample for different angles of incidence \( \theta \). For oblique incidence it reveals two minima centered at approximately 540 nm and 810 nm and that are absent in the case of bulk gold or porous alumina. The first resonance is caused by the transverse local plasmon resonance, corresponding to the electronic oscillations in the direction perpendicular to the long axis of nanorods [11]. Long-wavelength minimum is associated with the collective longitudinal oscillations of the electronic gas in metallic nanorods and vanishes for the normal incidence as there are no components of the fundamental electric field along the rods.

Metamaterials of this type are known to be highly reflective for all angles of incidence [2]. Reflection spectrum of the sample is shown in Fig. 3 demonstrating reflection bands near the pronounced resonance wavelengths.

Fig. 4 represents the wavelength-angular spectra of \( \rho \) measured in the Faraday geometry. For the clarity, several cross-sections of this graph are shown in Fig. 5. Two features can be seen here. The first one is a peak centered at approximately 540 nm with the quality factor decreasing with increasing \( \theta \). The maximal value of the magnetic contrast is about 1.5·10^2 that exceeds by the order of magnitude the corresponding value for the pure nickel film. The second spectral feature is located in the spectral
region 770-970 nm and has a complicated oscillating behavior. The noticeable property of the magnetic contrast in this spectral region is its rising with increasing angle of incidence.

The experimental spectrum of $\rho$ in the geometry of longitudinal magneto-optical Kerr effect for $\theta=45^\circ$ is shown in Fig. 6 (black line). Unlike the dependence attained for the pure nickel film (Fig. 6, red line), the spectrum for the HMM coated by Ni film oscillates in the spectral region under study.

Simulated transmission angular-wavelength spectrum is presented in Fig. 7. It demonstrates two absorption bands for oblique incidence in the spectral regions near 540 and 810 nm that coincides well with the experimental data. Simulated reflection spectrum (Fig. 8) demonstrates multiple oscillations associated with Fabry-Perot modes. The simulated spectrum of magnetic contrast in the Faraday geometry (Fig. 9) shows the enhancement and the sign-reversal of the magneto-optical effect in the spectral range near $\lambda=810$ nm for all nonzero $\theta$ that also coincides with the experimental data.

4. Discussion
First it should be emphasized that optical properties of the HMM composite are defined by two components $\varepsilon_\perp$ and $\varepsilon_\parallel$ of effective dielectric tensor, corresponding to perpendicular and parallel rods direction, respectively. We calculated their spectra in the framework of the effective medium theory [18], it was found that the two revealed features of transmission spectra of the considered metamaterial at about 540 nm and 810 nm are associated with ENP and ENZ points, respectively. Thus, the transition between elliptic and hyperbolic dispersion regimes is realized at $\lambda>810$ nm.

The behavior of the magneto-optical spectra in the vicinity of the ENZ point in the nanocomposite consisting of Ni thin film and hyperbolic medium is associated with the strong anisotropy of the optical properties of HMM in this region. In Faraday geometry p-polarized light falls onto the nickel film, polarization plane rotates. After the magnetic film the polarization plane is slightly inclined with respect to the plane of incidence. The nanorods array screws it up due to the absorption anisotropy of the sample as well as due to the difference between phase velocities of the ordinary and extraordinary beams. The analogous mechanism works in Kerr geometry, whereas the nickel film rotates the
polarization plane of the reflected beam and the HMM slab modifies this rotation. As the giant anisotropy of the optical properties of the structure (absorption and refraction coefficients) in vicinity of the ENZ point for the ordinary and extraordinary beams for oblique incidence, spectral behavior of the magnetic contrast is rather complicated and strongly depends on $\lambda$ and $\theta$. It should be noted that in common case the polarization of the outgoing beam is elliptical [9], this fact cannot be neglected and it does not allow us to calculate the polarization plane rotation from the magnetic contrast data, as frequently can be performed for ferromagnetic films. The amplification of the Faraday rotation in the spectral vicinity of the ENP point occurs due to the strong electric field localization and the effectivity of the light-matter interaction under the transversal plasmon resonance excitation. Similar effects were observed in nonhyperbolic magneto-plasmonic structures of various designs [14-16].

5. Conclusion
Optical and magneto-optical properties of the array of Au nanorods in porous anodic alumina matrix supplemented with continuous Ni film were studied experimentally and numerically. Epsilon-near-pole and epsilon-near-zero points were observed as minima in transmission spectrum of the composite structure. Strong enhancement of the magnetic contrast in Faraday and longitudinal Kerr geometries was revealed in the spectral vicinity of the ENZ and ENP points.

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