Symmetry resonance dependence of harmonic generation in metamaterials

J. Reinhold[1], M. R. Shcherbakov[2], A. Chipouline[1], V. I. Panov[2], T. Paul[3], C. Rockstuhl[3] F. Lederer[3], C. Helgert[1], E.-B. Kley[1], A. Tünnermann[1], A. A. Fedyanin[2] and T. Pertsch[1]

[1]Institute of Applied Physics, Abbe Center of Photonics,
Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany, email: reinhold@uni-jena.de
[2]Faculty of Physics, Lomonosov Moscow State University, Moscow 119991, Russia
[3]Institute of Condensed Matter Theory and Solid State Optics, Abbe Center of Photonics,
Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany

Abstract

The angular emission characteristics of the nonlinear response from metamaterials in the region of local plasmonic resonances are investigated experimentally and theoretically. An analytical model based on nonlinear dynamics of electrons inside the metallic structure of a metamaterial unit cell is proposed and used to describe the characteristic nonlinear radiation pattern at the excitation wavelength of magnetic resonance. The measured angular behavior of the third harmonic generation exhibits quantitative correspondence of the modeled results.

1. Introduction

The majority of remarkable effects in optical metamaterials (MM) appear due to the excitation of localized or surface plasmons in nano-structured metallic inclusions. This can result in for example highly localized electromagnetic field concentrations and leads to the enhancement [1, 2] of nonlinear (NL) responses, which makes the MM's attractive for NL optics. It was shown previously that the NL spectra obey the principles of the local-field enhanced NL response [3]. Although considerable effort was directed toward determining peculiarities of NL optical response caused by excitation of the magnetic resonances [4], an explicit evidence of magnetic mode symmetry-induced contribution still has not been demonstrated.

Here we show the results from experiments and numerical and analytical simulations of third harmonic generation (THG) from a fishnet MM. It turns out that the angular spectrum could neither be explained by means of local field description, nor with the linear transmission characteristics at the third harmonic wavelength. We introduce an analytical model based on dynamics of coupled NL oscillators which reveals the strong influence of the resonance symmetry on NL radiation pattern. With this model we demonstrate that, firstly, the angular behavior of the NL response of the MM's is strongly influenced by retardation effect, and secondly, it is specific to the magnetic resonance of the antisymmetric excitation mode.

2. Oscillator model

The NL radiation is considered to be caused by NL polarization of the metal. Other sources of the nonlinearities are neglected. Setting aside the discussion about the source of the nonlinearity we consider the motion of electrons within two conducting wires of a MM in the framework of the coupled oscillators model with weak nonlinearity. Within the chosen model the phase between the NL oscillations in two

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wires appears to be different at the antisymmetric (Fig. 1(a)) or symmetric (Fig. 1(b)) resonances and is equal to the phase of the relative linear motion multiplied by the order of harmonics n_h. For the antisymmetric resonance the phase is equal to π, for the symmetric one it is zero. We can write down the current densities for the resonances:

$$\mathrm{symmetric:}\quad \mathrm{j^s_y}(\mathbf{r},\mathrm{t}) = \mathrm{n_h}\omega \mathrm{q_0}\sin \mathrm{n_h}\omega \mathrm{t}[\delta(\mathrm{z-b})+\delta(\mathrm{z+b})][\Theta(\mathrm{y-a})-\Theta(\mathrm{y+a})] \tag{1}$$

$$\mathrm{antisymmetric:}\quad \mathrm{j^{as}_y}(\mathbf{r},\mathrm{t}) = \mathrm{n_h}\omega \mathrm{q_0}\sin \mathrm{n_h}\omega \mathrm{t}[\delta(\mathrm{z-b})-\delta(\mathrm{z+b})][\Theta(\mathrm{y-a})-\Theta(\mathrm{y+a})] \tag{2}$$

Here $\delta(y)$ is the Dirac delta function, $\Theta(y)$ is the Heaviside step function and q_0 is the maximum uncompensated charge achieved during the oscillations. a and b are the geometry parameters, as shown in Fig 1. The latter depends on the magnitude of the NL polarization and is proportional to $\chi^{(n_\mathrm{h})}_{\mathrm{eff}}$ and the local field factors at the harmonic frequency $L_{n_\mathrm{h}\omega}(\theta)$ and fundamental frequency $|L_\omega(\theta)|^{n_\mathrm{h}}$. The problem is considered as a two-dimensional one, i.e. x-independent. The solution of the $\mathbf{A}$ potential equation could be expressed with the delayed Liénard-Wiechert potential. Since $\mathbf{H} = \mathrm{curl}\mathbf{A}$ the magnetic field distribution for $\mathbf{H} = [H_x, 0, 0]$ in the antisymmetric case is expressed in cylindrical coordinates and in the far field as follows:

$$H_x = -4kq_0\tfrac{\sin\beta}{r\cos\beta}\sin(ka\cos\beta)\sin(kb\sin\beta)\sin(n_\mathrm{h}\omega t - kr), \tag{3}$$

where $k = n_\mathrm{h}\omega/c$ and $\beta = \theta + \pi/2$. The case of the symmetric resonance can be considered the same way. From this result one can get the angular radiation pattern $R(\theta)$ by the average electromagnetic intensity which the unit cell of the MM emits per unit angle. Taking into account that for a plane wave $\mathbf{r}\cdot(\mathbf{E}\times\mathbf{H}) = rH^2$ and time averaging give the angular radiation pattern for the resonances:

$$\mathrm{symmetric:}\quad \mathrm{R_{as}}(\beta) = \frac{2\mathrm{c}}{\pi}\left[\mathrm{kq_0}\tan\beta\sin(\mathrm{ka}\cos\beta)\sin(\mathrm{kb}\sin\beta)\right]^2 \tag{4}$$

$$\mathrm{antisymmetric:}\quad \mathrm{R_s}(\beta) = \frac{2\mathrm{c}}{\pi}\left[\mathrm{kq_0}\tan\beta\sin(\mathrm{ka}\cos\beta)\cos(\mathrm{kb}\sin\beta)\right]^2 \tag{5}$$

The elaborated model is a simplified one, i.e. it does not take the real phase velocity of the NL radiation inside the MM into account, considering pure symmetric/antisymmetric modes, assuming infinitely dense charge and current distributions etc. Nevertheless, the model gives an explicit way how one can distinguish between symmetric and antisymmetric resonances of the MM by means of its NL optical response. The polar plots in Fig.1(a) and (b) shows the normalized angular dependences of NL signal calculated using (4) and (5) for the antisymmetric and symmetric resonances, respectively, for the same set of parameters. In contrary to asymmetric resonance for the symmetric one there is no local extremum under tilted incidence.

3. Application of the oscillator model on fishnet structure

Here we want to apply the results from the model to describe the angular dependence of the third harmonic response of the fishnet structure at the magnetic resonance. The angular radiation pattern (4) is straightforwardly connected to the angular dependence of THG. Third harmonic radiation is emitted from each unit cell of the MM with the relative phase which depends on the pump angle of incidence. Radiation from each cell interferes to compose the diffraction pattern. The intensity of each diffraction lobe depends on the angle of diffraction via the radiation pattern dependence. If only the zeroth diffraction order is detected then the diffraction angle equals the angle of incidence and thus the radiation pattern is probed by measuring the angular dependence of THG.

The intensity function used is expressed as follows:

$$I(\theta) = B\left(|L(\theta)|^3\cot(\theta)\sin(ka\sin\theta)\sin(kb\cos\theta)\right)^2. \tag{6}$$

The parameters a, b were substituted with the measured values, B is a normalization constant and $|L(\theta)|^3$ describes the linear resonance feature, which is fitted from the measurements. In Fig. 1(c) the intensity function (6) for the third harmonic response is compared with data from experiments and numerical NL simulations [5]. The linear behavior could not explain the angular one of the third harmonic (Fig. 1(d), (e)).

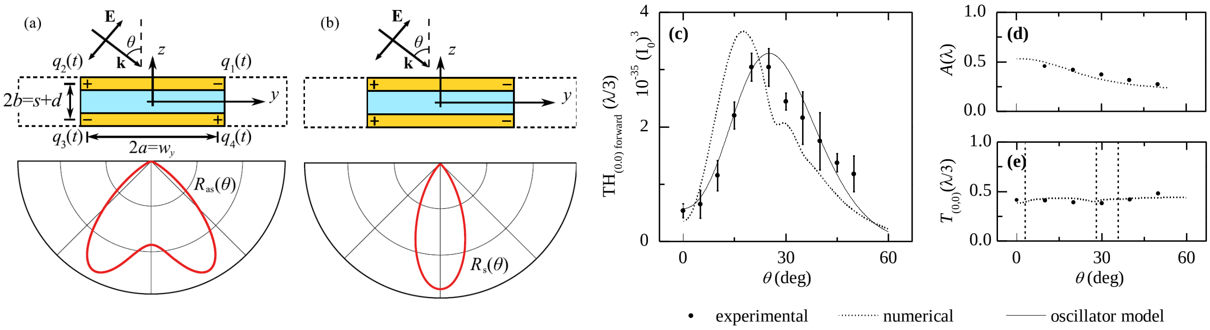

Fig. 1: (a), (b): parameters and far-field radiation pattern; (c): measured, simulated and modeled angular dependence of third harmonic for a fishnet structure at the magnetic resonance; (d), (e): measured and simulated linear angular dependence of absorption at fundamental wavelength and transmission at third harmonic wavelength, vertical lines in (e) corresponds to transmitted diffraction orders

4. Conclusion

An analytical model based on NL dynamics of electrons inside the conductive structure of a MM unit cell is proposed. It explain the specific angular pattern with the oscillations of NL sources which are the intrinsic property of plasmonic resonances. A magnetic-resonance contribution to third-order optical nonlinearities of the fishnet MM was shown. We get a good agreement with the experimental results, and with simulations using the NL Fourier modal method with and the oscillator model. Interference of radiation from separated third harmonic sources is shown to emerge as a local maximum in the angular spectra of the THG efficiency found at abnormal incidence. The results contribute to a better understanding of the possibilities of metamateral NL optical properties tailoring with plasmonic resonances of different symmetries.

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