Magnetic excitations in silver nanocrescents in the blue spectral range: experiments versus calculations

A. Shevchenko[1], R. Khakimov[1], V. Ovchinnikov[2], and M. Kaivola[1]

[1]Department of Applied Physics, Aalto University
P.O. Box 13500, FI-00076 AALTO, Espoo, Finland
Fax: +358- 947023155; email: andriy.shevchenko@tkk.fi
[2]MICRONOVA Nanofab, Aalto University
P.O. Box 13500, FI-00076 AALTO, Espoo, Finland
Fax: + 358–947026080; email: victor.ovchinnikov@tkk.fi

Abstract

We verify numerically the presence of resonant magnetic excitations in the blue spectral region in silver crescent-form split-ring resonators. The resonances originate from asymmetric plasmon modes that resemble electric quadrupole excitations. We also identify another resonance in the red or near infrared, which has a clear magnetic dipole character. These results support our previous experimental findings.

1. Introduction

A common approach to creating metamaterials that show magnetic properties at optical frequencies is to use metal nanostructures, such as split-ring resonators, in which light can excite magnetic dipole moments. Resonators of this type are interesting as they can provide the possibility to adjust the magnetic permeability of the material and to locally enhance the magnetic field component of the light.

The wavelength of the magnetic resonance is known to become shorter for smaller resonators, but if the characteristic size of the resonator approaches 100 nm, the resonance wavelength starts to saturate towards the value of ca. 800 nm. At the same time, the induced magnetic dipole moment decreases. This limiting wavelength has been shown to be approximately the same for split-ring resonators made of silver and gold [1]. Consequently, magnetic excitations at $\lambda < 800$ nm would seem to be unlikely in such resonators. In spite of this, a clear signature of magnetic excitations has been observed at $\lambda < 500$ nm in paired gold nanopillars by Grigorenko *et al.* [2] and at $\lambda < 400$ nm in silver nanocrescents by us [3]. A nanopillar pair can be qualitatively considered as a symmetric split-ring resonator with two slits and a nanocrescent as a split-ring resonator with a large slit.

In this work we give an explanation of the seeming discrepancy between the theory and our experiments with nanocrescents. We numerically calculate the magnetic and electric field distributions around individual nanocrescents illuminated with an optical wave. The calculations show that at λ varying from 370 to 460 nm, the observed magnetic excitations originate from electric quadrupole-like resonances. At long wavelengths, on the other hand, the excitation acquires a pure magnetic dipole character, such as the one described using a simple LC-model. Referring to [4], we note that simultaneous magnetic dipole and electric quadrupole resonances can appear in a metal nanostructure in the form of an asymmetric plasmon mode. Together with the magnetic dipole, the electric quadrupole can contribute not only to a local enhancement of the magnetic field, but also to the magnetization of the material composed of these resonators. Indeed, Maxwell's equations are invariant with respect to the interchange of the electric displacement $\mathbf{D} = \varepsilon_0\mathbf{E} + \mathbf{P} - \nabla\cdot\overleftrightarrow{\mathbf{Q}}$ and magnetic field $\mathbf{H} = \mathbf{B}/\mu_0 - \mathbf{M}$ with $\mathbf{D}'=\varepsilon_0\mathbf{E}+\mathbf{P}$ and $\mathbf{H}' = \mathbf{B}/\mu_0 - (\mathbf{M} + \mathbf{M}_Q)$, respectively. Here $\overleftrightarrow{\mathbf{Q}}$ is the electric quadrupole tensor, and its contribution $\mathbf{M}_Q$ to the overall magnetization $\mathbf{M} + \mathbf{M}_Q$ is found from the equation $\nabla\times\mathbf{M}_Q = -\partial(\nabla\cdot\overleftrightarrow{\mathbf{Q}})/\partial t$

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[4]. With the defined forms for **D**′ and **H**′, both the electric permittivity ε and the magnetic permeability μ of the medium remain independent of the wave vector **k**, which is usually the case when considering metamaterials.

2. Experiments versus calculations

In Ref. [3], we have reported on random arrays of silver nanocrescents that add an orthogonal component to a linearly polarized light passing through the array. Based on symmetry considerations, we attributed the appearance of this component to excitation of circulating electric currents in the nanocrescents. Considering the phase difference between the polarization components of the output light, we came to a conclusion that the structures behave as magnetic dipoles with their resonance wavelength fixed approximately at 370 nm. The array, its transmittance spectrum in crossed polarizers, and the polarization-rotation spectrum are shown in Figs. 1a, 1b, and 1c, respectively.

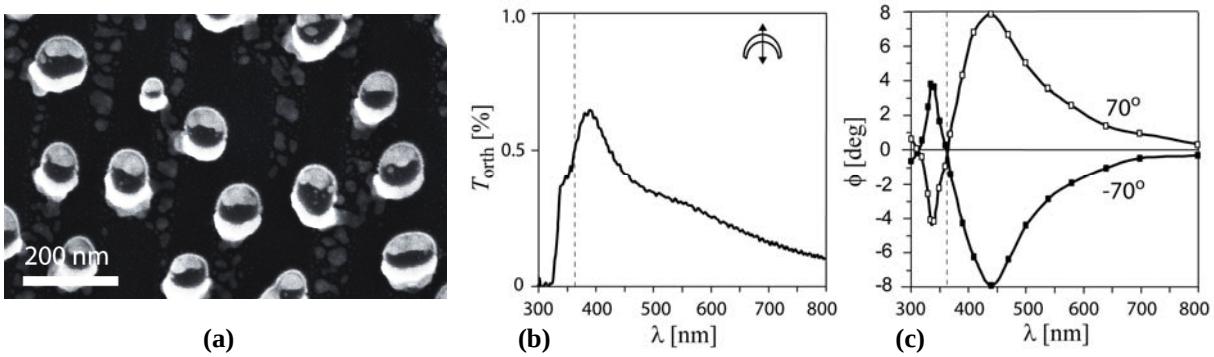

Fig. 1: (a) Top view of silver nanocrescents around dielectric nanopillars. (b) Spectral transmittance of the array through crossed polarizers. The input light is s-polarized (the polarization direction is shown in the inset) and incident on the array at θ = ±70°. It has an intensity of I_{inc}. The transmittance T_{orth} is defined as I_p/I_{inc}, where I_p is the intensity of the p-polarized light at the output. (c) The spectrum of the angle φ by which the principal axis of the polarization ellipse of the transmitted light is tilted with respect to the polarization plane of the incident light [3].

Since the nanocrescents are distributed randomly, periodic boundary conditions cannot be used in the calculations. Therefore, instead of computing the overall response of the array to the incident field, we calculate the response of individual nanocrescents with different sizes. For simplicity, we assume each nanocrescent to be infinitely long, and perform a 2D calculation which, obviously, can give only a qualitative result. Since magnetic excitations should be accompanied by a local enhancement of the magnetic field in the resonators, we calculate the normalized magnetic field intensity $|H|^2/|H_{inc}|^2$ along the inner surface of the nanocrescent and plot its average value as a function of λ (Fig. 2).

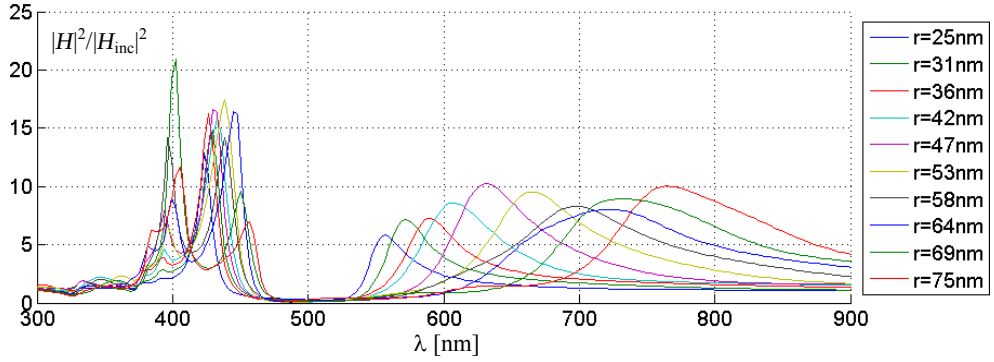

Fig. 2: Enhancement of magnetic field intensity at the inner surface of the nanocrescents. The radius of curvature of the inner surface is r. Its average value for the array of Fig. 1 is 50 nm.

Two sharp peaks in the wavelength range of 370 to 460 nm and one broader peak at a longer wavelength can be seen in the spectra of Fig. 2. These peaks testify for resonant magnetic excitations in the

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nanocrescents. For a nanocrescent corresponding to the average size, r = 50 nm, the magnetic field distributions at several wavelengths and phases of the incident wave are shown in Fig. 3. At a non-resonant wavelength λ = 330 nm (Fig. 3a), the magnetic field is not much disturbed by the resonator, and the electric field vectors at the surface of the crescent (the green arrows) are directed more along the incident electric field than along the surface. At on-resonance wavelengths, the magnetic field is significantly modified and enhanced by the crescent. The nature of the resonances can be inferred from the distribution of the electric field vector. It is seen that at λ = 650 nm (Fig. 3d), the electric current makes a full run along the surface of the crescent, corresponding to a magnetic dipole excitation. At λ = 440 nm (Fig. 3c), on the other hand, the excitation resembles an electric three-charge quadrupole – the electric current runs in opposite directions in the left and right halves of the crescent. Finally, the excitation at 400 nm consists of three opposite currents running along the crescent surface. This excitation is similar to that of an electric four-charge quadrupole.

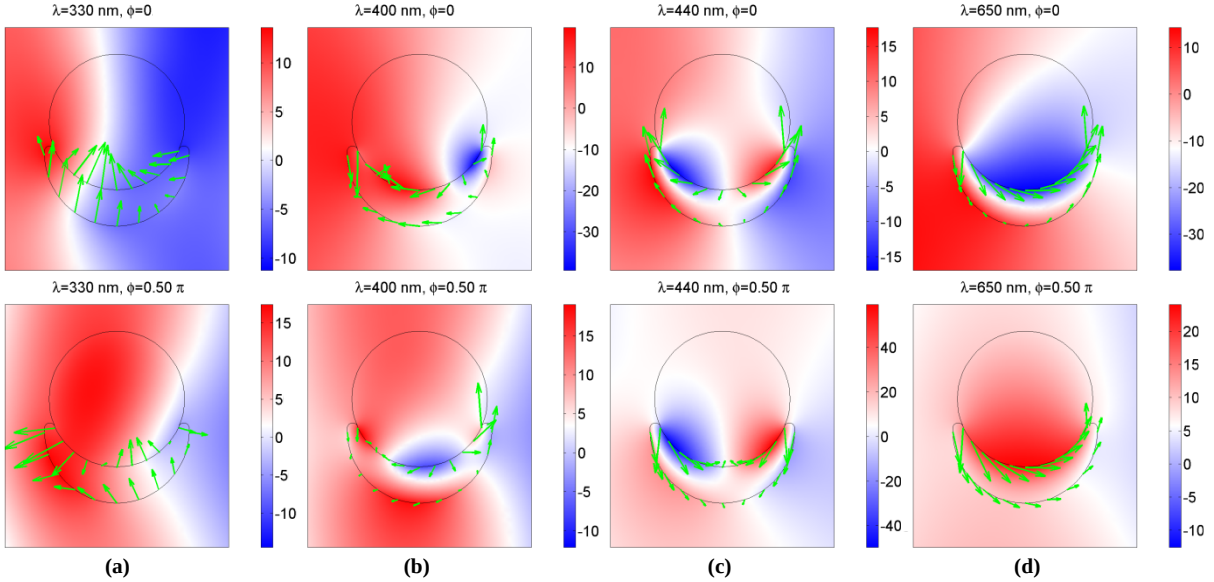

Fig. 3: Magnetic (red-blue colour) and electric (green arrows) field distributions at the phase values of φ = 0 and φ = π/2 of the incident wave at the center of the crescent. The wavelength of the wave is 330 nm in (a), 400 nm in (b), 440 nm in (c), and 650 nm in (d).

3. Conclusions

We have shown that magnetic excitations in silver nanocrescents can have their origin in asymmetric electric quadrupole-like modes in the crescents. These modes have a significant influence on the incident magnetic field, and they take place at shorter wavelengths than the wavelength of a magnetic dipole resonance. These results support our hypothesis about the magnetic character of the excitations observed previously in experiments. In general, it should be possible to obtain magnetic metamaterials at visible light frequencies by using electric quadrupole resonances like those described in this work.

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