Collective magnetic modes excitation in GaAs nanoclusters by azimuthally polarized vector beams

D F Yagudin, M K Kroychuk, A S Shorokhov and A A Fedyanin

Quantum Electronics Department and Quantum Technology Centre, Faculty of Physics, Lomonosov Moscow State University, Moscow 119991, Russia

E-mail: yagudin@nanolab.phys.msu.ru

Abstract. We investigate optical response of oligomers of dielectric nanodisks made of GaAs – anisotropic material with bulk quadratic nonlinearity. By using numerical approach, we analyse linear response of nanoparticle clusters in the vicinity of the magnetic dipole resonance for two types of incident radiation: linearly polarized plane wave and azimuthally polarized vector beams. Achieved results may be used to design highly efficient nonlinear optical nanoantennas with controllable radiation characteristics.

1. Introduction

In the recent years, the use of nanoparticles to control optical radiation at the nanoscale has attracted considerable interest from the international scientific community. At the same time, the implementation of new active nanophotonic devices capable of controlling light requires the development of structures with a high nonlinear optical response. In particular, the effects of optical harmonic generation can allow light frequency conversion at nanoscale. To enhance such effects on submicron sizes, amplification of electric fields in plasmon structures is usually used due to their resonant optical response. However, the observed efficiency of nonlinear frequency conversion in plasmon nanostructures remains small \cite{1}, on the order of \(10^{-11}\), mainly due to heat losses, additionally typical metals have centrosymmetric lattice, which restricts second-order nonlinear effects, such as the second-harmonic generation (SHG).

Recently, all-dielectric nanoantennas have been proposed as a possible way to increase the efficiency of frequency converters \cite{2}. Nanophotonic devices based on dielectric nanoparticles made of materials with a high refractive index feature an optically induced magnetic response due to multipole Mie resonances and thus can provide new possibilities for nonlinear optics. In contrast to plasmonic nanostructures, the electromagnetic field is localized within a dielectric nanoparticle, which leads to a more efficient use of the materials bulk nonlinearity. In addition, when using dielectrics, various materials with a wide range of optical nonlinearities and tensor symmetries become available. For example, metasurfaces made of III-V semiconductors with large second-order nonlinearities demonstrate strong second-harmonic generation induced by an electromagnetic field concentrated inside a nanoresonator \cite{3,4}.

Between adjacent resonant nanoparticles strong near-field interaction may also occur, manifested in a change of the nonlinear optical response of nanostructures due to excitation of collective modes in the whole structure, which can lead to further enhancement of harmonic generation while also imposing a dependence on polarization angle of incident light \cite{5}. Although this effect was studied for isotropic materials, which evidently only allow third-order harmonic generation, in this research we make a next step by analyzing near-field interaction in oligomers consisting out of cylindrical nanoparticles made
out of gallium arsenide – anisotropic material with strong second-order nonlinearity, which will not only allow more intensive harmonic generation, but will also theoretically introduce new interaction between orientation of crystalline structure of the material and geometrical symmetry of oligomer. We specifically analyze two regimes of optical excitation: illumination by a plane wave and by structured light, in particular - azimuthally polarized cylindrical vector beams (CVB). This beam type was chosen since it allows to increase SHG efficiency, furthermore we expect a presence of a collective mode induced by CVB in scattering spectra of oligomers, which may allow for a yet another increase of SHG [6].

2. Sample
The sample under study is a two-dimensional array of GaAs nanodisk clusters: trimers, quadrumers, pentamers and hexamers, arranged in a form of regular polygons (triangle, square, pentagon and hexagon, respectively). We also consider a singular disk in order to demonstrate how lack of near-field interaction will change the nonlinear response. The sample was designed to support magnetic dipole Mie resonances outside of the region of GaAs band-gap. For this task we employed multipolar expansion in accord with Mie theory. Out of all resonances that can be excited in these structures, the magnetic dipole (MD) resonance is the most interesting one, since it allows for a larger field localization within nonlinear material, thus leading to stronger SHG. The nanoparticles are etched from GaAs layer deposited on glass (SiO2) substrate. Distance between clusters is 10 μm, so each one can be considered individually, without taking interaction between them into account. Five different values for spacing between nanodisks within a cluster are considered: 100, 200, 300, 400 and 500 nm, to trace the excitation of the collective mode as the disks in the cluster become closer. The height of all GaAs disks was fixed at 200 nm in order to satisfy beforementioned requirements regarding position of MD resonance.

3. Numerical results
The first step of our calculations is the optimization of the sample’s geometry in the way it can support resonances described above. The numerical calculations were based on the finite-difference time-domain technique using Lumerical FDTD Solutions software package. Optical scattering spectra were simulated to see how they are modified for different incident beam types. As an example, Figure 1b shows a scattering spectrum of a quadrumer for linearly polarized plane wave excitation.

![Figure 1](image_url)

**Figure 1.** a) Electric field profile in the vicinity of magnetic dipolar resonance is shown, white arrows indicate polarization direction of pump beam. b) Optical scattering spectrum for quadrumer excited by a linearly polarized plane wave. Inset shows an example of geometric parameters for resonant nanoparticles, their height is fixed at 200 nm.
We expect that linear response of our structures will only depend on mutual orientation of pump beam polarization and direction of crystalline axis, in case of nonlinear response, however, we predict an additional modulation of signal attributed to geometric symmetries of each cluster and near-field interaction of disks. In case of excitation of oligomers by azimuthally polarized CVB the excitation of the collective mode is predicted to emerge in scattering spectra. Our numerical simulations prove that claim. Figure 2 shows locations of different modes in scattering spectra, and local electric field profile for structures of several types. This figure also demonstrates that in the vicinity of the collective resonance electric field is strongly localized inside nanodisks, which leads to more effective use of bulk nonlinearity and consequently, more intense SHG.

![Electric field distributions for quadrumer and pentamer clusters in proximity to collective mode resonance](image1.png)

![Optical scattering spectra for quadrumer and pentamer excited by azimuthally polarized CVB](image2.png)

**Figure 2.** a) Electric field distributions for quadrumer and pentamer clusters in proximity to collective mode resonance. b) Optical scattering spectra for quadrumer and pentamer excited by azimuthally polarized CVB, “OoPMM” (collective out of plane magnetic mode) - indicate positions of the collective magnetic mode, “MD” – location of the magnetic dipole resonance.

### 4. Conclusion
We numerically demonstrated how excitation by azimuthally polarized CVB can lead to strongly increased field localization in different oligomers made of GaAs. We have shown scattering spectra for different oligomer geometries while considering two types of excitation: by the azimuthally polarized structured light and by the linearly polarized plane wave. Obtained results can be used to design new very efficient nonlinear all-dielectric nanoantennas and subwavelength light sources with controllable radiation characteristics.
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