Ultrafast dynamics of light scattering in resonant GaAs nanoantennas

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Abstract. Active dielectric nanostructures have become one of the most popular trend lines in the modern dielectric nanophotonics due to great opportunities, which are offered by their numerous practical applications. Particularly, the concept of an optical switcher in nanophotonic circuitry can be realized by so-called all-optical mechanism, which could be carried out by a nanoantenna, such as an asymmetric dimer comprised of resonant nanoparticles made of material with high refractive index. In this case all-optical switching can be defined as the change of the trajectory of the femtosecond laser pulse after scattering on the nanoantenna, which was pumped by the other strong laser pulse. The realization of such effects would be a significant advance on the path to a novel technology. In this paper, we numerically demonstrate all-optical switching in the cylindrical asymmetric GaAs nanodimer, with the relaxation time of 10 ps and the deflection angle of the probe pulse of 7° for the relatively low intensities of the pumping (~15 GW cm⁻²).

1. Introduction
Optical metamaterials represent an intensively developing sector of modern nanophotonics. Aiming at widening the range of their performance, they have passed a long way from plasmonic to dielectric nanostructures [1—3]. Dielectrics, apart from low dissipative losses at optical frequencies, provide great opportunities in terms of the scattering properties. Indeed, the magnitude of magnetic dipole (MD) modes in dielectric nanoresonators is comparable with the magnitude of electric dipole (ED) modes and for specific geometries can even exceed it [4]. The interference of the modes in the far field results in various emission patterns of the scattered light. Unidirectional scattering, formed by the interaction of two dipole modes under certain conditions [4], is a particularly relevant example of this interference.

The construction of the conventional dielectric nanoresonators is not designed to provide for the change of the system’s configuration. For the case of unidirectional scattering that means that light of the same wavelength is always scattered in the same direction. Therefore, these nanostructures are passive and their optical properties cannot be externally manipulated. The development of active nanostructures can solve this problem.

Semiconductors can also be chosen as a material for a nanostructure. This class of materials preserves all advantages of dielectrics and is characterized by smaller values of the band gaps. The latter aspect is important for the effective free-carrier injection – phenomenon that leads to the changes of optical properties of the material due to varying concentration of electrons and holes in semiconductor.
The injection can be carried out by optical means. For instance, the optical response of the nanostructure can be changed by the strong laser pumping pulse. As a result, the trajectory of the probe pulse will be modulated, indicating this change. This effect is called all-optical switching. It was numerically demonstrated for an asymmetric Si-nanodimer, composed of two spherical nanoparticles: for the intensity of a pumping impulse \( I_{\text{peak}} = 40 \, \text{GW/cm}^2 \) the deflection angle of the probe pulse was \( \Delta \varphi = 20^\circ \) [5]. It is worth noticing that the considerable change of optical properties can be achieved for lower intensities of the pumping pulse. The measurements of the change in refractive index of the direct-gap semiconductor (GaAs) metasurface demonstrated the change of the reflection coefficient \( \Delta R = 0.35 \) for smaller peak intensities \( \sim 10 \, \text{GW/cm}^2 \) [6].

Taking into account the recent advances in the field of active photonic nanostructures, an idea of a new investigation emerges – experimental observation of all-optical switching in the asymmetric dimer. Experimental character dictates the choice of a new geometry (it is convenient to take disks instead of spheres) and of a new material (GaAs). This structure has never been considered before in the context of all-optical switching, therefore, numerical investigation of light scattering in it becomes attractive, paving the way to the future experimental studies.

2. Results and discussion
The calculations of dynamics of light scattering represent a complex task and should be conducted in several steps. The first one is the optimization of the sample’s geometry.

2.1. Geometry
The idea of the experiment is to transform initially asymmetric scattering indicatrix of the nanoantenna to a symmetric shape by pumping the sample with a femtosecond laser pulse. The symmetrization happens because of the different quantity of the injected free-carriers and, consequently, different changes of the refractive \( n \) and extinction \( k \) indices of the substance. That is why there must be the pronounced difference in the absorption cross-sections of two nanodisks in the vicinity of the pulse central wavelength (820 nm). At the same time, the resonance of the MD mode should be located in the same interval of the spectral domain. Moreover, the dimer must be entirely illuminated, and that fact also adds the condition on the distance between two cylinders.

To meet all the requirements, that were mentioned before, the following parameters were chosen (Fig. 1(a)): \( r_s = 95 \, \text{nm} \) and \( r_j = 80 \, \text{nm} \) (the radii of the larger \( r_s \) and smaller \( r_j \) disks); \( h = 200 \, \text{nm} \) (the height of the disks); \( d = 450 \, \text{nm} \) (the distance between the axes of symmetry of two nanoresonators).

2.2. Change of optical characteristics
The next step was the development of the physical model, which explains the change of \( n \) and \( k \) indices. The quantum description of the electron-hole transitions in semiconductors points out two effects, responsible for the change of optical properties – band filling and band-gap shrinkage. In addition, the classical Drude effect, describing optical properties of the electron gas in metals, should be also considered. The detailed analytical description of some phenomena can be found in literature [7]. To supplement the existing model and, hence, to get the accomplished analytical description, we added a formula for the change of the extinction coefficient, deduced on the basis of the classical Drude theory of metals [8]. The concentrations of electrons \( N \) and holes \( P \) are included as parameters in the analytical expressions for \( \Delta n \) and \( \Delta k \). In further calculations, we assume \( N = P \) (quasineutrality).

2.3. Dynamics of free-carrier concentration and light scattering
The free-carrier concentration dynamics \( N(t) \) can be found as the numerical solution of the Cauchy problem. The differential equation, applied for these calculations, contains terms, representing recombination of the electron-hole plasma, generated by the pumping pulse, and the process of
pumping as well. The information about coefficients for the recombination processes was taken from the literature [6, 9]. The term related to the pump process also was introduced in the equation. The calculations show that the typical time scales are $\sim 0.1 \, \text{ps}$ (for the pumping rate) and $\sim 10 \, \text{ps}$ (for the relaxation rate) for the peak intensities of the pumping pulses around $15 \, \text{GW/cm}^2$ (Fig. 1(b)), what is equivalent to the values of the injected free-carrier concentration $\sim 10^{19} \, \text{cm}^{-3}$.

By putting the calculated dynamics of concentration as a function of time in the expressions for $\Delta n$ and $\Delta k$, the dynamics of material’s dispersion data was obtained. In this way, it becomes possible to simulate the dynamics of light scattering in the asymmetric cylindrical nanodimer, which was carried out by means of 3D finite-difference time-domain simulations [10]. The results prove that the direction of the maximal intensity of the scattered radiation changes on $7^\circ$ for the pumping pulse with $I_{\text{peak}} = 15 \, \text{GW/cm}^2$ (Fig. 1(c)) with characteristic time scale of 10 ps.

3. Conclusion
We have numerically demonstrated all-optical switching in the asymmetric cylindrical GaAs nanodimer. The deflection angle of $7^\circ$ was numerically demonstrated for realistic experimental conditions.

It is remarkable that, pursuing the particular aim to calculate light scattering in the asymmetric dimer, we built the general model of the dispersion dynamics that could be used as a foundation for further numerical investigations of various active nanostructures. The general model for the material dispersion dynamics was developed and introduced to the numerical simulations by FDTD approach, which could be used as a foundation for further numerical investigations of various active nanostructures based on direct-gap semiconductors.
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