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Loss-Compensation in 3D Periodic Arrays of Nanoshells through Quantum Dots, and ε-Near-Zero Metamaterials

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Abstract: We compensate for the losses in 3D arrays of nanoshells through quantum dots embedded in the nanoshells' cores, to achieve *ɛ*-near-zero metamaterials at optical frequencies. Results show loss-compensation or gain-capability in a narrow frequency band. OCIS codes: (250.5403) Plasmonics; (160.3918) Metamaterials; (160.1245) Artificially engineered materials.

1. Introduction

Metamaterials have been proposed for several innovative applications and have laid the ground, for example, for the design of "perfect lenses" and invisibility cloaks. Under certain circumstances of polarization and excitation, a wave in the metamaterial could be described with good approximation as a TEM wave in a homogeneous material, which can in turn be represented by effective parameters (such as permittivity and refractive index). This paper deals with ε -near-zero (ENZ) metamaterials, which have been proposed as a viable way for a number of applications including cloaking, tunneling, and singularity-driven nonlinear phenomena [1]. In particular, we analyze a composite material made of a 3D periodic array of nanoshells at optical frequencies as in Fig. 1.



Fig. 1. Composite material made of a 3D periodic array of nanoshells embedded in a homogeneous medium with permittivity ε_h . Each nanoshell is an InP/ZnS QD (core) coated with a metal (shell). The InP internal radius is $r_{C,1}$, the ZnS has internal radius $r_{C,2}$; the core has an equivalent permittivity $\varepsilon_{\rm C}$; the shell outer radius is $r_{\rm S}$, with permittivity $\varepsilon_{\rm S}$; a, b and c are the periodicities along x-, y- and z-direction, respectively.

Losses in plasmonic-based metamaterials at optical frequencies have been found to be significantly large, which certainly hinder in part their interesting properties and thus limit their practical applications. It follows that loss mitigation mechanisms are inherently required to overcome this issue. A possible solution involves the use of active photonic materials: the gain experienced through the emission of an active medium is capable of counteracting the high absorption losses due to the presence of the metal. Optical loss compensation effects have recently been experimentally observed in [2] and [3], where Coumarin C500 and Rhodamine 6G fluorescent dyes were encapsulated into the dielectric shell of randomly dispersed nanoshell particles. It is worth stressing that molecular dyes have low emitting and absorption cross sections with respect to other active materials. Furthermore, high concentrations of dye molecules may impact in the overall compensation due to fluorescence quenching and other non-radiative phenomena. Loss-compensation has been also shown in [4] using the fishnet structure by using epoxy doped with Rhodamine 800 fluorescent dye molecules as the gain medium, where the experimental results, along with numerical simulations, directly demonstrated that the proposed sample was lossless and active. Effective parameters of metamaterials made of nanoshells with active gain materials embedded in the dielectric core have been simulated in [5] by artificially setting the imaginary part of the dielectric core to fixed ideal loss/gain conditions, and in [6] by using quantum dots (QDs), whose permittivity was modeled through the Lorentz-Drude model. However, no realistic parameters for the gain efficiency have been used in [6]. Theoretical estimations using realistic parameters of fluorescent dyes in 3D periodic arrays of nanoshells have been analyzed in [7] (and

references therein). Here, we assume that InP/ZnS QDs constitute the cores of the nanoshells (with realistic parameters taken from [8]), coated with a silver shell as in Fig. 1.

2. Simulation model and loss-compensation in epsilon-near-zero metamaterial

One of the key points for an effective loss-compensation is designing the metamaterial such that the frequency region of interest overlaps with the emission spectrum of the adopted gain material. The inclusion of QDs eases this requirement, because the emission frequency can be adjusted by modifying their physical dimensions (as a rule of thumb, the smaller the radius of the QD, the larger the energy bandgap, and so the emission frequency). The dielectric function of the QD is assumed to be homogeneous and is calculated using the formalism in [9] as $\varepsilon_{\text{OD}} = \varepsilon_b + \eta S / \left[\omega^2 - \omega_0^2 + i2\omega\gamma \right]$ with the realistic data for InP/ZnS QDs taken from [8]: background dielectric constant $\varepsilon_b = 5.9$, emission at $f_0 \approx 444$ THz ($hf_0 = 1.836$ eV, obtained by setting $r_{C,1} = 2$ nm and $r_{C,2} = 4$ nm), broadening parameter $\gamma \approx 7.59 \times 10^{12} \text{ s}^{-1}$ ($\hbar \gamma = 5 \text{ meV}$), and $S = 4\pi s f_0$, with transition strength $s \approx 1.38 \times 10^{13} \text{ rad/s}$ (hs = 9.1 meV). The population inversion η (see [8-9] for more details) defines the properties of the QD: full inversion ($\eta = 1$, gain) and transparency ($\eta = 0$). The effective relative permittivity of the metamaterial is computed following the steps provided in [7], using two different methods: Maxwell Garnett formulas (MG) and modal analysis in the 3D lattices, using the single dipole approximation (SDA) with nanoshell polarizability taken from Mie theory. The array in Fig. 1 has been designed to have the ε -near-zero frequency band around f_0 : $r_s = 5$ nm, $\varepsilon_h = 2.25$, a = b = c = 15 nm (simple cubic lattice), $\varepsilon_C = \varepsilon_{QD}$ and $\varepsilon_2 = \varepsilon_{\infty} - \omega_p^2 / \left[\omega (\omega + i\gamma_D) \right]$, with $\varepsilon_{\infty} = 5$, $\omega_p = 1.37 \times 10^{16}$ rad/s and $\gamma_p = 27.3 \times 10^{12} \text{ s}^{-1}$ for silver as in [7]. The effective relative permittivity for the 3D array assuming the QD in transparent condition is reported in Fig. 2 (blue and red curves). Note the good agreement between the MG and SDA methods. The effect of the gain provided from the QD in full inversion is reported in the two insets in the ENZ region (black and green curves). Note the resonant behavior of the real part, and the negative peak in the imaginary part which even shows over-compensation capabilities, thus opening up possibility to efficient loss-compensated designs. These results demonstrate the possibility to tailor the effective permittivity to approach virtually zero losses where the metamaterials shows the $\varepsilon = 0$ crossing point, thus creating favorable condition for a number of applications, including low-threshold nonlinear effects.



Fig. 2. ENZ frequency region in presence (shown in the insets) and in absence of loss-compensation, comparing results from MG and SDA.

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