OPTO-ELECTRONICS REVIEW **14**(3), 217–223

DOI: 10.2478/s11772-006-0029-6

Optical bistability of planar metal/dielectric nonlinear nanostructures

R.E. NOSKOV* and A.A. ZHAROV

Institute for Physics of Microstructures, Russian Academy of Sciences, 603950 Nizhny Novgorod, Russia

We study theoretically a nonlinear response of the planar metal/dielectric nanostructures constituted from periodical array of ultra thin silver layers and the layers of Kerr-like nonlinear dielectric. We predict hysteresis-type dependences of the components of the tensor of effective dielectric permittivity on the field intensity allowing the change in material transmission properties from transparent to opaque and back at extremely low intensities of the light. It makes possible to control the light by light in all-optical nanoscale devices and circuits.

Keywords: nonlinear metal/dielectric nanostructures, quasi-static resonance, tensor of effective dielectric permittivity, optical bistability.

1. Introduction

The development of linear and nonlinear optical devices to provide a light localization and light control at nanometer scales is highly important for perspective applications in integrated optical circuits. One of the promising features of the light propagation in a nonlinear medium is bi- or multistability when two or more possible stable self-consistent states of the field take place at the same intensity of external field [1]. This important nonlinear phenomenon is assumed to be used for ultrafast all-optical switching. The possible way to obtain bistability at low light intensities can be based upon using multilayered nonlinear structures. This is caused by electromagnetic field amplification because of resonances arisen due to the light reflections from the layers. There were a number of theoretical studies of nonlinear layered dielectric and metal/dielectric structures as far as the optical bistability is concerned [2–5]. Application of intrinsic electromagnetic resonances of another nature has a special interest because it extends the class of structures demonstrating bi- and multistability. The important possibility on this way opens at using the plane-layered periodic artificial structures consisting of ultrathin metal and dielectric layers with the thicknesses much less than optical wavelength in dielectric medium and than skin-layer width in metal. It is well-known that such a structure possesses, so-called, quasi-electrostatic resonance at optical frequencies (Refs. 6 and 7, and references therein) related to the charge separation in the adjacent layers with different signs of dielectric permittivity. It is well-known, at the same time, that such planar structures behave themselves like effective continuous anisotropic medium and can be described within effective medium approximation [8,9] which has been used in this paper.

*e-mail: rom-noskov@yandex.ru

In this paper we analyse, for the first time to our knowledge, nonlinear properties of such metal/dielectric layered nanostructures in terms of effective medium. We show that the components of the tensor of effective dielectric permittivity depend on intensity of the macroscopic electric field in a nontrivial way, allowing to reach the extremely strong nonlinear properties in optical frequency domain because of the, so called, "geometrical" quasi-static resonance in the system referring sometimes as plasmonic resonance.

2. Formulation of the problem

We consider a metal/dielectric periodical planar structure formed from ultra thin silver and Kerr-like nonlinear dielectric layers, shown in Fig. 1. We suppose that thicknesses of the layers making up the structure are much smaller than electromagnetic wavelength in a dielectric and skin-layer thickness in silver. This condition limits the silver layer thicknesses by several tens of nanometers [7]. Thus, to calculate a nonlinear response of the mentioned structure we can follow the standard approach treating the layered structure like an effective continuous anisotropic medium with effective tensor of nonlinear dielectric permittivity

$$\hat{\varepsilon}_{eff} = \begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix}. \quad (1)$$

(Coordinate axes correspond to those in Fig. 1). This problem is actually reduced to the problem of finding the relation between local and macroscopic field in composite materials containing the nonlinear inclusions (such a problem has been solved, in particular, for the left-handed metamaterials composed from direct metal wires and nonlinear split-ring resonators, Refs. 10 and 11).

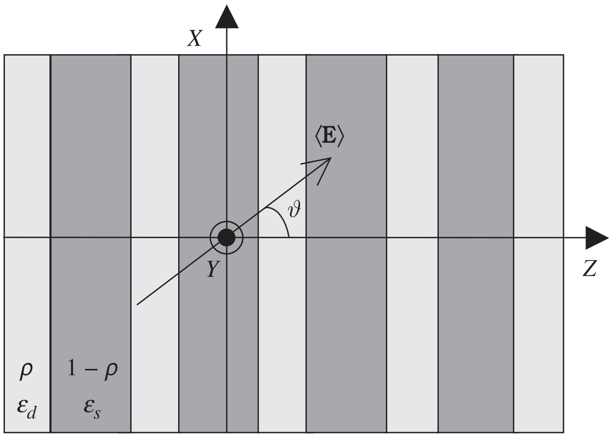

Fig. 1. Schematic of the metal/dielectric layered structure.

Dielectric permittivity of the silver can be described by means of Drude-like formula taking into account nonlinearity of metal nanolayers because of electron confinement and energy quantization effects [12,13]

$$\varepsilon_s = 1 - \frac{\omega_p^2}{\omega(\omega - iv)} + \frac{|\mathbf{E}|^2}{E_{CS}^2}, \quad (2)$$

where ω_p is the plasma frequency, v is the electron scattering rate, and E_{CS} is the characteristic field of silver nonlinearity. Within the wavelength band λ ~600–900 nm, Drude parameters of silver have the following values $\hbar\omega_p$ ≈8.85 eV and $\hbar v$ ≈0.14 eV [14]. Intensity dependent dielectric permittivity of Kerr-like dielectric

$$\varepsilon_d = \varepsilon_0 + \alpha|\mathbf{E}|^2 \big/ E_{CD}^2 \quad (3)$$

assumed to be frequency independent at optical band, where E_{CD} is the characteristic field of nonlinearity, $\alpha = \pm 1$ corresponds to the focusing (+) or defocusing (–) nonlinearity, respectively.

It is quite obvious that the considered structure can be treated as LC-oscillator in the frequency domain, where dielectric permittivities of the silver and dielectric layers have opposite signs. Indeed, the high-frequency electric field directed along the structure axis induces the charges of opposite signs at the interfaces of the layers with positive and negative permittivities (Fig. 2). Therefore one can attribute the usual positive capacitance to the dielectric layer with positive permittivity, whereas the metal layer with negative permittivity has negative capacitance (so it behaves itself like effective inductance). Hence, such a structure looks like a LC-oscillator, the eigen frequency of which depends on dielectric infilling factor $\rho = h_D/d$, where h_D is the thickness of dielectric layers and d is the structure period. In a nonlinear system, this eigen frequency depends on macroscopic field intensity, Eqs. (2) and (3). It is well known [15] that specific property of nonlinear oscillators is the response of a hysteresis (bistable) type. So, one can expect a hysteresis response of the structure as a whole at the frequencies about resonant value.

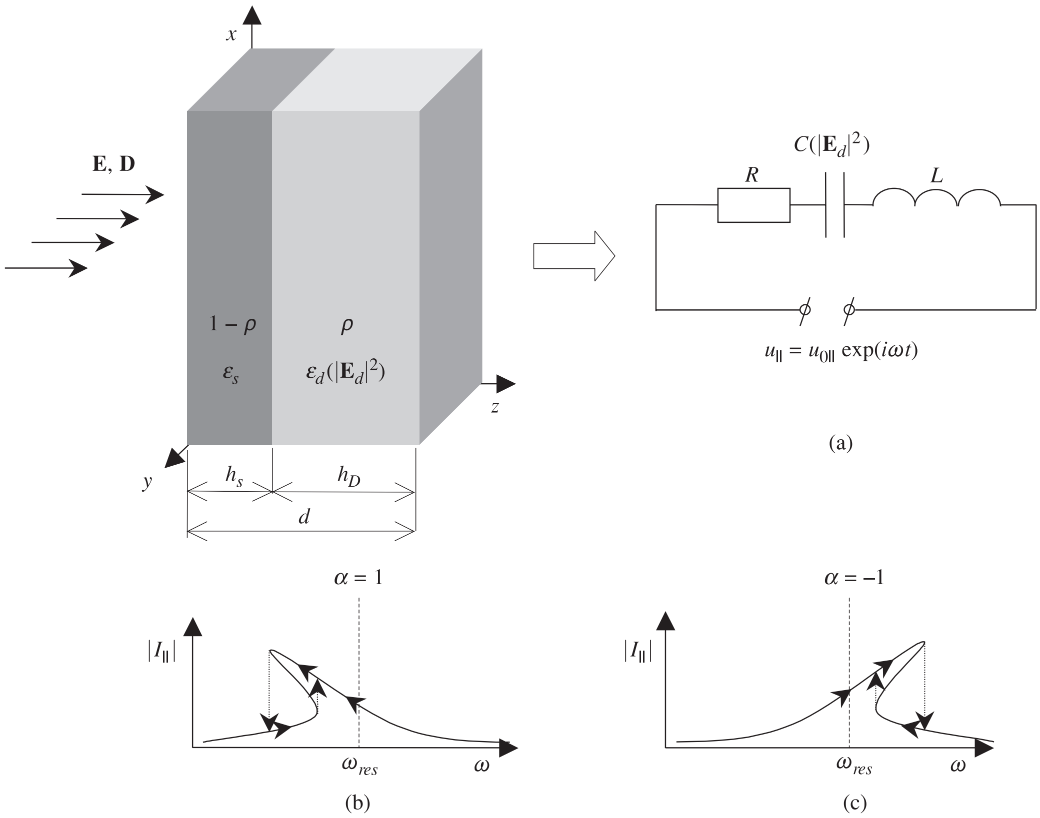

Fig. 2. Metal/dielectric planar structure and equivalent LC-oscillator (a). Resonance curve of nonlinear LC-oscillator with focusing (b) and defocusing nonlinearity (c). ω_{res} is the linear resonance frequency of the oscillator.

3. Results and discussion

In order to get the relation between the components of the tensor of effective dielectric permittivity and intensity of the macroscopic electric field we should satisfy the boundary conditions of the continuity of the tangential components of electric field and normal components of the electric displacement at interfaces between the layers and find out the relation between the local (microscopic) and macroscopic electric fields and electric displacements, which can be obtained by averaging the local fields in the layers over a period of the structure

$$\begin{cases} \langle \mathbf{E} \rangle = \rho \mathbf{E}_d + (1-\rho)\mathbf{E}_s \\ \langle \mathbf{D} \rangle = \rho \mathbf{D}_d + (1-\rho)\mathbf{D}_s \end{cases}, \tag{5}$$

where $\mathbf{E}_{d,s}$, $\mathbf{D}_{d,s}$ are the electric field tensions and electric displacement in dielectric and silver layers, respectively. Angle brackets denote averaging over the structure period. The relations between the local electric field and local electric displacement in the layers are

$$\begin{cases} \mathbf{D}_d = \varepsilon_d(|\mathbf{E}_d|^2)\mathbf{E}_d \\ \mathbf{D}_s = \varepsilon_s(|\mathbf{E}_s|^2)\mathbf{E}_s \end{cases}. \tag{6}$$

Tensor of effective dielectric permittivity, Eq. (1), gives the relation between macroscopic electric field and electric displacement

$$\langle \mathbf{D} \rangle = \hat{\varepsilon}_{eff} \langle \mathbf{E} \rangle. \tag{7}$$

Thus, using the averaged description we come to the equivalent homogeneous anisotropic medium instead of actual multilayered structure. Taking into account the above conditions we obtain the implicit parametric expressions for intensity-dependent components of effective dielectric permittivity tensor in the case of zero silver nonlinearity

$$\begin{cases} |\langle \mathbf{E} \rangle / E_{CD}|^2 = \dfrac{1}{\alpha}(\varepsilon_d - \varepsilon_0) \dfrac{|\rho\varepsilon_s + (1-\rho)\varepsilon_d|^2}{|\varepsilon_s|^2 \cos^2\vartheta + |\rho\varepsilon_s + (1-\rho)\varepsilon_d|^2 \sin^2\vartheta}, \\ \varepsilon_{zz} = \varepsilon_{\parallel} = \dfrac{\varepsilon_d \varepsilon_s}{\rho\varepsilon_s + (1-\rho)\varepsilon_d} \\ \varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{\perp} = \varepsilon_d \rho + (1-\rho)\varepsilon_s \end{cases} \tag{8}$$

where ε_d plays the role of the parameter, the variation of which gives the implicit dependences ε_{xx}, ε_{yy}, ε_{zz} on $|\langle \mathbf{E} \rangle|^2$ in accordance with Eq. (8) (at the focusing nonlinearity (α = 1) $\varepsilon_d > \varepsilon_0$ and at the defocusing one (α = –1) $\varepsilon_d < \varepsilon_0$), ϑ is the angle between macroscopic electric field $\langle \mathbf{E} \rangle$ and z axis (Fig. 1). The results for Ag/AlGaAs structure at $\vartheta = 0$ and $\vartheta = \pi/3$ are summarized in Figs. 3, 4, and 5, where intensity dependences of real and imaginary parts of the effective dielectric tensor components are shown. In these figures, the electric field strength is normalized upon critical field of dielectric nonlinearity E_{CD}.

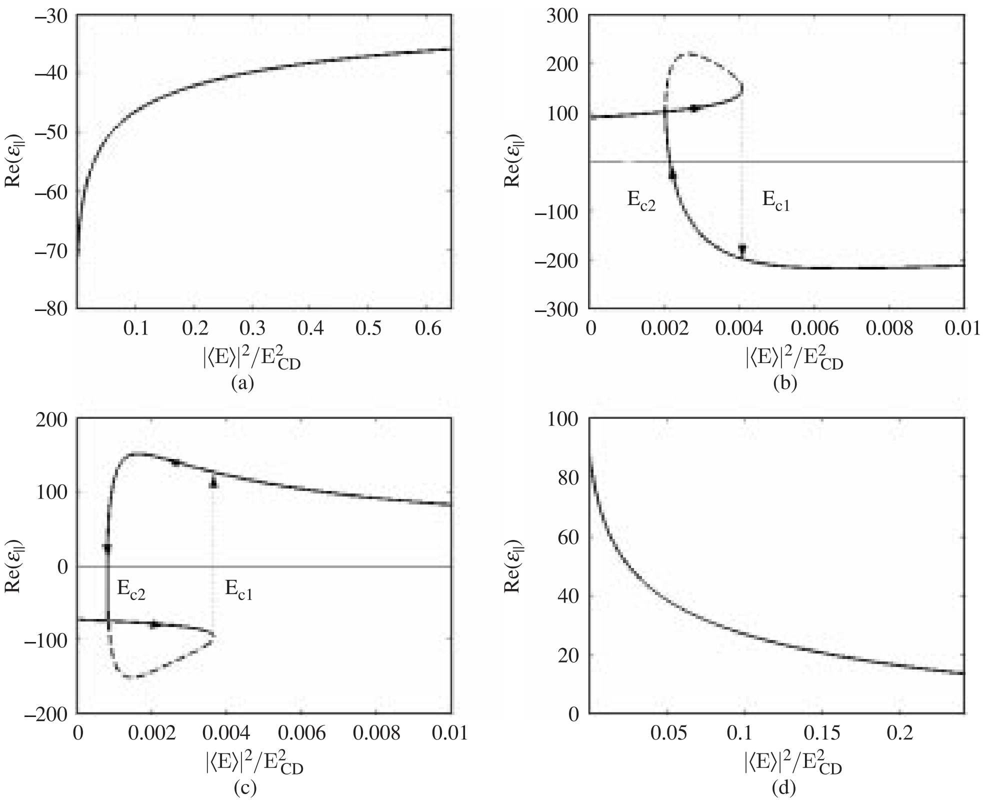

Fig. 3. Real part of the longitudinal component $\varepsilon_{\parallel}$ of the effective dielectric permittivity tensor vs. intensity of the average electric field in the case when $\varepsilon_0 = 8.0$, $\vartheta = 0$, (a) $\hbar\omega = 2.3$ eV, $\alpha = 1$, $\rho = 0.3$, (b) $\hbar\omega = 1.7$ eV, $\alpha = 1$, $\rho = 0.3$, (c) $\hbar\omega = 2.3$ eV, $\alpha = -1$, $\rho = 0.3$, (d) $\hbar\omega = 1.7$ eV, $\alpha = -1$, $\rho = 0.3$. The dashed curves show unstable branches.

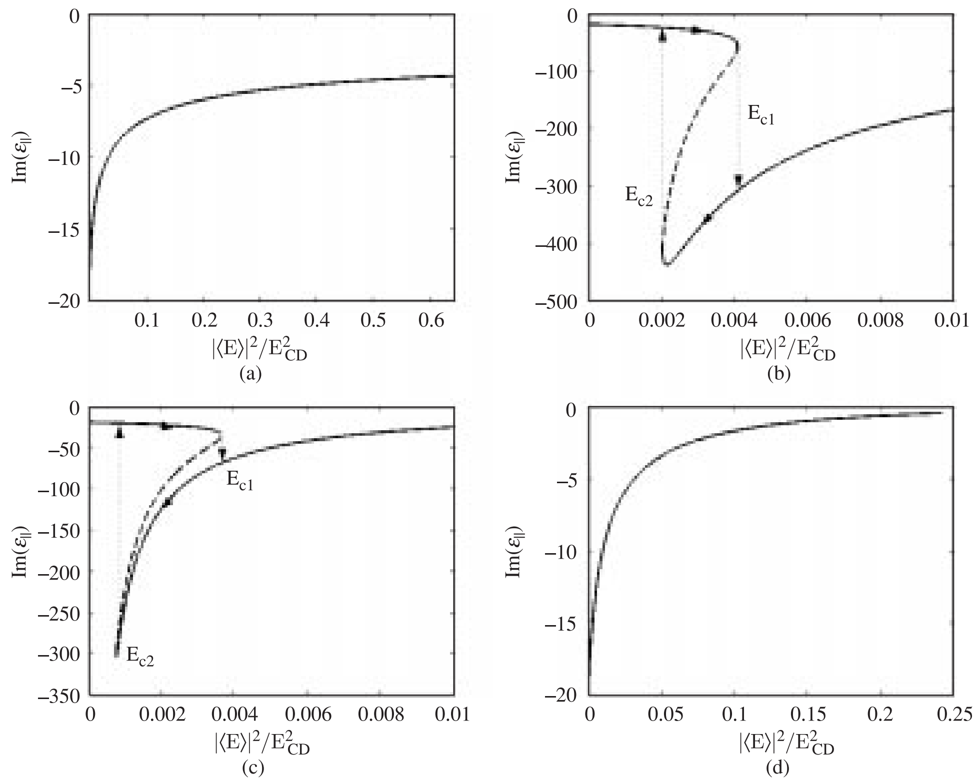

Fig. 4. Imaginary part of the longitudinal $\varepsilon_{\|}$ component of the effective dielectric permittivity tensor vs. intensity of the average electric field in the case when $\varepsilon_0 = 8.0$, $\vartheta = 0$, (a) $\hbar\omega = 2.3$ eV, $\alpha = 1$, $\rho = 0.3$, (b) $\hbar\omega = 1.7$ eV, $\alpha = 1$, $\rho = 0.3$, (c) $\hbar\omega = 2.3$ eV, $\alpha = -1$, $\rho = 0.3$, (d) $\hbar\omega = 1.7$ eV, $\alpha = -1$, $\rho = 0.3$. The dashed curves show unstable branches.

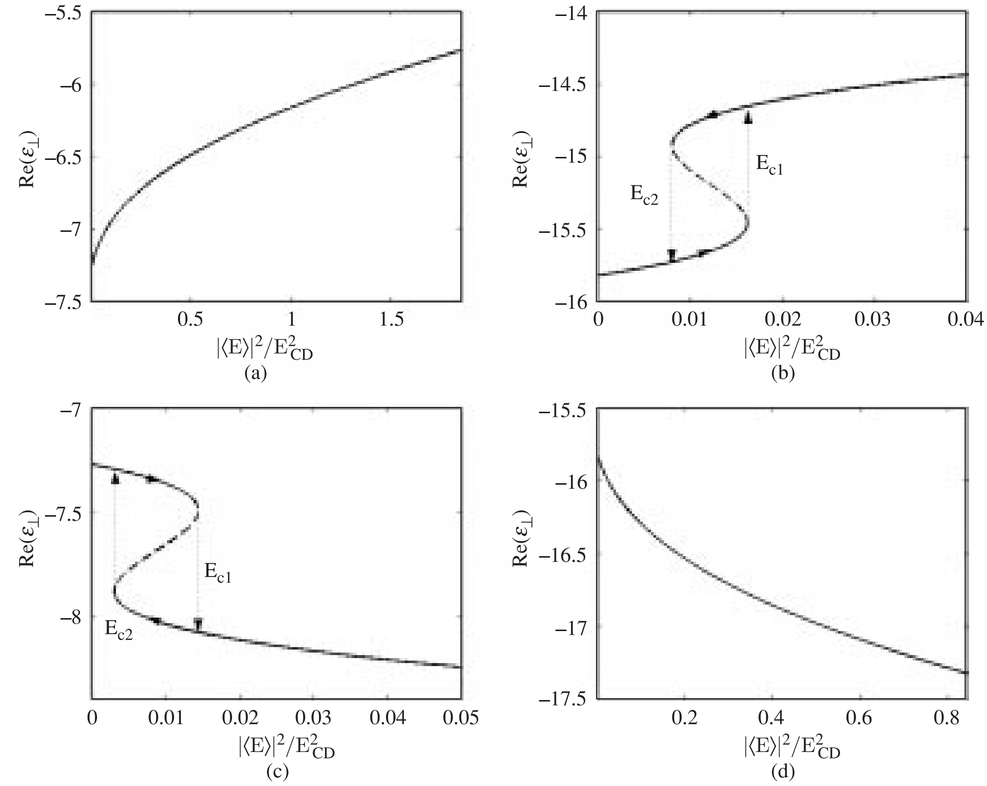

Fig. 5. Real part of the transverse $\varepsilon_{\perp}$ component of the effective dielectric permittivity tensor vs. intensity of the average electric field in the case when $\varepsilon_0 = 8.0$, $\vartheta = \pi/3$, (a) $\hbar\omega = 2.3$ eV, $\alpha = 1$, $\rho = 0.3$, (b) $\hbar\omega = 1.7$ eV, $\alpha = 1$, $\rho = 0.3$, (c) $\hbar\omega = 2.3$ eV, $\alpha = 1$, $\rho = 0.3$, (d) $\hbar\omega = 1.7$ eV, $\alpha = -1$, $\rho = 0.3$. The dashed curves show unstable branches.

The type of behaviour of the curves in Figs. 3, 4, and 5 strongly depends on the sign of nonlinearity and initial (linear) frequency detuning from linear resonance frequency value which corresponds to the zero of the denominator real part in the expression for $\varepsilon_{\parallel}$ in Eq. (8) and in the lossless case, the expression for the resonant frequency can be represented explicitly

$$\omega_{res} = \omega_p \Big/ \sqrt{1 + \frac{1-\rho}{\rho}\varepsilon_0}. \qquad (9)$$

The dependences $\omega_{res}(\rho)$ are shown in Fig. 6 for different dielectrics with different dielectric permittivities.

In the case of focusing nonlinearity (i.e., when $\alpha = 1$), the eigen frequency ω_{res} decreases with the intensity of the electromagnetic field because of a growth of the LC-oscillator capacitance. When $\omega_{res} < \omega < \omega_p$, the real part of the longitudinal component of the effective dielectric permittivity tensor $\mathrm{Re}(\varepsilon_{\parallel})$ monotonically increases being always negative, see Fig. 3(a).

More complicated behaviour of $\mathrm{Re}(\varepsilon_{\parallel})$ is observed for $\omega < \omega_{res}$, this is shown in Fig. 3(b). Here, in the linear limit $\mathrm{Re}(\varepsilon_{\parallel})$ is always positive, but the resonant frequency decreases with the growth of the electric field. Thus, the system is driven into resonance. In this case, $\varepsilon_{\parallel}$ is a three-valued function of the external electric field intensity and this result in jump of the longitudinal component of the effective dielectric permittivity tensor with the growth of the electric field at some $\left|\langle E_z \rangle\right|^2 = E_{c1}^2$ from positive to negative value. Thus, the initially transparent (for appropriate polarization of the light) medium becomes opaque. The reverse transition takes place when the electric field intensity decreases to the value $E_{c2} < E_{c1}$.

In the case of a defocusing nonlinearity (i.e., when $\alpha = -1$), the eigen frequency increases with the amplitude of the external field. That is why the resonance effects take place for $\omega < \omega_{res}$, as shown in Figs. 3(c), 3(d), 4(c), and 4(d). Here, we observe the opposite behaviour when the medium is switched from the opaque to the transparent state at the increasing field intensity. The reverse transition occurs at lower field intensities and in the case when $\omega < \omega_{res}$ $\mathrm{Re}(\varepsilon_{\parallel})$, monotonically decreases with the growing field intensity as shown in Fig. 3(d) and no bistabilities are observed.

Also our results show that the imaginary part of the longitudinal component of the effective dielectric permittivity tensor can be controlled by a proper choice of the external electric field intensity (Fig. 4).

Note that curves in Figs. 3 and 4 are qualitatively similar to those which have been obtained for the split-ring resonators array [10] that reflects the common properties of nonlinear oscillators.

It should be also noted that hysteresis behaviour can also take place for $\mathrm{Re}\varepsilon_{\perp}$ at $\vartheta \neq \pi/2$, as it is shown in Fig. 5 because of the nonlinear coupling of different (longitudinal and transverse) components of the electric field. At the same time, the imaginary parts of transverse components of

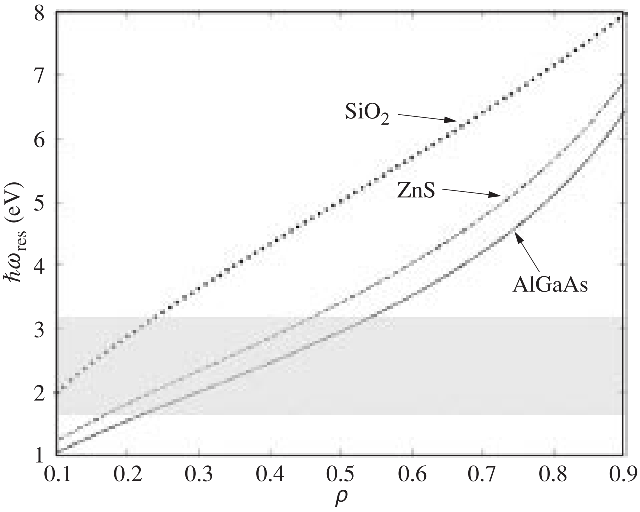

Fig. 6. The dependences of the resonant frequency of the silver/dielectric nanostructures on dielectric infilling factor for different dielectrics: AlGaAs ($\varepsilon_0 \approx 8.0$ – solid), SiO_2 ($\varepsilon_0 \approx 2.1$ – dotted), ZnS ($\varepsilon_0 \approx 5.7$ – dashed). The shaded band indicates the optical spectral range.

dielectric tensor do not depend on the field intensity according to Eq. (8).

The intensity thresholds of switching, shown in Figs. 3, 4, and 5 by arrows can be significantly reduced up to the values which are much less than characteristic nonlinear intensity E_{CD}^2 by the proper choice of operating frequency detuning. Figures 3, 4, and 5 correspond to the relatively large operating frequency detuning from resonance value, in Figs. 3(a), 3(c), 4(a), 4(c), 5(a), and 5(c) $\omega = 1.15\omega_{res}$ and in Figs. 3(b), 3(d), 4(b), 4(d), 5(b), and 5(d) $\omega = 0.85\omega_{res}$. For these cases, the estimations of the switching field strengths for the structures containing microcrystal films of AlGaAs (defocusing nonlinearity at the wavelength λ ~810 nm, $E_{CD} \approx 0.97\times10^6$ V/cm [16]) and the heterostructures GaAs-AlGaAs (focusing nonlinearity at the wavelength λ ~900 nm, $E_{CD} \approx 1.1\times10^6$ V/cm [17]) yield the values $E_{c1} \approx 6.1\times10^4$ V/cm and $E_{c1} \approx 6.9\times10^4$ V/cm, respectively.

In the case when silver nonlinearity is also taken into account, the expression for the tensor of effective dielectric permittivity can be obtained by the same way that has already used. The dependences of effective dielectric tensor components on field intensity are shown in Fig. 7 under assumption of linear dielectric. The hysteresis behaviour can also take place, however, the hysteresis loop is shifted towards significantly higher intensities. This is caused by two factors, first, the critical field of dielectric nonlinearity E_{CD} is much less than silver one ($E_{CS} \approx 1.5\times10^8$ V/cm [13]) and, second, a difference between the values of dielectric permittivities in metallic and dielectric layers in considered frequency band leads to suppression the local field intensity in metal layer in comparison to the dielectric one. Quasistatic approach gives the following factor $E_d/E_s \propto |\varepsilon_s/\varepsilon_d| \approx 2.2$ in our particular case. It means that when both dielectric

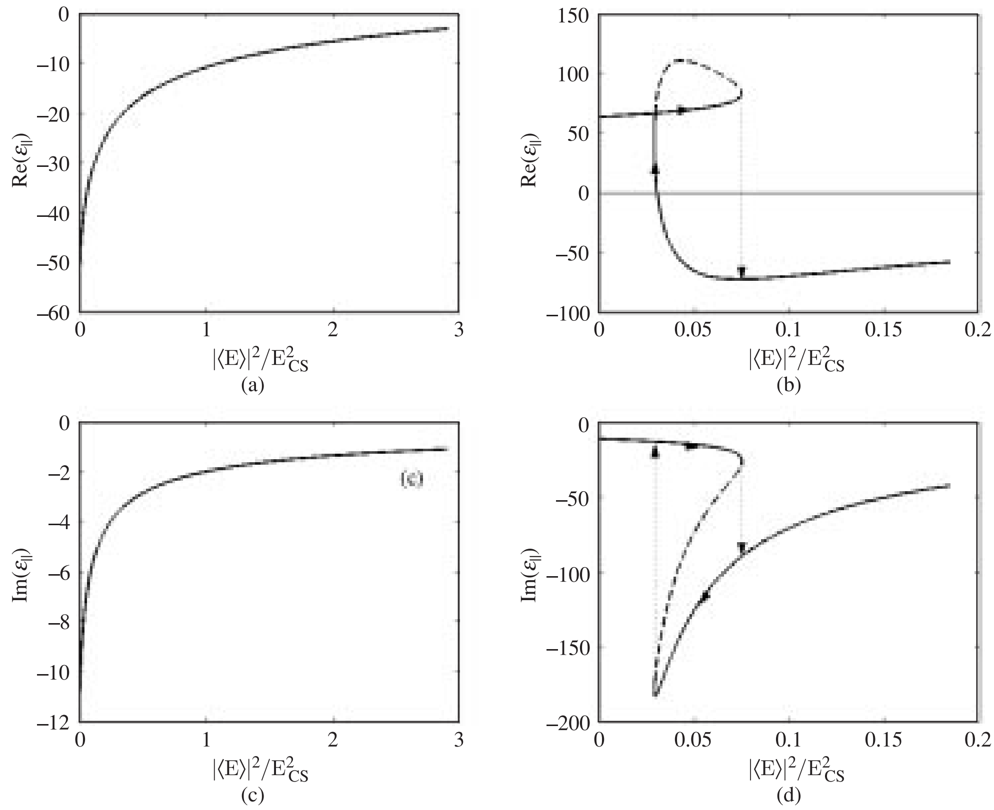

Fig. 7. The dependences of real and imaginary parts of the effective dielectric tensor component $\varepsilon_{||}$ on light intensity for the nonlinear silver/linear dielectric (ZnS) structure for $\vartheta = 0$, (a) and (c) $\hbar\omega = 2.67$ eV, $\alpha = 1$, $\rho = 0.3$, (b) and (d) $\hbar\omega = 1.97$ eV, $\alpha = 1$, $\rho = 0.3$. The dashed curves show unstable branches.

and silver in the structure possess nonlinearities the corresponding dependences of effective dielectric tensor components actually coincide with the curves shown in Figs. 3, 4, and 5 and they can be distinguished only after essential magnification of the scales at the coordinate axes.

4. Conclusions

In conclusion, we demonstrate the optical bistability of the nonlinear metal/dielectric layered nanostructures. The nature of this bistability is the nonlinear quasistatic resonance appeared due to the different signs of the permittivity of dielectric and metal layers that is equivalent to the nonlinear LC-oscillator. We show that the transmission properties of the structure can be changed from transparent to opaque and back at extremely low intensities of the light that opens a perspective to use such composite materials for nanophotonic applications.

Acknowledgements

The authors acknowledge RFBR for support through grant No 05-02-16357.

References

1. H.M. Gibbs, *Optical Bistability: Controlling Light by Light*, Academic, Orlando, 1985.
2. E. Lidorikis, O. Li, and C.M. Soukoulis, "Wave propagation in nonlinear multilayer structure", *Phys. Rev.* **B54**, 10249 (1996).
3. P.K. Kwan and Y.Y. Lu, "Computing optical bistability in one-dimensional nonlinear structures", *Opt. Commun.* **238**, 169 (2004).
4. R.S. Bennik, Y.K. Yoon, R.W. Boyd, and J.E. Sipe, "Accessing the optical nonlinearity of metals and metal-dielectric photonic bandgap structures", *Opt. Lett.* **24**, 1416 (1999).
5. W. Chen and D.L. Mills, "Optical response of nonlinear multilayer structures: Bilayers and superlattice", *Phys. Rev.* **B36**, 6269 (1987).
6. G.B. Smith, A.I. Maaroof, and A.S. Scheim, "Optical response of nanostructed metal/dielectric composites and multilayers", *Proc. SPIE* **5508**, 192–205 (2004).
7. U. Kreibig and M. Vollmer, *Optical Properties of Metal Clusters*, Springer-Verlag, Berlin, 1995.
8. N.A. Khizhnyak, "Artificial anisotropic dielectrics", *Zhurnal Tekh. Fiziki* **27**, 2006 (1957). (in Russian)
9. Y.B. Fainberg and N.A. Khizhnyak, "Artificially anisotropic media", *Zhurnal Tekh. Fiziki* **29**, 711 (1959). (in Russian)
10. A.A. Zharov, I.V. Shadrivov, and Y.S. Kivshar, "Nonlinear properties of left-handed metamaterials", *Phys. Rev. Lett.* **91**, 037401 (2003).
11. N.A. Zharova, I.V. Shadrivov, A.A. Zharov, and Y.S. Kivshar, "Nonlinear transmission and spatiotemporal soli-

tons in metamaterials with negative refraction”, *Opt. Express* **13**, 1291 (2005).
12. K. Uchida, S. Kanenko, and S. Omi, “Optical nonlinearities of a high concentration of small metal particles dispersed in glass: copper and silver particles”, *J. Opt. Soc. Am.* **B11**, 1236 (1994).
13. V.P. Drachev, A.K. Buin, H. Nakotte, and V.M. Shalaev, “Size dependent $\chi^{(3)}$ for conduction electrons in Ag nanoparticles”, *Nano Lett.* **4**, 1535 (2004).
14. P.B. Johnson and R.W. Christy, “Optical constants of the noble metals”, *Phys. Rev.* **B6**, 4370 (1972).
15. M.I. Rabinovich and D.I. Trubetskov, *Oscillations and Waves in Linear and Nonlinear Systems*, Dordrecht, Kluwer, 1989.
16. M.J. La Gasse, K.K. Anderson, C.A. Wang, H.A. Haus and J.G. Fujimoto, “Femtosecond measurements of the non-resonant nonlinear index in AlGaAs”, *Appl. Phys. Lett.* **56**, 417–419 (1990).
17. C. Kutsche, P. LiKamWa, J. Loehr, and R. Kaspi, “Quasi-CW self-guided optical beams in GaAs-AlGaAs double heteroastructure slab waveguides”, *Electron. Lett.* **34**, 906–907 (1998).