Steplike Transmission of Light through a Metal-Dielectric Multilayer Structure due to an Intensity-Dependent Sign of the Effective Dielectric Constant

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We numerically study light propagation through a specially designed nonlinear nanoscale metal-dielectric multilayer structure with a linear effective dielectric constant just below zero. The calculated dependence of the output intensity on the input intensity shows a steplike behavior. It rests upon an intensity-dependent change of the effective dielectric constant from negative (low-transmission state) to positive (high-transmission state) values, corresponding to a transition of the optical properties from metalliclike to dielectriclike. The study of the transient behavior of the structure demonstrates a switching time of around 1 ps.

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The interaction of light with matter in metal-dielectric nanostructured materials brings about many fascinating phenomena leading to insights which facilitated the recent progress in nanoscience, nanotechnology, and plasmonics (for recent reviews see, e.g., [1,2]). The dielectric constant of metals behaves like that of a plasma, which is negative below the plasma frequency. This property can be used to tailor composite materials with a desired dielectric constant. Control of the value and sign of the dielectric constant and the magnetic permeability is the central idea of the fast-growing field of negative-refraction optics [3–5]. Potential applications of this field are superlens imaging with a resolution below the diffraction limit [5] as well as, for example, focusing of scanning beams to a subdiffraction spot [6]. A negative dielectric constant also supports collective electron oscillations (plasmons) near the surface of a metal, responsible for several remarkable effects. Examples are the dramatic local field enhancement in a subwavelength volume used, e.g., for the detection of single molecules [7] or the enhanced transmission through subwavelength hole arrays [8]. Among the different geometrical structures studied up to now, a structure composed of alternating metallic and dielectric layers is a particularly simple and easy-to-create system. Linear transmission properties in such 1D photonic crystals were studied in Ref. [9], where high transmission has been shown even when the total thickness of the metal significantly exceeds the skin depth. Recently, imaging with subwavelength resolution by periodic metal-dielectric layered structures was investigated [10–12]. The nonlinear optical response of a Cu/SiO_2 stack has been studied both theoretically [13] and experimentally [14], demonstrating a strongly enhanced nonlinear transmission compared with the bulk metal with the same total thickness.

In this Letter we apply the degree of freedom of composite metal-dielectric structures connected with the negative dielectric constant of metals and its large nonlinear susceptibility (several orders of magnitude larger than those of dielectrics). By choosing an appropriate volume fraction of the metal and the dielectric, an effective linear dielectric constant of the composite slightly below zero can be achieved. In this case, the composite behaves like a metal with a very small linear transmission. However, a field inside of the slab can increase the effective dielectric constant to positive values due to the optical nonlinearity of one or both components. We numerically study light propagation through such specially designed nonlinear metal-dielectric multilayer structures and predict a steplike bistable transmission which sharply increases with the intensity, due to the change of the effective dielectric constant from negative values with a metal-like low-transmission state to positive values with a dielectriclike high-transmission state.

In Fig. 1(a) the considered structure is presented, which consists of 9 silver layers alternating with fused-silica layers. The thicknesses of the layers are $d_{\mathrm{Ag}} = 13$ nm and $d_{\mathrm{SiO}_2} = 104$ nm, $\Lambda = d_{\mathrm{Ag}} + d_{\mathrm{SiO}_2} = 117$ nm is the pitch of the structure, and $L = 950$ nm is the total thickness. Silver has a linear dielectric constant of $\varepsilon^L_{\mathrm{Ag}}(\omega) = 1 - \omega_p^2/(\omega^2 + i\nu\omega)$ with $\omega_p = 11.5\ \mathrm{fs}^{-1}$, $\nu = 0.083\ \mathrm{fs}^{-1}$ and that for fused silica is $\varepsilon^L_{\mathrm{SiO}_2} = 2.1$ [15] for wavelengths in the range of 500–700 nm. We have chosen silver among the metals due to its relatively low loss as well as the high nonlinear susceptibility in films $\chi_3 = 2.49 \times 10^{-8} + i7.16 \times 10^{-9}$ esu [16], which has a real part notably larger than the imaginary part, in contrast to other metals with higher nonlinearities, such as copper. The nonlinear dielectric constant of silver is given by $\varepsilon^{\mathrm{NL}}_{\mathrm{Ag}} = \varepsilon^L_{\mathrm{Ag}} + \epsilon_0\chi_3 E^2$, and the nonlinearity of SiO_2 is neglected. In order to avoid the plasmonic resonance in the response of the material, we consider a linearly polarized wave with a polarization parallel to the metal films. A plasmonic resonance occurs for metal-dielectric surfaces perpendicular to the polarization, for example, for inclusions in the form of spheres, and would strongly enhance the nonlinearity but also enhance the loss. In the quasistatic

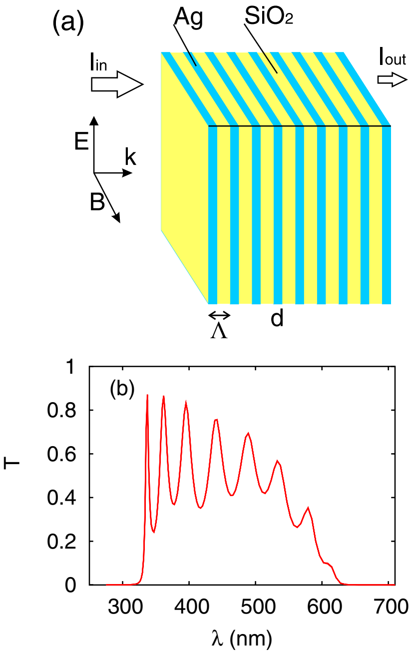

FIG. 1 (color online). Geometry of the metal-dielectric multilayer slab (a) and its linear transmission as a function of the wavelength (b). In (b) the parameters are $d_{\mathrm{Ag}} = 13$ nm, $\Lambda = 117$ nm with 9 layers of silver.

limit $\Lambda \ll \lambda$ for a polarization parallel to the films the effective linear dielectric constant of the slab is determined from the theory of composites simply as

$$\varepsilon_{\mathrm{eff}}^{L} = \frac{d_{\mathrm{Ag}}}{\Lambda}\varepsilon_{\mathrm{Ag}}^{L} + \frac{d_{\mathrm{SiO_2}}}{\Lambda}\varepsilon_{\mathrm{SiO_2}}^{L}. \tag{1}$$

The nonlinear effective dielectric constant $\varepsilon_{\mathrm{eff}}^{\mathrm{NL}}$ can be determined in a similar way, using $\langle\varepsilon_{\mathrm{Ag}}^{\mathrm{NL}}\rangle$ instead of $\varepsilon_{\mathrm{Ag}}^{L}$ in Eq. (1) with averaging over the space. If $\varepsilon_{\mathrm{Ag}}^{L}$ and $\varepsilon_{\mathrm{SiO_2}}^{L}$ have different signs, $\varepsilon_{\mathrm{eff}}^{L}$ with near-zero values can be obtained by choosing appropriate d_{Ag} and $d_{\mathrm{SiO_2}}$.

First we study the linear optical properties of the composite structure. In Fig. 1(b) the linear transmission is presented as the function of the wavelength. The finite-difference time-domain method with perfectly matched layer absorbing boundary conditions was employed to calculate the linear and nonlinear transmission of the structure. The spatial resolution was up to 2000 points per wavelength, to resolve the nanometer-scaled silver films and account for the rapidly changing field at the metal-dielectric interface. One can see a sharp increase of transmission with decreasing wavelength, followed by oscillations. For large wavelengths, the effective dielectric constant is negative; therefore the structure has optical properties of a metal film and, correspondingly, an extremely low transmission. For smaller wavelengths, the absolute value of the $\varepsilon_{\mathrm{Ag}}^{L}$ decreases, which leads to a positive $\varepsilon_{\mathrm{eff}}^{L}$, dielectriclike properties of the structure and a significant transmission. The oscillations are due to the Bragg interference between the waves reflected from the two *outer* surfaces of the structure. For wavelengths in the range of 300 nm, a photonic band gap appears due to the Bragg reflection from the interfaces *inside* the structure, and the effective-medium approach is no longer applicable. However, in this region high silver losses inhibit the studied effects. Note that a plasmonic resonance at 400–600 nm does not appear in the transmission for the here considered case of a polarization parallel to the films, as expected. The sharp feature of the transmission at around 630 nm suggests the use of this spectral region for a study of a possible enhancement of nonlinear properties.

For a qualitative understanding of the nonlinear behavior of the studied metal-dielectric structure we first use the idealized approach of the effective-medium approximation. Let us consider the propagation of a monochromatic wave through a *homogeneous* slab of the material with nonlinear Kerr-like response $\varepsilon_{\mathrm{eff}}^{\mathrm{NL}} = \varepsilon_{\mathrm{eff}}^{L} + \epsilon_0\langle\chi_3\rangle E^2$. The propagation of a cw in a nonlinear homogeneous lossless slab can be described by the formalism developed in Ref. [17], which uses the ansatz $E(z) = |A(z)|\exp[i\phi(z)]\exp(i\omega t)$ to transform the wave equation

$$\frac{d^2E}{dz^2} + \varepsilon_{\mathrm{eff}}^{L}\frac{\omega^2}{c^2} + \frac{\omega^2}{c^2}\langle\chi_3\rangle|E|^2E = 0 \tag{2}$$

into a first-order differential equation

$$\left(\frac{dA}{dz}\right)^2 = B - \left[\frac{W^2}{A^2} + \varepsilon_{\mathrm{eff}}^{L}\frac{\omega^2}{c^2}A^2 + \frac{1}{2}\frac{\omega^2}{c^2}\langle\chi_3\rangle A^4\right] \tag{3}$$

with $d\phi/dz = W/A^2$, where B and W are constants depending on the input and structure parameters. Such an approach is equivalent to exactly solving the Maxwell equations in the slab. However, the loss is not included in this formalism. Figure 2 shows the transmission of the slab with parameters given in the caption, which for a certain range of input intensities exhibits *three* solutions demonstrating a bistable behavior with two stable (red and green solid curves) and one unstable (gray dashed curve)

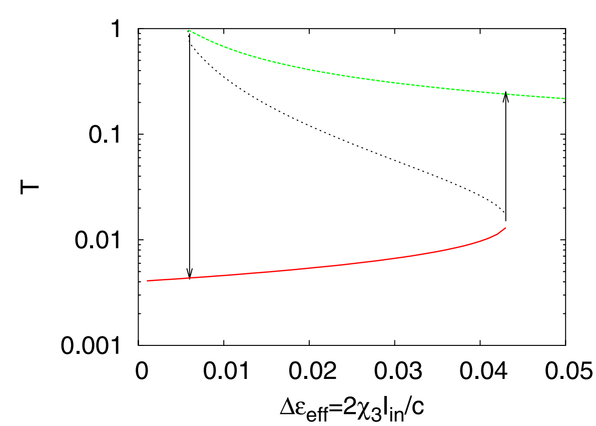

FIG. 2 (color online). Transmission of a homogeneous nonlinear slab as a function of the normalized input intensity for $\varepsilon^L = -0.01$ and a slab thickness of 5λ.

branches and with a transmission contrast between the stable branches of more than 2 orders of magnitude. The bistability mechanism is easily understood. A slab of a material with dielectric constant $\varepsilon_{\rm eff}^{L}$ which is negative but close to zero is nontransparent in the linear regime, for a slab thicknesses in the order of wavelength or more. On the other hand, if an electric field is present in the slab, then due to the nonlinear effects $\varepsilon_{\rm eff}^{\rm NL}$ increases and becomes positive, the optical properties of the slab now correspond to that of a dielectric, and the slab will be transparent. In the latter case the finite transmission sustains the presence of the electric field inside of the slab, thus resulting in bistability.

Optical bistability has attracted significant interest over more than three decades due to possible applications in all-optical information processing [18]. Recently, all-optical switching was achieved with greatly reduced power requirements using challenging and carefully designed resonant nonlinear microcavities in a photonic crystal [19–21]. In these studies, the nonlinear modification of the refractive index shifts the resonance frequency of a high-quality cavity to the frequency of the input wave, resulting in a positive feedback. In contrast to typical bistable systems, the system considered here is much simpler and compact.

The effective-medium approach used in Fig. 2 requires that loss be negligible as well as that the wavelength be large in comparison to the pitch Λ. However, the optical properties of a real metal-dielectric structure strongly depend on the loss of the metal layers. For a more accurate description of the metal-dielectric structure illustrated in Fig. 1(a), now we use the finite-difference time-domain method, taking the linear and nonlinear loss of silver into account.

We consider a cw wave with a wavelength of $\lambda = 633$ nm (e.g., He-Ne laser) normally incident on a structure shown in Fig. 1(a). With parameters as given above the effective linear dielectric constant of the composite is $\varepsilon_{\rm eff}^{L} = -0.11 + i0.052$ at 633 nm. In Fig. 3, the output transmitted intensity is shown as a function of the input intensity. The transmission slowly grows until the input intensity of 2.5 GW/cm^2, and then sharply increases with a contrast of more than 4. In a narrow range of input intensities from 2.5 to 2.58 GW/cm^2 bistability is predicted, with only stable branches observable in the numerical simulations. In comparison with Fig. 2, the loss of the composite reduces the width of the two bistable branches and the contrast, but the sharp steplike increase in the transmission remains. The maximum modification of the silver dielectric constant at the threshold $|\varepsilon_{\rm Ag}^{\rm NL} - \varepsilon_{\rm Ag}^{L}|$ is 2.6. In contrast to dielectrics where nonlinear modification of ε in the order of unity is out of reach, such large nonlinear contribution has been experimentally demonstrated [22] for metals. In Fig. 3(b), the distributions of the squared field amplitudes in states A and B with correspondingly higher or lower transmission are shown. It can be seen that in the higher-transmission state the field penetrates farther

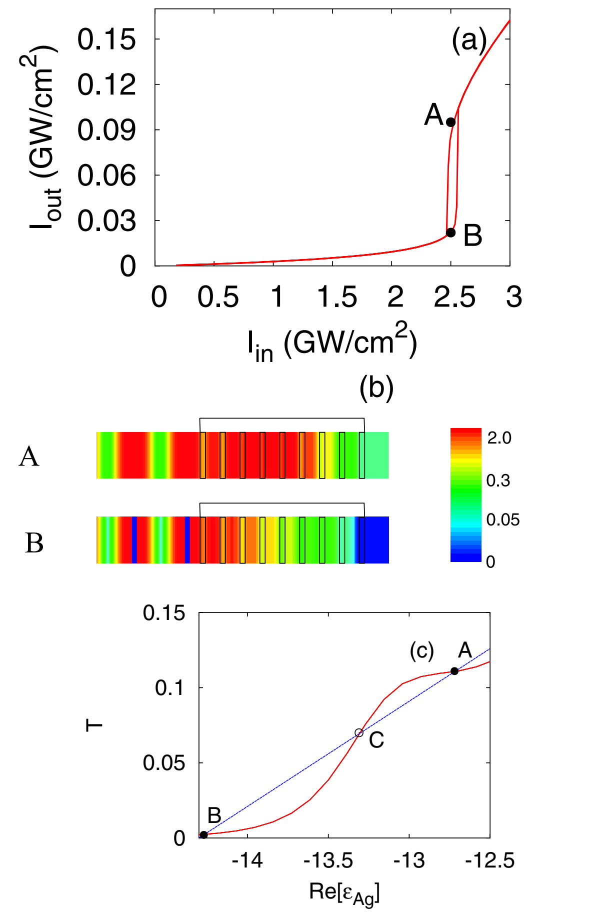

FIG. 3 (color online). Output intensity dependent on the input intensity (a) and the spatial distributions of the squared amplitude of the electric field inside of the structure (b). The geometry of the structure is presented in Fig. 1(a), with parameters given in text. In (b), high fields are denoted by red (gray), low fields by blue (dark gray), the size of the structure is shown by the square bracket above the field distribution, and the metal layers by the small rectangles. In (c), the linear transmission of the structure as a function of Re[$\varepsilon_{\rm Ag}$] (red curve) and the dependence of Re[$\varepsilon_{\rm Ag}$] on transmission (blue curve) are presented.

in the structure. For the lower-transmission state, the intensity of the wave in the part of the structure near the output surface is low, and this part remains effectively "metallic." The mechanism of bistability is not based on a resonance in a cavity, but on the transition of the optical properties from metal-like to dielectriclike. The bistability is further qualitatively illustrated in Fig. 3(c). Consider the silver dielectric constant Re[$\varepsilon_{\rm Ag}$] as an unknown internal parameter which, for qualitative understanding, is assumed to be the same in all the silver layers. Then, we have two equations for the unknown transmission T and Re[$\varepsilon_{\rm Ag}$]. The first equation is given by the transmission T as a function of Re[$\varepsilon_{\rm Ag}$] determined from the linear propagation [red curve in Fig. 3(c)]. In turn, due to the nonlinearity, Re[$\varepsilon_{\rm Ag}$] is proportional to the squared field inside the structure and therefore to the transmission T, providing

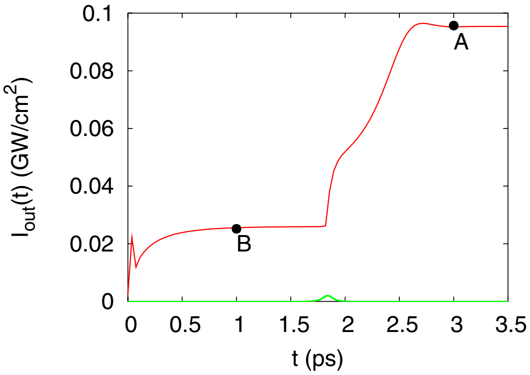

FIG. 4 (color online). The transmitted intensity of the cw wave [dark gray (red) curve] dependent on time. The light gray (green) curve denotes the low-energy pulse which switches the system from low-transmission state B to high-transmission state A.

the second equation [blue straight line in Fig. 3(c)]. The graphical solution is given by the points where both curves cross, and for a sufficiently high input intensity we obtain three crossings A, B, and C, of which only A and B correspond to stable solutions.

The periodicity of the layers and the one-dimensional geometry considered here are not required for the observed effects. We numerically found that random variation of the distances between the layers below 10% does not influence the transmission curve (not shown). We have also considered a 2D system of rods with diameters and distances between them much smaller than the wavelength and observed a bistability similar to that in Fig. 3 (not shown). The predicted effects can also be observed at other wavelengths with corresponding adjustment of the layer thicknesses in order to reach $|\varepsilon_{\rm eff}^L| \ll 1$, $\varepsilon_{\rm eff}^L < 0$, although far from the optimum around 600 nm the loss of silver can become too high.

The transient response of the considered system is determined not by the response time of the bulk silver nonlinearity but by the feedback of the multilayer structure. To determine the switching time, a cw signal is launched into the composite with an input intensity of 2.55 GW/cm^2, which is 1% below the higher edge of the bistability range in Fig. 3(a). At $t \sim 1.8$ ps, a second pulse (shown by the green curve along the lower axis in Fig. 4) with a FWHM duration of 70 fs, a peak intensity of 50 MW/cm^2 (2% of the cw field intensity), and a central wavelength of 600 nm enters the medium. As a result, switching to the A state occurs with a transition time on the order of 1 ps. The high loss of the structure leads to high requirements in respect to the modification of the dielectric constant, but it also has the advantageous effect of allowing ultrafast (sub-ps-range) response. Such fast operation is hardly possible in systems based on cavities with quality factors on the order of 10^3–10^4. Previous studies on the ablations threshold for silver in the picosecond regime [23–25] provide values of about 0.5 J/cm^2 and above, only weakly depending on the pulse duration. Taking into account the film thickness of the studied structure [25], threshold values of roughly 0.2 J/cm^2 and above can be estimated. For an intensity of 2 GW/cm^2, fluences in the range from 0.01 to 0.1 J/cm^2 correspond to pulse durations from 5 to 50 ps, which are larger than the response time. Therefore, ablation of the silver films will not be critical for the considered parameters.

In conclusion, the transmission of a metal-dielectric multilayer structure with a linear effective dielectric constant slightly below zero is studied. The predicted steplike dependence of the output intensity on the input intensity is connected with the intensity-dependent sign of the effective dielectric constant and a transition from metalliclike to dielectriclike properties of the composite. The highly nonlinear behavior of the proposed metal-dielectric structure can find applications, for example, in all-optical information processing as well as in pulse cleaning for high-power chirped-pulse amplification.

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