Accessing the optical nonlinearity of metals with metal–dielectric photonic bandgap structures

Ryan S. Bennink, Young-Kwon Yoon, and Robert W. Boyd

Institute of Optics, University of Rochester, Rochester, New York 14627

J. E. Sipe

Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada

Received June 11, 1999

Metals typically have very large nonlinear susceptibilities ($\sim 10^6$ times larger than those of typical dielectrics), but because they are nearly opaque their nonlinear properties are effectively inaccessible. We demonstrate numerically that a multilayer metal–dielectric structure in which the metal is the dominant nonlinear [$\chi^{(3)}$] material can have much larger intensity-dependent changes in the complex amplitude of the transmitted beam than a bulk sample containing the same thickness of metal. For 80 nm of copper the magnitude of the nonlinear phase shift is predicted to be as much as 40 times larger for the layered copper–silica sample, and the transmission is also greatly increased. The effective nonlinear refractive-index coefficient n_2 of this composite material can be as large as $(3 + 6i) \times 10^{-9}$ cm^2/W, which is among the largest values for known, reasonably transmissive materials.

OCIS codes: 190.0190, 190.4400.

Materials with strong optical nonlinearities hold great promise for science and technology. Because metals typically have nonlinear susceptibilities that are orders of magnitude larger than those of dielectrics, they hold promise as nonlinear optical materials. Their large attenuation constants, however, render even samples that are just tens of nanometers thick essentially opaque. As a result, much research has been directed toward metal–dielectric composite materials.[1–6] Such studies have considered effective-medium composites, either in Maxwell Garnett[7] or in Bruggeman[8] geometries, in which local-field effects enhance the nonlinear polarization while keeping the absorption at an acceptable level. An alternative approach, proposed in this Letter, is to use a layered metal–dielectric geometry in which a photonic band resonance both maximizes the transmission and increases the nonlinear phase shift.[9]

A distinction crucial to our approach is that the attenuation constant of a medium is proportional to the imaginary part of the refractive index, whereas the absorption constant is proportional to the imaginary part of the dielectric constant.[10] Since the imaginary part of the refractive index in a metal is typically larger than the imaginary part of the dielectric constant, the large attenuation of light is due more to reradiation in the backward direction than to absorption. If this light could be redirected in the forward direction (say, through Bragg reflection), it should be possible to greatly increase the transmission of the sample. The concept of using photonic bandgap (PBG) structures[11] to greatly increase the linear transmittance of a given thickness of metal was recently demonstrated by Bloemer and co-workers.[12,13] In this Letter we apply this concept to the design of composite nonlinear materials and show numerically that the increased transmission afforded by layered metal–dielectric structures can lead to intensity-dependent changes in the complex amplitude of the transmitted beam that are very much larger than in a bulk sample containing the same thickness of metal.

Because exact analytical solutions to the wave equation in inhomogeneous media are not known for the general case, we integrate Maxwell's equations for $\mathbf{E}$ and $\mathbf{H}$ numerically. We consider a plane wave propagating in the z direction, normally incident on a multilayer structure [Fig. 1(a)]. We choose the field to be polarized in the x direction so that $\mathbf{E} = E(z)\hat{\mathbf{x}}$ and $\mathbf{H} = H(z)\hat{\mathbf{y}}$. Then, for a nonmagnetic material with

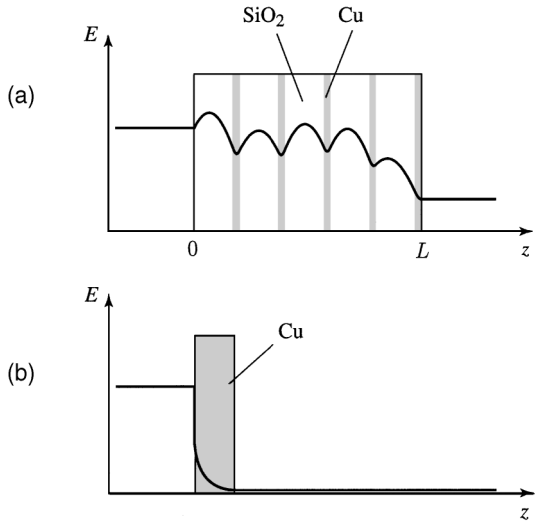

Fig. 1. Cross sections of (a) a silica and copper PBG structure and (b) a bulk sample of copper, with representative distributions of the electric field superimposed. The particular distribution shown in (a) corresponds to a sample with copper and silica layer thicknesses of 16 and 100 nm, respectively, at a wavelength of 650 nm.

0146-9592/99/201416-03$15.00/0

a $\chi^{(3)}$ nonlinearity, the relevant equations (in Gaussian cgs units) are

$$dE/dz = ikH\,, \tag{1}$$

$$dH/dz = ik[\epsilon_{\mathrm{lin}}(z) + 12\pi\chi^{(3)}_{1111}(z)|E|^2]E\,, \tag{2}$$

where $k = \omega/c$ and ϵ_{lin} is the linear dielectric constant. The integration process automatically ensures the continuity of the electric and the magnetic fields across layer boundaries, so that only the external boundary conditions need be considered. Provided that the external medium is linear, these conditions are

$$E_i = \frac{1}{2}[E(0) + H(0)/n_0], \tag{3}$$

$$E_r = \frac{1}{2}[E(0) - H(0)/n_0], \tag{4}$$

$$E_t = \frac{1}{2}[E(L) + H(L)/n_0], \tag{5}$$

$$0 = \frac{1}{2}[E(L) - H(L)/n_0], \tag{6}$$

where n_0 is the refractive index of the external medium, E_i is the complex amplitude of the incident light, E_r is the amplitude of the reflected light, and E_t is the amplitude of the transmitted light. Integrating Eqs. (1) and (2) backward from $z = L$ to $z = 0$ yields E_i and E_r as a function of E_t, from which the reflection and transmission coefficients, $r = E_r/E_i$ and $t = E_t/E_i$, respectively, are easily determined.

Figure 2(a) shows the calculated linear transmission properties of a sample consisting of five identical pairs of alternating copper and silica layers at $\lambda = 650$ nm. Copper was chosen because the imaginary part of its permittivity reaches a relatively small value in the visible and because its cubic nonlinearity is orders of magnitude larger than that of other metals commonly used in studies of metal–dielectric composites. At this wavelength the (linear) refractive indices of copper[14] and silica[15] are $n_{\mathrm{Cu}} = 0.216 + 3.64i$ and $n_{\mathrm{SiO_2}} = 1.456$. Our results are normalized to be independent of the value of $\chi^{(3)}$; it was assumed only that the nonlinearity of silica is negligible compared with that of copper. The transmission is plotted as a function of $d_{\mathrm{SiO_2}}$, the thickness of the silica layers, when the copper layers each have a thickness $d_{\mathrm{Cu}} = 16$ nm. One finds a series of transmission resonances between regions of negligible transmission, which are the stop bands. A striking feature is that the transmission becomes as large as 50%, whereas in a bulk copper sample of the same thickness (80 nm) one would expect a transmission $T \sim \exp[-2(\mathrm{Im}\, n_{\mathrm{Cu}}) \times 2\pi/\lambda \times 80\ \mathrm{nm}] = 0.3\%$. The field distribution within the multilayer sample (with $d_{\mathrm{Cu}} = 16$ nm and $d_{\mathrm{SiO_2}} = 100$ nm) is displayed in Fig. 1(a). Whereas in the bulk sample [Fig. 1(b)] the magnitude of the field exhibits a rapid exponential decrease, in the multilayer sample the field remains at a significant fraction of its incident amplitude.

To make effective use of the nonlinearity of the metal one should maximize the field strength in the metal layers. But, in general, increasing the field strength in the metal tends to decrease the transmission, since more energy passes through the absorbing material. Thus for an optimally tuned structure some trade-off exists between transmission and effective nonlinear response. As an additional consideration, the sensitivity of the output to variations in the phases and amplitudes of the internal fields will vary greatly as the device is tuned through resonance. Our studies showed that local maxima in transmission, average metal-layer field strength, and phase sensitivity all occurred at similar values of $d_{\mathrm{SiO_2}}$ (Fig. 2). In Fig. 2(b), $\langle|E_{\mathrm{Cu}}|^2\rangle$ is a spatial average of the field over the metal layers and

$$F \equiv \frac{\Delta\phi_{\mathrm{pbg}}}{\frac{2\pi}{\lambda}\int_0^L \Delta n\, dz} \tag{7}$$

is the phase sensitivity, defined as the ratio of the phase change at the output to the phase change that one would expect from the change in optical path length alone. Here $\Delta n(z)$ is the intensity-dependent change in refractive index of the metal layers.

To demonstrate the advantage of the PBG structure over a bulk sample consisting of an equal thickness (80 nm) of copper we take the ratio $\Delta\phi_{\mathrm{pbg}}/\Delta\phi_{\mathrm{bulk}}$ as a figure of merit, where $\Delta\phi$ is the intensity-dependent change in the complex phase of the transmitted beam. The complex phase ϕ is defined in terms of the transmission coefficient given above by $t \equiv \exp(i\phi)$. Figure 3 shows the magnitude of $\Delta\phi_{\mathrm{pbg}}/\Delta\phi_{\mathrm{bulk}}$ as a function of the PBG silica-layer thickness. Comparing Figs. 2 and 3, one can see that the nonlinear phase shift of the layered sample greatly exceeds that of the bulk sample when the layered sample is tuned to a transmission resonance. The greatest enhancement occurs near the band edge ($d_{\mathrm{SiO_2}} = 98$ nm), for which the intensity transmission $|t_{\mathrm{pbg}}|^2 = 12\%$ and the

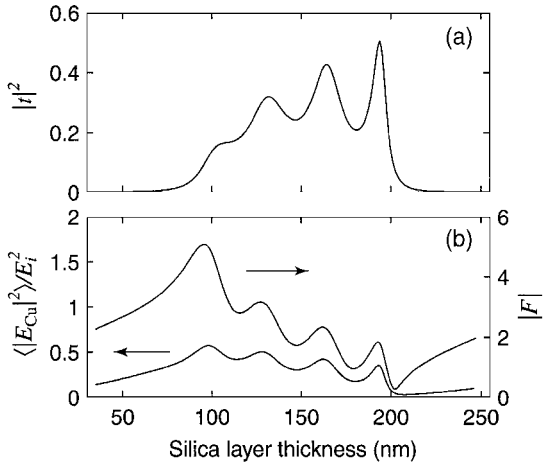

Fig. 2. (a) Linear transmission spectrum, as a function of silica layer thickness, of the silica–copper PBG structure, with $d_{\mathrm{Cu}} = 16$ nm and $\lambda = 650$ nm. (b) Average of the squared field strength in the metal layers (normalized to the incident field), along with the phase sensitivity F [defined in Eq. (7)].

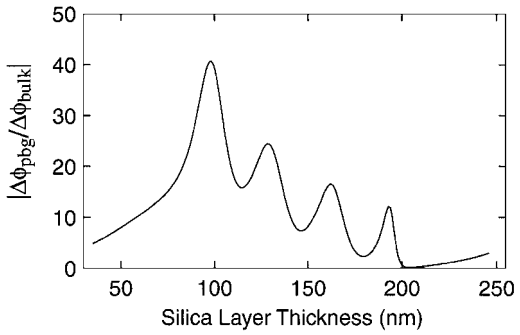

Fig. 3. Ratio of the nonlinear phase shift of the PBG structure to that of a bulk copper sample, under the same conditions as in Fig. 2.

magnitude of the nonlinear phase shift is more than 40 times larger than that of the bulk sample. We note that $\Delta\phi_{\mathrm{pbg}}/\Delta\phi_{\mathrm{bulk}}$ is approximately proportional to the product of the two functions plotted in Fig. 2(b). This relationship makes sense, because to first order in the nonlinearity, $\Delta\phi_{\mathrm{pbg}}$ is proportional to the product of $\chi^{(3)}|E_{\mathrm{Cu}}|^2$ (through Δn) and F [cf. Eq. (7)]. Since F is complex, the phase of the effective nonlinearity of the structure can be different from the phase of the intrinsic nonlinearity of the metal. This phase difference generally varies with tuning, and consequently we can adjust it to minimize nonlinear attenuation.

In photonics applications the net phase shift that one can achieve with a reasonable intensity of light is the primary quantity of interest. With this in mind, we define the effective second-order refractive index $n_{2\mathrm{eff}}$ for the PBG structure as

$$\Delta\phi_{\mathrm{pbg}} = \frac{2\pi}{\lambda} n_{2\mathrm{eff}} I L , \tag{8}$$

where I is the incident intensity. We now suppose that the $\chi^{(3)}$ of copper is imaginary with a reported magnitude[4] $|\chi^{(3)}| \approx 1.5 \times 10^{-6}$ (esu), which corresponds to $|n_2| \approx 7.5 \times 10^{-8}\ \mathrm{cm}^2/\mathrm{W}$. With this supposition, $n_{2\mathrm{eff}} = (3 + 6i) \times 10^{-9}\ \mathrm{cm}^2/\mathrm{W}$ at the point of greatest enhancement, a value whose magnitude is smaller than that of bulk copper but is unprecedentedly large for a reasonably transmissive material. The real part of this quantity corresponds to a phase shift of $\sim 5\pi$ per $\mathrm{GW/cm}^2$ of incident intensity. Thus PBG structures can improve the optical properties of metals to the point where optical limiting, bistability, and switching become possible.

In conclusion, we have shown that a metal–dielectric multilayer PBG structure can exhibit extremely large nonlinearities while remaining reasonably transmissive. Our numerical studies revealed that for the particular five-layer-pair structure discussed in this Letter one could increase the transmittance by much more than an order of magnitude and achieve nonlinear phase shifts as much as 40 times larger than with a bulk sample consisting of the same thickness of metal. Based on this effect, we believe that metal–dielectric PBG composite materials could be used in possible new classes of nonlinear optical devices.

The authors gratefully acknowledge useful discussions with C. M. Bowden and R. L. Nelson. The portion of this work conducted at the University of Rochester was supported by U.S. Air Force Office of Scientific Research grant F49620-00-1-0061 and by the sponsors of the university's Center for Electronic Imaging Systems. R. W. Boyd's e-mail address is boyd@optics.rochester.edu.

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