10-4-2007

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# Passive all-optical diode using asymmetric nonlinear absorption

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Saturable and reverse saturable absorptions are well-known phenomena, originating from the imaginary component of the third order nonlinear optical susceptibility. We note that structures with an axially asymmetric nonlinear absorption can be easily realized from saturable and reverse saturable absorption materials arranged in tandem. In this paper, the basic transmission behavior of such a structure is worked out. Detailed numerical simulations demonstrate passive all-optical diode behavior, and the results are verified experimentally. The principle will work for all light polarizations, has no phase-matching restrictions, and can be extended to a large number of available nonlinear media for possible applications. © 2007 American Institute of Physics.
[DOI: 10.1063/1.2794015]

Spatially nonreciprocal passive devices that allow unidirectional propagation of an optical signal are currently receiving much attention as an alternative to the conventional optical isolator, which is based on linear polarizers and a magneto-optical Faraday rotator. These "optical diodes" include fluorescent dyes with a concentration gradient,[1] absorbing multilayer systems,[2] and second harmonic generators with a spatially varying wavevector mismatch.[3] However, most reported optical diodes are generally based on photonic crystals, which are dielectric or metallodielectric structures, with a spatial periodicity in their refractive index. This periodicity affects the propagation of electromagnetic waves in the same way as the periodic potential in a semiconductor crystal affects electron motion by defining allowed and forbidden electronic energy bands. The absence of allowed propagating electromagnetic modes in a range of wavelengths gives rise to a host of optical phenomena such as inhibition of spontaneous emission, high reflectivity, and low-loss waveguiding. Such photonic bandgaps have also been suggested for optical diode applications. For example, a one-dimensional photonic crystal structure having a nonlinear optical medium response[4,5] and a two-dimensional photonic crystal waveguide with an asymmetric array of nonlinear defect rods[6] are found to display nonreciprocal effects. An electrotunable optical diode based on liquid-crystal heterojunctions, showing nonreciprocal transmission of circularly polarized light at the photonic bandgap regions, has been reported.[7] In another configuration using liquid crystals, linearly polarized light is used.[8] However, since the basic physical phenomenon is based on diffraction, the periodicity of the photonic crystal structure has to be in the same length scale as half the wavelength of the electromagnetic waves used. For photonic crystals operating in the visible part of the spectrum, this would be approximately 200–350 nm, which makes the fabrication cumbersome and complicated.

In this context, it would be interesting to investigate the transmission of optical pulses through structures in which the longitudinal nonlinear absorption coefficient, rather than the linear or nonlinear refractive index, is varied in some fashion. One motivation for this enquiry is the fact that the fabrication of such a structure is much easier compared to that of a photonic crystal. It can be intuitively argued that the spatial variations in the nonlinear absorption should lead to spectacular transmission effects, the nature of which can be calculated using appropriate propagation equations. Indeed, from numerical simulations, here we show that it is possible to obtain a high contrast, passive nonreciprocal transmission from a simple structure having a single asymmetry in the nonlinear absorption coefficient. It turns out that the layer thickness used can be much larger than the light wavelength.

In practice, the simplest form of asymmetry would be a sharp step in the nonlinearity, which can be achieved by placing a layer of saturable absorber material adjacent to a layer of reverse saturable absorber material. Photons incident on an absorbing material will transfer electrons from the ground state to an upper energy state, thereby depleting the ground state population. If the absorption cross section of the excited state is smaller than that of the ground state, the transmission of the system will be increased at high intensities, making it a saturable absorber (SA). On the other hand, if the absorption cross section of the excited state is larger than that of the ground state, then the system is less transmissive at high intensities, and it is called a reverse saturable absorber (RSA). In general, two-photon and multiphoton absorptions can also contribute to reduced transmission at high intensities. Our simulations show that if a SA is placed in tandem with a RSA, in that order (called the forward bias configuration), the light transmission can be enhanced at higher intensities. If the order is changed (i.e., RSA followed by SA, called the reverse bias configuration), then the transmission can be attenuated. Results are verified from nonlinear transmission measurements carried out in a semiconductor doped glass–copper phthalocyanine (CuPc) solution combination. Optical nonlinearities in semiconductor doped glasses[9] and metal phthalocyanines are well known. At the

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excitation wavelength of 532 nm, the glass we used is a strong saturable absorber, while CuPc is a reverse saturable absorber due to its large excited state absorption cross section.[10] A structure of this kind has the advantages that it is all-optical and polarization independent and can withstand high optical intensities. Furthermore, its organization is extremely simple.

The nonlinear absorption coefficient of a SA is given by[11]

$$\alpha_{SA}(I) = \alpha_{0SA}/(1 + I/I_s), \qquad (1)$$

where $\alpha_{0SA}$ is the linear absorption coefficient, $I$ is the intensity within the medium, and $I_s$ is the saturation intensity (intensity where the absorption drops to half its original value). The corresponding propagation equation can be written as

$$\frac{dI}{dz'} = -\left(\frac{\alpha_{0SA}}{1 + I/I_s}\right)I, \qquad (2)$$

where $z'$ is the position coordinate along the axis of propagation in the medium (of length $L$) and $I$ is $I(z',t)$. Equation (2) can be solved to give the transmission of a two level saturable absorber at steady state, in the homogeneously broadened case, as

$$T = \exp(-\alpha_{0SA}L)\exp[(I_{in}/I_s^\Delta)(1 - T)], \qquad (3)$$

where $I_s^\Delta = I_s[1 + (\Delta\omega T_2)^2]$, with $\Delta\omega$ being the excitation frequency detuning and $T_2$ the phase coherence lifetime. $I_{in}(t)$ is the incident intensity at $z' = 0$.

For a RSA based on excited state absorption, the nonlinear absorption coefficient is given by

$$\alpha_{RSA}(I) = \alpha_{0RSA} + \sigma N(I)I, \qquad (4)$$

where $\alpha_{0RSA}$ is the linear absorption coefficient, $\sigma$ is the absorption cross section, and $N(I)$ is the population density of the excited state. As a generalization to include two-photon absorbers, $\sigma N(I)$ can be replaced by $\beta I$, where $\beta$ is the two-photon absorption coefficient. The corresponding propagation equation is given by

$$\frac{dI}{dz'} = -(\alpha_{0RSA} + \beta I)I, \qquad (5)$$

which, when solved for a pulsed Gaussian beam, gives the transmission as

$$T = [\exp(-\alpha_{0RSA}L)/\sqrt{\pi}q_0]\int_{-\infty}^{+\infty} \ln[\sqrt{1 + q_0\exp(-t^2)}]dt, \qquad (6)$$

where $q_0$ is given by $\beta I_0 L_{eff}$. $I_0$ is the on-axis peak intensity, and $L_{eff}$ is given by $[1 - \exp(-\alpha_{0RSA}L)]/\alpha_{0RSA}$.

In our simulation, $I_s$ and $\beta$ are taken as the nonlinear parameters. $I_s$ values are kept in the range of $10^{12}$–$10^{14}$ W/m$^2$ and $\beta$ values in the range of $10^{-11}$–$10^{-9}$ m/W, which match well with practical situations. For the forward bias simulation, a temporally Gaussian laser pulse of 3 ns full width at half maximum and energy $E_{in}$ is chosen. The intensity of the Gaussian pulse $[I_{in}(t)]$ is then propagated through the SA, and the output intensity is calculated using Eq. (2). This output intensity is then fed as the input of the RSA, and the output of the RSA $[I_{out}(t)]$ is calculated using Eq. (5). $I_{out}(t)$ is then time integrated to calculate the output energy $E_{out}$, and the transmission of the
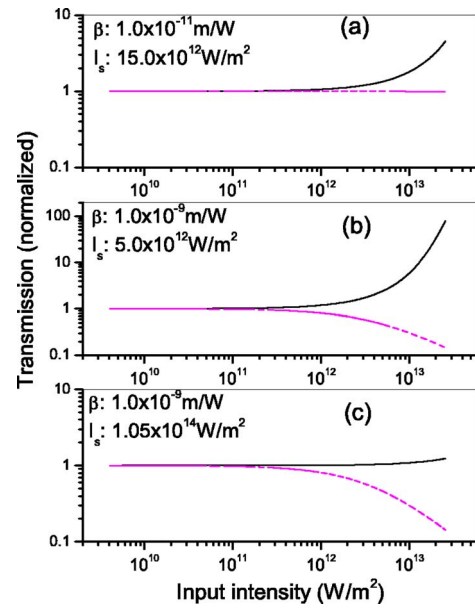


FIG. 1. (Color online) Simulated optical transmission of the system in the forward and reverse bias configurations for different nonlinear parameter values. Solid curves correspond to forward bias and dashed curves correspond to reverse bias. Plotted transmission values are normalized to the linear transmission of the system.

device is given by $E_{out}/E_{in}$. This process is repeated for intensity values within the range of $10^9$–$10^{14}$ W/m$^2$ (which can be achieved in practice from a focused Nd:yttrium aluminum garnet (YAG) laser). The same procedure is adopted for the reverse bias simulation, except that in this case Eq. (5) is solved first, which is then followed by Eq. (2).

Typical results obtained are given in Fig. 1. The nonlinear transmission of the system is plotted in the forward and reverse bias configurations for different values of $I_s$ and $\beta$. The transmission is reciprocal for excitation intensities less than $10^{11}$ W/m$^2$ in all cases, but nonreciprocity is achieved by different degrees at higher excitation intensities. For example, in Fig. 1(a), the transmission goes up in the forward bias while it remains almost a constant in the reverse bias. In Fig. 1(b), the transmission is substantially enhanced in the forward bias and attenuated in the reverse bias. In Fig. 1(c), the transmission increases slightly in the forward bias, while the reverse bias transmission is substantially reduced. Thus, by a proper choice of the nonlinear parameters, it is possible to tailor the desired nonreciprocal transmission. The maximum contrast ratios achieved can be quite large, as it reaches nearly 1000:1 in Fig. 1(b).

To verify these results experimentally, we used an orange colored glass filter of 3 mm thickness that shows high saturable absorption at 532 nm as the SA, and a diluted solution of CuPc dissolved in a polymethyl-methacrylate-chloroform base, taken in a 1 mm cuvette, as the RSA. The CuPc solution has a linear transmission of 58% at 532 nm. The light source used is a Nd:YAG laser producing laser pulses of 3 ns pulsewidth at the second harmonic wavelength of 532 nm. The output beam is spatially and temporally Gaussian. A half-wave plate and a linear polarizer are kept in the beam path before the sample. By rotating the half-wave plate, the energy reaching the sample can be controlled over a large range. To make the nonlinear transmission measurements, the transmission of the sample was measured for different angular positions of the half-wave plate.
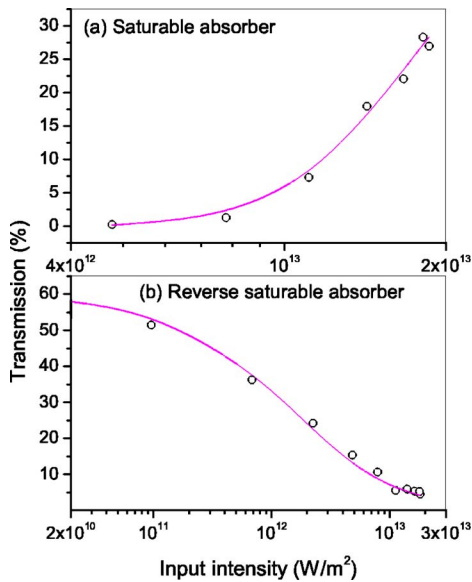
FIG. 2. (Color online) Transmission of the (a) colored glass filter (SA) and (b) Cu-phthalocyanine solution (RSA) as a function of the input intensity. Circles are experimental points, while curves are numerical fits to Eq. (2) (for SA) and Eq. (5) (for RSA). The calculated values are $I_s = 9.75 \times 10^{11}$ W/m$^2$ and $\beta = 1.1 \times 10^{-9}$ m/W.

To determine the nonlinear parameters of the SA and RSA, separate measurements were first performed. Figure 2(a) shows the strong nonlinear transmission obtained in the colored glass filter. By fitting the experimental data to Eq. (2), the best-fit $I_s$ value was calculated. Figure 2(b) shows the nonlinear transmission for the CuPc solution, and the experimental data were fitted to Eq. (5) to get the best-fit $\beta$ value. After thus obtaining both nonlinear parameters, the two materials were kept together by an adhesive tape, and the measurements were done in the forward and reverse bias configurations. The results obtained are given in Figs. 3(a) and 3(b), respectively. The $I_s$ and $\beta$ values calculated above were used for fitting these data to the corresponding equations.
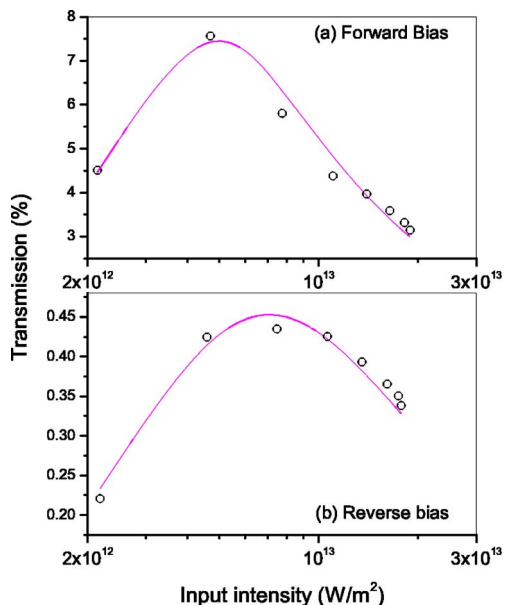


FIG. 3. (Color online) Transmission of the SA+RSA combination in the forward and reverse bias configurations. Circles are experimental points, while the curves are simulations.

Clearly, there is an order of magnitude difference between the transmissions in the forward and reverse bias conditions, and the maximum contrast ratio obtained is about 18. The curves have a different shape compared to the simulations (shown in Fig. 1). This is due to the fact that the nature of nonreciprocity is heavily dependent on the $I_s$ and $\beta$ values, which we have verified by simulating the transmission for a large number of combinations of $I_s$ and $\beta$. It should be possible to realize higher contrast ratios by a careful choice of nonlinear materials.

These results clearly demonstrate that the idea of employing the nonlinear absorption coefficient for optical nonreciprocity is practically viable. The saturable absorber and the reverse saturable absorber used in these measurements are commercially available. We subjected these materials individually and jointly to a maximum intensity of $1.8 \times 10^{13}$ W/m$^2$ several times, without observing any optical damage. There are a large number of nonlinear materials that can be used likewise, without involving any sophisticated fabrication process. By choosing highly nonlinear materials such as liquid crystals and optical fibers, it should be possible to operate the device at different wavelengths and much lower laser intensity levels.

In conclusion, we have studied the optical transmission of a medium characterized by an asymmetry in its longitudinal nonlinear absorption coefficient. It is shown numerically that a passive all-optical diode of high contrast can be realized in such structures. Experimental demonstration of nonreciprocal transmission is obtained from a simple commercial colored glass and a 1 mm cell containing copper phthalocyanine solution, which are taped together in tandem. While fabrication of photonic crystals proposed for optical diode applications in the visible region is cumbersome due to dimensional constraints, the present device does not have this limitation, since the nonlinear layers can be as thick as a few millimeters. Moreover, unlike most of the other suggested devices, it works equally well for all light polarizations. There are no phase-matching requirements, and since the effect is based on optical nonlinearity, these can withstand high optical intensities. Since saturable and reverse saturable materials are available for applications over a wide range of intensities (solution, polymer films, quantum dots, metal nanoparticles, etc.), it should be possible to develop the device as laboratory on a chip for waveguide and fiber optic applications.

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