Large nonlinear phase shifts in low-loss Al$_x$Ga$_{1-x}$As waveguides near half-gap

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We study the instantaneous nonlinear index change in Al$_{0.2}$Ga$_{0.8}$As waveguides below the two-photon absorption edge and find $\pi$ phase shifts with 80 pJ, 0.4 ps pulses at wavelengths near 1.6 $\mu$m. These large phase shifts are obtained with less than 1 dB of loss from multiphoton absorption. Our results indicate that AlGaAs waveguides, which have a mature fabrication technology, can be used as compact nonlinear elements in switching and quantum optics applications in the near-infrared. Further optimization of the waveguide geometry should result in useful nonlinear phase shifts and low losses for pulse energies approaching a picojoule.

Nonlinear optical processes in condensed matter systems are often limited by their time response, heating, or loss when they result from the excitation of charge carriers in the material. It is an advantage for many photonics applications if the nonlinear process is dominated by a refractive index change that instantaneously follows the pump field without any loss to the material. Unfortunately nonlinear index changes are inextricably bound to nonlinear losses. How does one optimize the nonlinear index change-to-loss ratio? The important practical question is, "How does one obtain a nonlinear phase shift near $\pi$ (or equivalently a high nonlinear reflection) with less than 3 dB of absorption from both linear and nonlinear processes?"

The maximum nonlinear phase shift during a pump light pulse for a third-order optical nonlinearity $n_2$ can be written

\[ \Delta \phi = 2\pi n_2 I / \lambda \]  

(1)

where $I$ is the interaction length, $I$ is the light intensity at the temporal peak of the pulse averaged over the spatial beam mode, and $\lambda$ is the wavelength. Glass fiber is clearly one material system that answers the practical question. Glass fibers have one of the lowest optical nonlinearities because of their large band gap. The third-order nonlinear susceptibility $n_2$ roughly scales inversely as the fourth power of the band-gap $E_g$. The miracle for optical fibers is that one is able to make low-loss optical fibers many kilometers in length so that despite the low nonlinearity, large phase shifts can be obtained. Large enough nonlinear phase shifts with low loss can also be achieved with second-order nonlinearities in a number of materials, however the factor of two difference in frequency between the pump and signal frequencies in this case is often a difficulty because of the complicated apparatus and phase matching of the pump and signal.

Our experiments demonstrate that by using semiconductor waveguides less than one centimeter in length one can achieve large phase shifts by operating at photon energies just below half of the band-gap energy in order to avoid two-photon absorption. The nonlinear susceptibilities in semiconductors are nearly two orders of magnitude larger than in optical fibers because of the smaller band gap and the $E_g^{-4}$ scaling. A recent analysis shows that one expects a positive peak in $n_2$ just below the half-gap associated with the two-photon absorption edge. This is a relatively broad spectral peak extending, for example, in Al$_{0.5}$Ga$_{0.5}$As from 1.6 to 2.0 $\mu$m. Half-picosecond pulses in this wavelength range propagate with negligible dispersion broadening for distances of one centimeter. High quality ridge waveguides can also be fabricated for this system with linear losses less than a few tenth of a dB/cm.

A schematic drawing of the experimental apparatus is shown in Fig. 1. A ridge waveguide is formed on a 2.55 $\mu$m thick layer of Al$_{0.2}$Ga$_{0.8}$As. We choose this material composition so that the 1.6 $\mu$m wavelength excitation lies 100 meV below the half-gap energy, sufficiently far below to avoid two-photon absorption from all frequency components of the pulse. The 1.6 $\mu$m wavelength was chosen because of the available pulsed laser source and its proximity to wavelengths of interest for communications and switching applications. Guiding is provided by a 2.55 $\mu$m underlying layer of Al$_{0.5}$Ga$_{0.5}$As whose refractive index is 0.15 less than the actively layer. A MOCVD growth process was used to form this layered structure with sources and vacuum conditions chosen to minimize oxygen impurities that result in midgap states that can cause pump induced absorption. The ridge height is 0.7 $\mu$m, its width is 4.5 $\mu$m and the waveguide length is 7.1 mm for most of the measurements reported here. These dimensions were chosen so that simple optics could be used for coupling into the guide. The index difference is small enough so that only a few modes are supported by these waveguides and careful excitation results in the lowest order mode dominating the
FIG. 1. A schematic drawing of the apparatus used to measure the nonlinear optical properties of Al$_{0.5}$Ga$_{0.5}$As waveguides. The pump beam from a mode-locked NaCl color-center is focused into the ridge waveguide (W) by a microscope objective. A portion of the pump is split off to an acousto-optic modulator (AOM) to form a signal beam at a frequency shift of 80 MHz with variable modulation amplitude at 40 kHz. A detail of the front face of the waveguide shows the 0.7 μm high and 4.5 μm wide ridge and the layered structure. Both guiding layers are 2.55 μm thick. Light from the output of the 7.1 mm waveguide is collimated by a second microscope objective, frequency analyzed with a grating spectrometer (OSA) and detected using a photodiode detector (D) and a lock-in amplifier.

transmitted light as determined by its spatial distribution. Total coupling efficiencies of nearly 45% were obtained with microscopic objectives at the input and output of the waveguide. Both input and output surfaces are antireflection coated for reflections near 1% per surface. The total loss in the input and output microscope objectives is 6%. All of the remaining loss is easily accounted for by the input and output mode coupling losses. The dominant linear loss is scattering from nonuniformities in the waveguide ridge and this loss is less than 5% for the present experiments due to a uniform etching process utilizing the orientation of the crystal axes. There is no measurable birefringence in these guides.

A passively mode-locked NaCl color center laser is used to excite the waveguides. The pulses are 360 fs full-width half-maximum in duration and are separated by 11.9 ns. The lasing wavelength is tuned between 1.5 and 1.7 μm using a birefringent tuner and different saturable absorbers. Peak powers as high as 300 W were measured at the output of the waveguide corresponding to peak intensities in the waveguide near 10 GW/cm$^2$. A probe pulse was formed by frequency shifting a portion of the input beam by 80 MHz using an acousto-optic modulator. The polarization orientation, time delay, and amplitude of the probe were varied to measure the nonlinear optical properties of the guide. The frequency spectra of the output pulses were measured with a grating spectrometer and the output pulse shapes are measured with an optical correlator.

Large nonlinear phase shifts in the waveguides are apparent in the optical spectra of the output pulses shown in Fig. 2. At phase shifts of π and larger self-phase modulation dramatically broadens the spectrum and forms a series of spectral peaks. A series of spectra corresponding to (a) the input spectrum, (b) Δφ = π, (c) Δφ = 1.5 π, and (d) Δφ = 2.5π are shown in Fig. 2. Nearly identical spectra as a function of input intensity were obtained for wavelengths of 1.69 and 1.62 μm. These spectra are expected in the presence of strong self-phase modulation due to an instantaneous nonlinearity where there is negligible dispersion, and similar spectra have previously been observed in optical fibers. Asymmetry of the spectra probably results from slight asymmetries of the input pulse shape. Phase shifts as large as 4.5π were observed at shorter pulse durations but dispersion effects associated with the broad spectral widths in this case make it difficult to interpret the results with a simple model. Similar spectra were measured for a probe beam orthogonally polarized with respect to the pump beam. The probe intensity was maintained at one tenth of the pump intensity. The effective cross-phase modulation is two-thirds of the self-phase modulation as expected for the symmetry of the crystal. The intensity required for a π phase shift is 3.1 ± 0.5 GW/cm$^2$ which yields a value of $n_2 = + 3.6 (± 0.5) \times 10^{-14}$ cm$^2$/W for self-phase modulation. The uncertainty in the measured values is primarily a result of estimating the pulse intensity in the multimode waveguide. This is in qualitative agreement with the scaling predicted near the two-photon absorption edge when compared with the measured value of $n_2 = - 4 \times 10^{-13}$ cm$^2$/W at a wavelength of 1.06 μm. The sign of $n_2$ is positive for our waveguides, as deduced from the pulse broadening at high intensities. The sign of $n_2$ changes from negative to positive as one decreases the excitation energy

FIG. 2. A series of spectra from the waveguide output are shown as a function of wavelength. All spectra are plotted on a linear scale and are normalized to a unit peak spectral intensity. At low input intensities the spectra is the Fourier transform of the input pulse shape as shown in (a). For intensities corresponding to phase shifts of approximately π (b), 1.5π (c), and 2.5π (d), the spectra broaden and develop a series of peaks.
from above to below the two-photon edge in agreement with theoretical predictions.\cite{1}

Even at the large phase shift of 2π the measured nonlinear loss for the pump was only 15%. A more accurate study of the nonlinear loss using the probe beam is shown in Fig. 3. We measure the total (pump + probe) energy loss for the light propagating through the guide by measuring the 40 kHz modulation amplitude due to the probe as the pump is turned on and off. This measurement technique eliminates the need to correct for four-wave-mixing gain effects. We fit this data to

$$\alpha_{NL} = 4\alpha_2 \langle I \rangle + 9\alpha_3 \langle I \rangle^2$$

(2)

where $\alpha_2$ is the pump two-photon absorption coefficient, $\alpha_3$ is the pump three-photon absorption coefficient, $\alpha_{NL}$ is the net nonlinear absorption coefficient for the signal beam, and the $\langle \rangle$ brackets on $I$ indicate the appropriate spatial and time averages. The fact that we measure a small signal absorption in order to obtain the pump absorption coefficients accounts for the factors of 4 and 9 in Eq. (2). Neither two-photon nor three-photon absorption alone yield a good fit to the data. The fitted values obtained are $\alpha_2 = 0.26 \times 10^{-4}$ cm/MW and $\alpha_3 = 0.004$ cm$^3$/GW$^2$. The two-photon absorption coefficient is more than two orders of magnitude below that obtained for above half-gap excitation.\cite{6} As we had hoped there is a sufficient reduction of the two-photon absorption component below half-gap to allow for our large measured phase shifts. In fact, the nonlinear absorption for phase shifts above 1.5π is dominated by three photon absorption. For example, at a phase shift of 4.5π the nonlinear absorption for the pump is 37%, largely due to three-photon processes. Our measured three-photon absorption coefficient agrees with theoretical estimates\cite{7,8} within a factor of two. We also checked for free-carrier generation by delaying the probe pulse. At phase shifts near 2π, a 5%-10% component of the absorption persisted for approximately 50–100 ps after the pump pulse. This is in order of magnitude agreement with a free-carrier absorption from carriers at densities near $10^{16}$/cm$^3$ generated by two and three photon absorption. This free carrier component should rise very rapidly with $I^3$ and ultimately lead to damage of the waveguide. However it is a negligible effect in the interesting region of phase shifts near π. Finally we checked for thermal effects with time constants of the order of milliseconds and found none. It is important for many applications\cite{9,10} to obtain energies of the order of or less than a picojoule for phase shifts in the range from 0.1π to π. For the relatively large waveguide used in our initial experiments and a length of 7.1 mm, a π phase shift requires nearly 80 pJ. Reduction of the waveguide dimensions should allow one to achieve a π phase shift with pulse energies near 10 pJ. For some interesting switching applications a phase shift of only 0.1π is sufficient.\cite{9,10} Waveguides based on the results reported here seem quite promising for these applications. Increasing the band gap of the waveguide will only decrease $n_2$, and increase $\alpha_3$ until one reaches one-third of the gap where three-photon absorption will decrease sharply and $n_2$ will have decreased by roughly a factor of four due to the $E_0^{-4}$ scaling. This is probability another good operating point for obtainable lengths of semiconductor waveguide.

In summary, we have demonstrated an operating region for semiconductor waveguides where interesting nonlinear phase shifts can be obtained with moderate pulse energies and low loss. These phase shifts are “ultrafast,” i.e., they respond at the optical pump frequency. The nonlinear response is broadband so that the pump frequency selection and stability are not critical. These waveguides can be incorporated into a number of interesting optical switches.\cite{8,10} In a fundamental sense it is interesting to verify that multiphoton absorption is the limiting process for nonlinear phase shifts in semiconductors and that there are optimum operating regions just below each multiphoton absorption edge.

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