Title: Semiconductor-Doped Glasses for Nonlinear Integrated Optics (Unclassified)

The properties of all-optical switching devices, as applied to nonlinear directional couplers in semiconductor-doped CdS \textsubscript{x}Se \textsubscript{1-x} glasses were investigated. This program achieved five distinct goals:

1. The operating characteristics of, and material requirements for, a variety of all-optical switching devices were investigated theoretically and defined for the first time. The devices included nonlinear directional couplers, Mach-Zehnder interferometers, and distributed feedback gratings; the nonlinear phase shifts required for device switching operation were found to be $4\pi \approx 2\pi$ and $\pi$, respectively. A material figure of merit, $W = \Delta n \lambda / 2\alpha$, was defined for various devices.

2. Parameters for forming ion-exchange waveguides in CdSSe-doped glasses and color filter glasses were evaluated. Single-mode channel waveguides and directional couplers were designed, fabricated, and tested in these materials.

3. The non-linear properties, the intensity-dependent refractive index, and absorption changes and their time evolution in channel waveguides in the semiconductor-doped glasses were evaluated experimentally, using a novel pump-probe hybrid Mach-Zehnder interferometer, and compared with the predictions of plasma theory. The relative contributions of the electronic and thermal nonlinearities were measured.

4. All-optical switching in a nonlinear directional coupler was demonstrated experimentally with a 20-ps turn-off time for the first time. Thirty percent switching was obtained, in excellent agreement with theory.

5. Figures of merit for a series of semiconductor materials were calculated, based on the solid-state properties of the materials.
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ABSTRACT

The properties of all-optical switching devices, as applied to nonlinear directional couplers in semiconductor-doped CdS_xSe_{1-x} glasses were investigated. This program achieved five distinct goals:

1. The operating characteristics of, and material requirements for, a variety of all-optical switching devices were investigated theoretically and defined for the first time. The devices included nonlinear directional couplers, Mach-Zehnder interferometers, and distributed feedback gratings; it was found that the nonlinear phase shifts required for device switching operation were 4\pi, 2\pi, and \pi, respectively. A material figure of merit, W = \Delta n_{\text{sat}}/\alpha\lambda was defined for various devices.

2. The parameters for forming ion-exchange waveguides in CdSSe-doped glasses and color filter glasses were evaluated. Single-mode channel waveguides and directional couplers were designed, fabricated, and tested in these materials.

3. The nonlinear properties, the intensity-dependent refractive index, and absorption changes and their time evolution in channel waveguides in the semiconductor-doped glasses were evaluated experimentally, using a novel pump-probe hybrid Mach-Zehnder interferometer, and compared with the predictions of plasma theory. The relative contributions of the electronic and thermal nonlinearities were measured.

4. All-optical switching in a nonlinear directional coupler was demonstrated experimentally with a 20-ps turn-off time for the first time. Thirty percent switching was obtained, in excellent agreement with theory.

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INTRODUCTION

All-optical devices have the potential for performing switching and logic operations on picosecond and sub-picosecond time scales. Such operations can be used in the manipulation of information in ultrafast data systems, coding and decoding of information, and so forth. At the start of this program, no such device had been implemented, the operating characteristics were unknown, and the material requirements for such devices were not known.

The goal of this program was to fabricate ultrafast, nonlinear guided-wave devices for all-optical signal processing using semiconductor-doped glasses (SCDG). This original choice of material was based on:

1. Some original four-wave mixing measurements of the nonlinearities by Jain and Lind [JOSA 73, 647 (1983)] that indicated usefully large nonlinearities and sub-nanosecond nonlinearity turn-off times;
2. The author's own four-wave mixing and fluorescence lifetime measurements which indicated nonlinearity turn-off times in the tens of picoseconds;
3. The fact that the material was readily available in the form of color filter glasses;
4. The sodium content of the host glass, which made the fabrication of waveguides by ion exchange with potassium feasible.

Any interferometric or mode-coupled integrated optics device can be made into an all-optical device by incorporating a nonlinear material in one or more of the waveguides. A nonlinear directional coupler was chosen for study because it is a four-port device with two input and two output ports. The fraction of the input power appearing in each of the output channels can be controlled by varying the input power into one or both of the channels.

This research program covered three years and a great deal of progress was made, not only toward the specific goal of ultrafast all-optical switching waveguide devices in semiconductor-doped glasses, but also toward the general understanding of such all-optical devices. Some of the early conclusions led to experiments in optical fibers and the development of nonlinear organic materials as attractive alternatives. The short-term goals of this research program evolved as new issues were raised. In order to supplement our own expertise and capabilities, this program included many collaborations, principally with Professor C. N. Ironside of the University of Glasgow, and also with Douglas Hall of Corning Glass, Lars Thylen of Ericsson (Sweden), S. Trillo and S. Wabnitz of Fonazione Ugo Bordoni, and A. C. Walker of Heriot-Watt University.
The program evolved as follows (technical details will be discussed in succeeding sections):

1. Ion-exchange waveguides were fabricated in special Schott glasses with high sodium content.
2. Short directional couplers were fabricated and their linear properties tested. No all-optical switching was observed.
3. The existence of nonlinearities in ion-exchanged glasses was established by degenerate four-wave mixing.
4. The effects of saturation in the nonlinear index change on the performance of a nonlinear directional coupler were analyzed and found to be critical. The effects on a large range of other all-optical guided-wave switching devices were also examined theoretically.
5. A hybrid Mach-Zehnder interferometer was built for measuring the real and imaginary parts of the third-order nonlinearity and its evolution in time in channel waveguides.
6. The nonlinear properties of channel waveguides in CdSSe-doped glasses were evaluated, including both the thermal and electronic contributions to the nonlinearity.
7. New nonlinear directional couplers were designed and all-optical switching demonstrated.
8. Calculations were performed on a number of semiconductor materials to evaluate their suitability for guided-wave all-optical switching.

The results of this program allowed optimum materials for all-optical switching devices to be identified; their testing and device implementation is the subject of a renewal proposal.

The following sections describe in more detail the results of this program.

WAVEGUIDE FABRICATION

The first goal was to fabricate high-quality waveguides in semiconductor-doped glasses. Slab waveguides were made by exchanging the sodium for potassium in the surface region of the color filter glass by immersing the glass into molten potassium nitrate. The sodium content of the commercially available Schott color filters was found to be too small to make waveguides easily via ion exchange with potassium. As a result, special melts were ordered from Schott with a sodium content of approximately 10% to 12%. The surfaces were first polished to optical tolerances (± λ/10) using standard techniques. The ion exchange of the sodium in the color
filter slabs in potassium nitrate solution has been performed at temperatures of about 200°C, just above the melting point of the potassium nitrate. No damage in the waveguide surface was observed with an optical microscope, and propagation losses of less than 0.2 db/cm have been measured in the best samples at the HeNe wavelength of 0.633 μm (far from the semiconductor bandgap resonance). A series of multimode samples were fabricated and the mode coupling angles were measured for both TE and TM polarizations. These experiments allowed measurement of the diffusion parameters for sodium-potassium exchange, as well as the small anisotropy produced in the refractive index during the exchange process. Further, using the inverse WKB method we were able to determine the refractive index depth profile created by the diffusion process. The results are summarized in Publications 4, 14, and 28.

Channel waveguides and dual directional couplers then were fabricated in these glasses, based on the best estimates for the waveguide and diffusion parameters, as discussed above. Assuming that the diffusion is isotropic, single-mode channel waveguides were designed, the appropriate masks built (at the University of Glasgow), and channel waveguides fabricated by ion exchange through the masks. The channel waveguide propagation losses were quite low, the best again having an attenuation of less than a 1 dB/cm. No significant depolarization of the light on transmission through the waveguides was observed. Using a computer model developed at the University of Glasgow (UG), directional couplers were designed with coupling lengths varying from one to a few centimeters. The procedure relies on calculating the lowest-order symmetric and antisymmetric modes of the two parallel waveguide structures, based on the previously measured diffusion profiles. The coupling length is inversely proportional to the difference in the propagation constants between the two modes. The required masks were fabricated at UG and directional couplers were made both at UG and the University of Arizona (UA). The experimentally measured coupling lengths were in good agreement with the theoretical predictions.

High-power laser beams with wavelengths near the bandgap of the semiconductor crystallites were propagated in the waveguides and a damage threshold of 100 MW/cm² was established for nanosecond pulses. This increases to a few gigawatts per square centimeter for picosecond pulses.

We were also able to make high-quality waveguides in the Corning glasses (somewhat to our surprise) by sodium-potassium exchange, although the diffusion parameters were substantially different than found previously for the Schott glasses. The initial sodium concentration was low in these glasses, typically 4% to 5%, which
required exchange times in the tens of hours to obtain even single mode waveguides. Because of these long times and the difficulty in controlling the processing, only the Schott glasses were used in the channel waveguide and directional coupler experiments.

**INITIAL EVALUATION OF NONLINEAR WAVEGUIDE PROPERTIES**

When we attempted to observe power-dependent changes in the transfer coefficient in the directional coupler, no power-dependent change in the output of the two channels was observed. This result could be interpreted as an absence of the nonlinearity inside the waveguide. However, the nonlinearity in the bulk glass was confirmed by degenerate four-wave mixing. Furthermore, it was confirmed by four-wave mixing that the heat cycling of the glass involved in the ion-exchange process does not affect the nonlinearity of the bulk glass. Finally, ESCA studies of the glass surface before and after the ion-exchange process revealed that the selenium which is bonded to other constituents (versus dissolved) had changed. Since the wavelength of the bandgap (and hence the optimum nonlinearity) depends on the sulphur to selenium ratio in the crystallites, it was initially concluded that the wavelength dependence of the nonlinearity is different in the bulk and waveguide regions.

Further experiments were carried out on ion-exchanged samples. A shard (approximately 20-μm thick) of the semiconductor-doped glass was ion exchanged from both sides. Based on our previous investigation of the ion-exchange parameters, the process required a few days to obtain exchange throughout the volume of the shard. Degenerate four-wave mixing experiments were performed on both the ion-exchanged shard and a non-ion-exchanged control sample. The same signal was obtained from both to within the experimental error. This experiment clearly demonstrated the presence of nonlinearity in the ion-exchanged waveguides. The relaxation time of the four-wave mixing signal was less than the laser pulse width of approximately 15 ns.

Other tests were performed to establish the presence of a nonlinearity in the ion-exchanged regions. We had previously developed a technique for studying nonlinearities in waveguides by measuring the efficiency of the process by which an external radiation field is coupled into a planar waveguide. Furthermore, by monitoring the shape of the output pulses for pulsed laser inputs, the nature of the nonlinearity (thermal versus Kerr-law) could be established. This technique was applied to the ion-exchanged waveguides. For 15-ns laser pulses, the pulse profiles were indicative of a slow, thermal, integrating nonlinearity. When the experiments were repeated with picosecond pulses, the nonlinear response also exhibited features
associated with electronic nonlinearities, band filling in this case. These experiments established conclusively that there were electronic nonlinearities in the waveguide region of the Schott samples.

The nonlinear prism coupling technique was also applied to the the new Corning ion-exchanged waveguides using mode-locked, Q-switched, picosecond pulse trains, with pulse widths of approximately 100 ps. Interference effects between the thermal (slow) and electronic (fast) nonlinearities were observed, proving that the nonlinearities were also present in the Corning waveguides.

At this stage, there were clear indications that the nonlinearities associated with the color filter glasses were present in the ion-exchange waveguides. However, no evidence of switching was found in the directional couplers.

EFFECTS OF SATURATION ON NONLINEAR GUIDED-WAVE DEVICES

While on sabbatical with our collaborators in the U.K., the author raised the question of saturation effects in nonlinear waveguides as the cause of the negative result in the directional couplers. That is, what happens if there is a limit to how much the refractive index of a material can be changed by the application of an intense light beam? The saturation power required and the maximum change in index depends on the physical mechanism which is responsible for the nonlinearity in the first place. This question is important because a nonlinear phase change of $4\pi$ is required for a nonlinear directional coupler to switch between two channels.

A simple model was assumed for the nonlinearity and the response of the directional coupler was calculated using coupled-mode theory. A two-level model was used, which includes a saturating index change and a bleachable absorption. We found numerically that the quantity $w = \Delta n_{sat}/L/\lambda$ must be greater than two for efficient switching to occur, where $L$ is the length of the device and $\Delta n_{sat}$ is the maximum change in refractive index. For $w < 1$, essentially no all-optical switching occurs at any input optical power. Absorption is of course important because it limits the useful length of a device and its throughput, leading to the definition of a materials figure of merit, $W = \Delta n_{sat}/\alpha \lambda$. For more than 80% throughput, a value $W > 10$ is needed for a nonlinear directional coupler.

The conclusion from these calculations was that saturation effects could explain all of the results to date, namely the four-wave mixing results, the null device results, and the nonlinear prism coupling results. Based on the previous experiments of Jain and Lind, the saturation value of index appears to be on the order of $10^{-4}$. For four-wave mixing, the signal did not disappear when it saturated. For the nonlinear prism coupling experiment, the fast electronic nonlinearity saturates out...
very quickly for the small beam radii used (< 100 µm), leaving only the thermal nonlinearity at higher powers, as observed. For our devices, which are nominally 5-mm long, preliminary calculations showed that no measurable switching should occur. In the nonlinear directional couplers used to date, both channels saturated before a useful nonlinear phase shift was obtained and the nonlinear directional coupler reverted to a linear directional coupler.

More detailed calculations of the effects of saturation were made. Interesting phenomena emerged. First, the number of discrete switching powers doubles in the presence of saturation for a one-beat-length coupler. Second, there are two dimensionless parameters that describe the operating characteristics of this device and there is a trade-off between saturation and absorption. Although the most detailed calculations have been made for media consisting of saturating two-level atoms, the general conclusions are valid for all absorptive nonlinearities, specifically for the semiconductor-doped glasses once the absorption and saturation index are known at a given wavelength.

Other potential all-optical switching devices were also studied. The key question was what value for the figure of merit is necessary for all-optical switching in devices such as nonlinear Mach-Zehnder interferometers or nonlinear distributed feedback gratings. Detailed analyses showed that the minimum value is different for each device. In fact, the nonlinear distributed feedback grating requires a value for W of 0.5, where the Mach-Zehnder needs a minimum value of approximately 1.0.

Degenerate four-wave mixing studies on SCDG were carried out on a variety of bulk Schott and Corning glasses to investigate the saturation in the index change. The first problem encountered was the photodarkening of color filter glasses with increasing exposure to laser energy, eventually leading to a steady-state darkening. The Corning glasses were found to require larger total fluxes for damage to occur than the Schott glasses, and the total darkening was larger in the Schott than the Corning glasses. Subsequent experiments all were performed with "darkened" glasses whose nonlinear properties no longer changed with time.

The degenerate four-wave mixing experiments did show saturation in the signal with increasing incident power. Unfortunately, the four-wave mixing signal becomes progressively less sensitive to saturation as the power is increased. However, the values for the nonlinearity and its saturation change were in reasonable agreement with the predictions of the plasma model provided by Stephan Koch, establishing it as a predictive tool for studying trade-offs in material parameters.
MEASUREMENTS OF CHANNEL WAVEGUIDE NONLINEARITIES

To verify that band filling is the dominant nonlinearity in the channel waveguides, we measured with picosecond pulses the change in transmission with wavelength of channel semiconductor-doped Schott glass waveguides at various power levels. The glasses were first exposed to high-energy laser pulses until the change in transmission stabilized, meaning that the photodarkening had saturated. Unless this is done, the channel transmission varies not only with power, but also with accumulated pulse energy. A clear blue-shift of the transmission curves with increasing power indicated that the nonlinearity is the result of band filling in the semiconductor-doped crystallites, establishing the electronic nature of the nonlinearity.

Pump-probe experiments as a function of wavelength were also made. Here the resonant absorption is bleached out by a strong pump pulse and then probed by a weak beam delayed in time relative to the pump pulse to monitor the recovery of the absorption. This gives directly the nonlinearity relaxation time and the fraction of the total absorption which is attributable to the resonant nonlinearity. We found that up to 90% of the absorption could be bleached out in a few cases, and that the recovery time of the nonlinearity was approximately 20 ps. More typically, less than 50% of the absorption could be bleached out at high powers.

The key question for the semiconductor-doped glasses was now the exact value of $\Delta n_{\text{sat}}$, and whether it was large enough to allow implementation of all-optical switching devices. Using a novel pump-probe form of a hybrid Mach-Zehnder interferometer, we were able to measure with a single apparatus the magnitude and sign of the maximum nonlinear phase shift, the attenuation and its variation with intensity, and the nonlinearity relaxation time (providing that it is larger than one picosecond), all inside a waveguide. All of these quantities were measured as a function of detuning from the bandgap in the doped-glass channel waveguides.

The maximum optically induced change in the refractive index was measured as a function of wavelength. We found $\Delta n_{\text{sat}}$ to increase with increasing wavelength above the bandgap, reach a maximum, and then decrease with subsequent increases in wavelength. The largest $\Delta n_{\text{sat}}$ measured was approximately $5 \times 10^{-5}$. The materials figure of merit, $W$, was also measured and found to increase monotonically with wavelength, reaching a value of approximately 0.5 where the waveguide scattering loss equaled the loss caused by the band-filling effect. This value of $W$ is too small to allow implementation of a nonlinear directional coupler purely based on nonlinear refractive effects.

The recovery time of the nonlinearity was measured by delaying the probe beam relative to the pump beam. For both the refractive index and absorption
changes, the measured relaxation time was $20 \pm 1$ ps. By delaying the pump relative to the probe by more than 100 ps, the thermal contribution to the refractive index change was also measured.

ALL-OPTICAL SWITCHING IN A NONLINEAR DIRECTIONAL COUPLER

All of the preceding characterization experiments led to the demonstration of switching for the first time in a nonlinear directional coupler with picosecond recovery times. It is the fastest all-optical integrated-optics switch reported to date. New (longer) directional couplers were designed and fabricated. Two-picosecond, Q-switched, mode-locked pulses from a dye laser pumped by a doubled Nd:YAG laser were used at the input of one of the channels. Pulses were obtained at low powers at the output of both channels. When the pulse power was increased to near the saturation threshold for the nonlinearity, the ratio of the power emerging from the two channels changed by approximately 30%, corresponding to all-optical switching. The "turn-on" was essentially instantaneous, and the coupler nonlinearity relaxed in approximately 20 ps, meaning that switching can be implemented for pulse trains with pulse-to-pulse separations of approximately 50 ps, for crosstalk less than 20 dB.\textsuperscript{26,31}

The physical origin of the switching was differential bleaching of the absorption between the two channels, as expected from the figure of merit for this material. It was not a refractive but rather an absorptive effect. The response of the nonlinear directional coupler was modeled in this limit and excellent agreement with experiment was obtained.\textsuperscript{26,31}

We have also induced a change in the transmission of a nonlinear Mach-Zehnder interferometer of approximately 20% with increasing input power. The mechanism also was bleaching of the absorption.

FIGURES OF MERIT FOR OTHER SEMICONDUCTORS

As part of theoretical program, the nonlinear response was evaluated for a variety of semiconductors.\textsuperscript{33} The analysis was based on the Banyai-Koch theory, known as the plasma model. Input values are typical solid-state parameters such as free electron mass and band-gap energy. This theoretical work has shown that the semiconductor CdTe has a figure of merit about five times that of CdS, and therefore should prove useful in making nonlinear directional couplers around the wavelength region of GaAs.\textsuperscript{33} As a result, we have obtained color filter glasses which supposedly contain CdTe from Corning, Schott and Hoya. Unfortunately the Na-ion content is not known and we have to measure it ourselves in order to make
ion-exchange waveguides. The picosecond laser has already been converted to operation on Styryl 9 in the 0.8 to 0.9 \( \mu m \) wavelength region.

We also have begun a program to evaluate semiconductor materials for all-optical switching guided-wave devices in the 1.3 to 1.5 \( \mu m \) wavelength regions. The projected figures of merit (W) are on the order of 10 to 20, although the nonlinearity relaxation times may ultimately prove too long for switching of serial pulse trains. We have obtained from CSELT in Italy single-mode channel waveguides fabricated from InAlGaAs on InP. The band gaps are in the 1.2 to 1.4 \( \mu m \) region. We have been able to observe waveguiding in these samples. Furthermore, we have extended our collaboration with Charles Ironside (UG) to include switching devices in the 1.3 to 1.5 \( \mu m \) range and expect him to bring some waveguide samples in April. (This is the same group with which we collaborated in the CdSSe work.) A color center laser is being installed to access this wavelength range.

CONCLUSIONS

All-optical integrated optics switching devices have been investigated, both as a generic class of devices, and in particular as nonlinear directional couplers in CdSSe-doped glasses. Figures of merit have been defined and evaluated for semiconductor-doped glasses by measuring the third-order nonlinearities. Nonlinear directional couplers have been designed and all-optical switching demonstrated.

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