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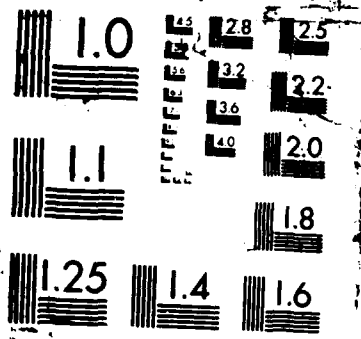
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SIGNAL PROCESSING WITH DEGENERATE FOUR-WAVE MIXING

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PROGRAM GOALS

The original goals of this program were:

1. To demonstrate degenerate four-wave mixing in planar waveguides,
2. To fabricate nonlinear organic waveguides using the Langmuir-Blodgett (L-B) technique, *not*
3. To demonstrate all-optical signal processing based on degenerate four-wave mixing.

During the course of this program, it became clear that nonlinear organic waveguides based on L-B deposition techniques would never exhibit losses low enough for guided-wave degenerate four-wave mixing. As a result, it was decided:

4. To develop semiconductor-doped glass waveguides.
5. To demonstrate degenerate four-wave mixing in these waveguides, *not*
6. To demonstrate all-optical signal processing in semiconductor-doped waveguides. *✓*

RESEARCH PERFORMED

Phase 1: Initial Degenerate Four-Wave Mixing Experiment

Degenerate four-wave mixing (DFWM) was demonstrated in a planar waveguide geometry for the first time. The sample geometry consisted of a thin high-index glass guiding film deposited on a fused-silica substrate, with liquid CS_2 as the cladding medium above the film. The details of the geometry are shown in Ref. 1. The liquid was held in a special cell which was optically contacted to the thin-film surface. Three separate guided waves were excited by means of three separate coupling prisms. The probe beam was introduced at a right angle to the pump beams. The nonlinear interaction occurred in the CS_2 because of the weak evanescent fields which penetrate into the cladding. A reflectivity of 10^{-9} was measured.

This experiment showed that DFWM was possible. However, two major drawbacks contributed to the very small reflectivity. First, the guided-wave fields in the cladding medium were relatively weak since only a few percent of the guided-wave power actually existed in the nonlinear cladding. Further, the peak field occurred inside the film. Obviously, a nonlinear cladding is not an efficient utilization of the guided-wave power.

The second problem was the weak nonlinearity associated with CS_2 . Although this material is the standard against which nonlinearities in general are compared, it is still a very small nonlinearity for implementing efficient degenerate four-wave mixing. More nonlinear materials were clearly required.

Phase 2: Fabrication and Testing of Langmuir-Blodgett Films

The fabrication of thin films by means of the Langmuir-Blodgett technique was implemented. A Vickers system was purchased and installed, along with a special water de-ionizing and filtration system, in a clean-room environment. The graduate student on this phase of the project, C. Naselli, spent two summers at IBM in San Jose with J. Swalen and his group, learning to operate the system. After approximately eighteen months, state-of-the-art Langmuir-Blodgett films up to 400 layers ($1\text{ }\mu\text{m}$) thick were being produced here.

Although guiding in L-B films was accomplished (that is, guided-wave streaks were visually verified), the propagation losses were very high. The best films had attenuation coefficients greater than 10 dB/cm . After consulting various groups with much greater experience in this field, it was concluded that L-B films are unlikely to prove useful for low-loss waveguides, especially of the nonlinear variety which typically involve more complex molecular systems than those usually used in L-B films. In fact, the work of the GTE group with polydiacetylenes showed propagation losses of 100 dB/cm , causing them to abandon the L-B approach.

Films produced with our L-B tank did prove useful for other research. Specifically, Brillouin scattering was used for the first time to measure the elastic properties of L-B films.

Phase 3: Identification of Semiconductor-Doped Glasses for DFWM

The initial experiments with CS_2 -clad waveguides made it clear that efficient DFWM in waveguides requires that the nonlinear medium be the guiding layer and that the nonlinearities be a few orders of magnitude larger than those of CS_2 .

It was decided that color-filter glasses, which contain small ($100\text{ }\text{\AA}$ in diameter) crystallites of $\text{CdS}_x\text{Se}_{1-x}$ semiconductor, should be investigated as a nonlinear waveguide medium. According to the original work of Jain and Lind, the nonlinearities are on the order of $n_2 \cong 10^{-14}\text{ m}^2/\text{W}$ (subsequently verified here). Our work, by both DFWM and luminescence decay, has shown that the nonlinearity "turn-off" times can be as short as a few tens of picoseconds. Further, the host

glasses typically contain 5% to 10% sodium, making them excellent candidates for waveguide fabrication via ion-exchange in silver or potassium nitrate melts.

At this point we requested that the scope of the program be enlarged to include semiconductor-doped glasses as a potential material system for implementing degenerate four-wave mixing in waveguides. Simultaneously, examination of nonlinear organic systems for waveguide degenerate four-wave mixing was continued.

Phase 4: Fabrication and Characterization of Semiconductor-Doped Glass Waveguides

The first waveguides in semiconductor-doped glasses from the Schott Glaswerkes were made from silver-nitrate melts. The silver ions diffused in, exchanging for the sodium ions, which diffused out. It was possible to produce single-mode waveguides, but the waveguiding losses were large because the silver ions tended to form clusters, using the crystallites as nucleation sites. However, these efforts did show that waveguides could be produced by ion-exchange.

At this point a collaboration was established with workers at the Department of Electrical Engineering at the University of Glasgow, who have extensive experience in ion-exchange waveguide fabrication. Special color-filter glasses with a large sodium concentration (10% to 12%) were ordered from Schott Glaswerkes. The ion-exchange parameters for potassium-sodium exchange were established experimentally and the waveguides fully characterized with respect to field structure and losses. The best waveguides exhibited waveguiding (versus absorptive) losses of 0.1 to 0.3 dB/cm.

Simultaneously, bulk degenerate four-wave mixing experiments were performed on these and other glasses manufactured by Schott. These glasses were found to darken on continuous exposure to high-energy laser pulses. This is commonly called solarization and is accompanied by a loss of nonlinearity, effectively ruling out the Schott materials as candidates for these waveguide experiments. The Corning glasses did not solarize easily, requiring orders of magnitude more cumulative laser energy for darkening to occur. This is because the host glass is a borosilicate which is much less susceptible to solarization. The nonlinearities of the Corning filter glasses were found to be comparable to those measured for the Schott filters.

It was originally believed that Corning color-filter glasses could not be successfully ion-exchanged because of their low sodium content. However, one of the graduate students took the initiative, and after some experimentation, found that a single-mode ion-exchanged waveguide could in fact be made in these glasses, but that the ion-exchange time had increased by approximately one order of magnitude. The

losses in these waveguides were quite low, again on the order of tenths of a dB per centimeter. It is these guides in which efficient waveguide degenerate four-wave mixing was ultimately implemented.

Phase 5: Efficient Waveguide Degenerate Four-Wave Mixing

Degenerate four-wave mixing was implemented in the semiconductor-doped waveguides. An actively Q-switched, frequency-doubled Nd:YAG laser produced 10-ns 0.53- μm pulses. Prism coupling was used to excite the pump and probe beam guided modes, and to remove the conjugate signal from the waveguide. Reflectivities up to 1% were obtained before saturation occurred. Analysis of this saturation effect is still not complete. To date, it has been shown that the saturation can be modeled adequately using a carrier-density-dependent relaxation time. It is felt that this approach is more reasonable than the hypothesis suggested by Flytzanis et al., who attribute this saturation to Auger processes.

Waveguide DFWM has also been implemented with picosecond pulses. In this case 10% reflectivities were obtained before the onset of saturation. The effects of saturation appear to be quantitatively different and this difference is currently under investigation.

Although this contract has now expired, prior to the implementation of all-optical signal processing functions, we are now on the threshold of investigating this aspect, especially since a good nonlinear system has already been developed. Investigation will continue.

Phase 6: Nonlinear Organic Waveguides

The suitability of organic-film waveguides for all-optical signal processing has been under investigation in an effort paralleling the semiconductor-doped glass waveguide work. Initially these investigations undertook definition of the organic-film properties necessary for these applications. The key parameters were found to be the saturation change in refractive index (Δn_{sat}), the magnitude and sign of the nonlinearity (n_2), the waveguide loss coefficient (α), the "turn-off" time of the nonlinearity, and the material damage threshold. These parameters have been compiled from available literature and it is clear that the necessary information is simply not available and therefore must be measured.

Nonlinear organic materials, supplied by various sources and prepared in this laboratory, are being assessed. To date, the most promising appears to be PMMA

spun-on films, doped with MNA up to 50% by volume. It has been determined that such waveguides can have propagation losses of less than 1 dB/cm, and nonlinearities on the order of 10^{-17} m²/W. Degenerate four-wave mixing in these waveguides will be attempted using picosecond pulses.

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