**REPORT DOCUMENTATION PAGE**

1. **AGENCY USE ONLY (Leave blank)**
2. **REPORT DATE**
   12/18/96
3. **REPORT TYPE AND DATES COVERED**
   Final 8/1/93 - 7/31/96

4. **TITLE AND SUBTITLE**
   PHOTOREFRACTION AS A DIAGNOSTIC TOOL FOR InP

5. **FUNDING NUMBERS**
   F49620-93-C-0452
   61103 D
   34841TS

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8. **PERFORMING ORGANIZATION**
   AFOSR-TR-97-
   0005

9. **SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)**
   AFOSR/NE
   110 DUNCAN AVENUE, ROOM B-115
   BOLLING AFB DC 20332-8080

11. **SUPPLEMENTARY NOTES**
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12a. **DISTRIBUTION / AVAILABILITY STATEMENT**
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12b. **DISTRIBUTION CODE**

13. **ABSTRACT (Maximum 200 words)**
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14. **SUBJECT TERMS**
15. **NUMBER OF PAGES**
16. **PRICE CODE**
17. **SECURITY CLASSIFICATION OR REPORT**
    UNCLASSIFIED
18. **SECURITY CLASSIFICATION OF THIS PAGE**
    UNCLASSIFIED
19. **SECURITY CLASSIFICATION OF ABSTRACT**
    UNCLASSIFIED
20. **LIMITATION OF ABSTRACT**
    UL
FINAL TECHNICAL REPORT

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ABSTRACT

The phase shift between the intensity pattern and the index pattern in two-wave mixing experiments has been determined from measured values of photorefractive two-wave mixing gain and diffraction efficiency in a bulk InP:Fe crystal as a function of the total incident intensity. Remarkably, the phase shift varies from 0 to 180 degrees with a value of 90 degrees at the intensity for which the two wave mixing gain peaks. Comparison between theory and experiment yields values for sample Fe$^{2+}$ and Fe$^{3+}$ connections at $6 \times 10^{15}$/cm$^3$ and $4.8 \times 10^{16}$/cm$^3$ respectively.
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I. PROBLEM STUDIED

Diagnostic tools to determine local dopant concentration and local mobility can make possible the development of ultra-fast electronic and photonic devices. We have investigated the potential of the use of the photorefractive effect to provide an unambiguous measurement of the concentration of Fe$^{2+}$ and Fe$^{3+}$ in InP.

II. SUMMARY OF RESULTS

Studies of the photorefractive effect in InP:Fe show unusual behavior [1,2]. The two wave mixing (TWM) gain coefficient, in the presence of an applied d.c. electric field shows a sharp resonance as a function of the total incident intensity. In addition, the resonance peak is found to shift to higher intensities which increase in crystal temperature. These observations are unexpected since a characteristic feature of the photorefractive effect in oxides is that its magnitude is independent of pump intensity for an incident intensity exceeding a critical value depending on the dark irradiance of the material. This intensity dependent behavior is, however, successfully explained by an elegant model developed by Picoli et al [3]. The resonance peak is seen when thermally generated electrons are matched by photogenerated holes. In addition to the observed intensity dependent TWM gain, the model also predicts an intensity dependent phase shift between the intensity spatial pattern and the corresponding index grating. The predicted phase varies from 0 to 180 degrees as the incident intensity is changed. This prediction is also unexpected since another characteristic feature of the photorefractive effect in oxides is that the phase is intensity independent, although its value is predicted to depend on the magnitude of the
applied field. This predicted intensity dependent phase behavior in InP:Fe has, to our knowledge, never been observed.

As a result during this project, we report the first observation of an intensity dependent phase shift between the photorefractive index pattern and the intensity pattern for InP:Fe crystals. The phase shift varies from near 0 degrees to near 180 degrees, reaching a value of 90 degrees for an intensity corresponding to the peak gain. Our results, although conceptually surprising, are in good agreement with the Picoli model [3] and add further credibility to its application.

The InP crystals studied were produced by magnetically stabilized Kyropoulos growth [4] at Hanscom Air Force Base. In this technique, crystals are grown in an applied magnetic field of 2000 gauss which results in improved uniformity of dopant distribution from seed to tail of the crystals. For these crystals the residual donor impurity concentration is of the order of mid $10^{15}$ cm$^{-3}$. This impurity is likely to be silicon which enters the melt from either the charge material or the Silica crucible. The InP crystals were grown with a controlled amount of iron dopant to give a range of $2.10^{16}$ to $2.10^{17}$ atoms.cm$^{-3}$ in the crystals. Figure 1 is an energy level diagram showing the Fe deep acceptor level when empty, Fe$^{3+}$, and when filled, Fe$^{2+}$, and the shallow impurity levels. Under illumination the carriers are both electrons and holes although thermal excitation of electrons is dominant over optical excitation while optical excitation of holes is dominant over thermal excitation [5,6,7].

Different techniques can be used to measure the phase shift between the intensity pattern and the index pattern [8-13]. We determined the phase shift by measuring the gain and the diffraction efficiency in a TWM experiment. For our experiment the grating vector was oriented along <001> and the optical beams propagated along <110> and were polarized along $\langle\bar{T}10\rangle$ (Figure 2).
Experimentally, the gain $\Gamma$ is found from the TWM signal amplification. To measure the diffraction efficiency $\eta$, the diffracted part of the pump in the signal direction is measured during the TWM experiment by switching off the signal beam. Since the decay time for the photorefractive index grating with only the pump beam is about 50 ms for an intensity of 100 mW/cm$^2$, a shutter speed of 500 $\mu$s was used to switch off the signal beam while a digital oscilloscope acquired the signal diffracted from the pump beam. The measurement was performed with a titanium-sapphire laser tuned to 1.02 $\mu$m. To limit the intensity variation and consequently the phase variation along the crystal due to absorption, a short 1.7 mm long crystal was used. In this case, the phase shift is nearly constant throughout the sample since the absorption coefficient at 1.02 $\mu$m is about 1 cm$^{-1}$. The intensity ratio between the pump beam and the signal beam is 20 at the entrance face of the crystal and the grating with Peltier coolers and a 10 kV/cm d.c. electric field is applied in the $<001>$ direction. The measured TWM gain and the phase shift determined from the gain and measured diffraction efficiency are shown as a function of total intensity in Figure 3 assuming the phase is 0 for low intensity. The sample exhibits a sharp resonance with a maximum gain greater than 10 cm$^{-1}$. As predicted the phase shift varies from 0 to near 180 degrees as a function of intensity and reaches a value of 90 degrees at the resonance intensity. Since the measurement determines only $\sin (\phi)$, it is not possible to differentiate a phase angle of $\alpha$ from a phase angle of 180-$\alpha$ but previous work on TWM with a moving grating [14] has shown that the phase shift changes from less than 90 degrees to greater than 90 degrees as the intensity varies from one side of the resonance intensity to the other side. The quantitative fit of the experimental points using $n_{T0}$ and $p_{T0}$ as fitting parameters is shown in figure 3. The fit yields values of $n_{T0} = 6 \times 10^{15}$ cm$^{-3}$ and $p_{T0} = 4.8 \times 10^{16}$ cm$^{-3}$. These values are
consistent with the range of $n_{70}$ and $p_{70}$ measured by different techniques in similar samples grown in our lab [15].

In conclusion, we have demonstrated experimentally for the first time that the phase shift between the index grating and the illumination grating for TWM experiments in InP:Fe with a d.c. applied field is strongly intensity dependent. The phase shift is 0 for low intensity, then increases to 90 degrees at the resonance intensity and reaches 180 degrees for high intensity. This result gives further credibility to the Picoli model and are used to determine the Fe$^{2+}$ and Fe$^{3+}$ concentrations in InP samples. For example, in the sample used to produce the data in Fig. 3, the Fe$^{2+}$ concentration is measured at $6 \times 10^{15}$/cm$^3$ while the Fe$^{3+}$ concentration is determined to be $4.8 \times 10^{16}$/cm$^3$.

III. REFERENCES

FIGURE CAPTIONS

Fig 1: Energy level diagram for InP:Fe including deep Fe acceptor and shallow donors and acceptors.

Fig 2: Optical set-up to measure the phase shift between the intensity pattern and the index pattern.

Fig 3: Gain and phase shift as a function of intensity in a 1.7mm long InP:Fe crystal. Fitting curves using parameters

\[ \sigma^e_0 = 6 \times 10^{-18} \text{ cm}^2, \sigma^p_0 = 1 \times 10^{-17} \text{ cm}^2, T = 293 \text{ K}, n_{70} = 6 \times 10^{15} \text{ cm}^{-3}, p_{70} = 4.8 \times 10^{16} \text{ cm}^{-3}. \]
Shutter and lenses

Polarization

N. D. Filter

<001> x

<110> z

Signal beam

Pump beam

FIGURE 1
FIGURE 2
FIGURE 3