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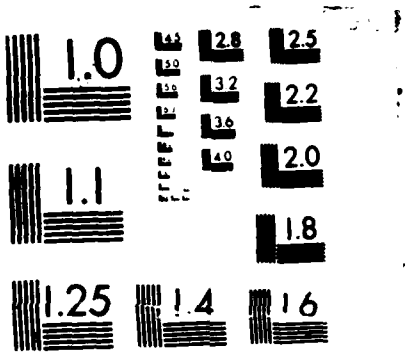

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NONLINEAR OPTICAL PROPERTIES AND SUBPICOSECOND DYNAMICS OF EXCITONS AND ELECTRON–HOLE PLASMAS IN MULTIPLE QUANTUM WELL STRUCTURES

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Time resolved photoluminescence measurements are reported for MBE-grown $Al_xGa_{1-x}As$ samples having alloy compositions which result in both direct gap and indirect gap materials. Quantitative results have been obtained for bandgap renormalization induced at high carrier densities and the results compared with a theoretical model. Ultrafast spectroscopy measurements on an MBE-grown x=0.52 sample following optical excitation well above the bandgap show presistent bleaching of the direct absorption, induced absorption below the fundamental absorption edge and a blue shift by Fabry-Perot fringes in the transparent spectral region.

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INTRODUCTION

This document reports progress on the AFOSR contract entitled "Nonlinear Optical Properties and Subpicosecond Dynamics of Excitons and Electron-Hole Plasmas in Multiple Quantum Well Structures" for the period August-December 1986. Progress on internal IR&D projects on which we have agreed to provide AFOSR unrestricted access to is also presented. The primary achievements during this time period have been in three areas: Task 1 of the descoped work statement--*Time-Resolved Photoluminescence and Transient Absorption Spectroscopy of MBE-grown Ternary Alloys*, and two internal IR&D projects--*Ultrafast Photorefractive Effects* and *Photoreflective Characterization of MBE-Grown Structures*. A brief summary of each follows.

Time-Resolved Photoluminescence and Transient Absorption Spectroscopy of MBE-Grown Ternary Alloys

The principle objective of this task is to apply picosecond time-resolved photoluminescence and ultrafast spectroscopy to investigate optical nonlinearities associated with band-gap narrowing, alloy scattering, nonlinear diffusion, and bulk and surface recombination in MBE-grown heterostructures for potential application to high-speed nonlinear optical devices.

Previously we reported (August 1986 Annual Report) results of time-resolved photoluminescence on a direct-gap LPE-grown $Al_xGa_{1-x}As$ sample with x=0.23 and two MBE-grown samples--one direct-gap sample with x=0.38 and one indirect-gap sample with x=0.52. The luminescence from the sample with the alloy composition of x=0.23 was typical of direct gap materials. The samples with the larger x-values, however, displayed luminescence peaks from both direct and indirect gaps. In each case, the features of the luminescence provided qualitative evidence for the influence of alloy scattering and multiple-valley electron occupation on band gap narrowing in

these alloy semiconductors.

Since our last reporting, we have performed time-resolved photoluminescence measurements on two additional samples (x=0.49 and 0.41) and have continued to analyze the volumes of data obtained from the five crystals. This study has produced quantitative results for the band-gap renormalization induced at high carrier densities, and a comparison of the renormalization to that expected from the universal formula of Vashishta and Kalia[1] (VK) has been made. The renormalization of the direct gap in the sample with a low aluminum concentration (x=0.23) was found to be in excellent agreement with this universal formula. For x values near the direct-to-indirect gap crossover ($x \simeq 0.43$), the electrons are distributed among both the direct and indirect valleys, and multiple luminescence peaks are observed. This distribution of the carriers amongst the various conduction band minima and the admixture of the heavy X-valleys with the lighter central valley that results from alloy scattering both influence the observed renormalization.

Specifically, we find that, for indirect gap samples with x=0.49 and 0.52 and carrier densities in the range $2.5\text{-}6\times10^{19}$ cm^{-3}, the renormalization of the X-valleys is in excellent agreement with the universal formula. The direct gap renormalization, however, is found to be approximately 47 meV less than predicted by the VK formula. This is a consequence of the low electron densities in the direct valley. The renormalization of the direct gap is then governed by hole exchange and electron and hole correlation--the electron exchange should be negligible. Indeed, we find that subtracting the electron exchange energy from the renormalization predicted by the VK formula produces excellent agreement with our data.

The observed renormalization of the direct gap in direct-gap samples with x=0.38 and 0.41 and carrier densities in the range $1\text{-}5\times10^{18}$ cm^{-3} is found to be approximately 56 meV in excess of that predicted by the VK formula. This can be attributed to the admixture of the light central valley with the heavy side valleys

through alloy scattering. This band mixing can be accounted for phenomenologically by increasing the optical mass and subsequently the density of states mass of the central valley. The increased mass leads to an effective increase in the electron concentration in the central valley and, thereby, a larger renormalization. Our analysis indicates that a mass enhancement factor of 4.3 results in a calculated renormalization that is in excellent agreement with our data. This enhancement factor is consistent with that obtained by Pearah *et al.* from excitonic absorption measurements.[2]

Our ultrafast spectroscopy measurements have been concentrated on the MBE-grown x=0.52 sample. One section of this sample was prepared for transmission measurements by Dr. C. A. Lee and his graduate students at Cornell University. Well-defined windows were etched through the sample substrate leaving regions of semitransparent 2-μm-thick AlGaAs. These regions were then excited with 30-psec pulses at 532 nm (well above the direct gap). The excited region was probed at various time delays by a weak broadband picosecond continuum pulse generated by tightly focusing a 40 psec 1-μm pulse into a water cell. The transmitted probe was dispersed in a 0.25-m spectrometer and then directed into a streak camera whose output was detected with a vidicon and optical multichannel analyzer. A reference continuum pulse, which did not pass through the sample, was delayed by a few hundred picoseconds and then arranged to travel collinearly with the probe. The data were normalized to this reference to eliminate shot-to-shot fluctuations in the continuum spectra. Although temporal resolution in this measurement was achieved by mechanically changing the path length that was traversed by the probe, several benefits were obtained by using the streak camera. The camera provided a convenient means of separating the probe and the delayed reference pulse. The high sensitivity of the streak camera greatly enhanced signal-to-noise in the measurement. Finally, for delays greater than the excitation pulsewidth, scattered light from the

excitation source was completely eliminated.

Three significant features were observed in the transmitted probe spectra for 532-nm excitation of this sample at 15K: (i) a significant and persistent (hundreds of picoseconds) bleaching of the direct absorption, (ii) induced absorption below the unexcited fundamental absorption edge, (iii) a blue shift of the Fabry-Perot fringes in the transparent spectral region. The persistent bleaching arises from band filling in the valence band and is consistent with our estimate of the quasi-Fermi level for holes obtained in the analysis of our luminescence data for similar excitation levels. The induced absorption at energies below the unexcited absorption edge arises from band gap renormalization. The blue shift of the Fabry-Perot fringes is consistent with a negative change in the refractive index.

Ultrafast Photorefractive Effects

The principal objectives of this task are to use ultrafast optical techniques for the study of ultrafast photorefractive effects in various materials for possible device applications and for the control and characterization of the speed and performance of various electronic and optical devices. Recent efforts have focused on two tasks: Our continued theoretical modeling of the voluminous and varied experimental data associated with our observations of picosecond photorefractive effects in GaAs and our experimental measurements of similar effects in $BaTiO_3$.

Recently, we (together with collaborators at North Texas State University) completed the first experimental investigations of photorefractive effects in GaAs on subnanosecond time scales. In these studies, the initial experimental configuration was a two-beam coupling geometry in which the energy transferred between two 43-ps, 1.06-μm pulses that were spatially and temporally coincident in the GaAs sample was measured as a function of the ratio of energies in the two pulses, the total fluence, time delay between the two pulses, and crystal orientation. We found

the direction of energy transfer between two equal beams to depend definitely on crystal orientation--an unambiguous signature of the photorefractive effect. However, for these pulsewidths, the accompanying contributions of two-photon absorption and free-carrier transient energy transfer (through the free carrier index) to the probe loss or gain were also evident. In subsequent experiments, a three-pulse transient-grating geometry was used. In the latter, two coincident pulses interfered in the sample to produce a grating with a period of 1.7 μm. The grating formation and decay was interrogated by measuring the diffraction efficiency of a third, time-delayed probe pulse that was counter-propagating to one of the writing pulses.

Initially, we concentrated our theoretical efforts on interpretation of the measurements of the two-beam coupling as a function of the total fluence and crystal orientation, and we ignored the complications introduced by time delays between the pump and the probe pulses. Our theoretical calculations of the photorefractive energy transfer in GaAs on these time scales are based on simple rate equations for carrier generation and diffusive charge transport. By including the effects of transient energy transfer and probe loss through two photon absorption, we have successfully fit the net probe gain over the fluence range of 0.01 to ~10 mJ/cm^2 both for the crystal oriented for photorefractive energy transfer from strong-to-weak beam and for the crystal oriented for photorefractive energy transfer from weak-to-strong beam. As observed, the model predicts a net probe gain (or loss) dominated by photorefractive beam coupling for fluences below ~1 mJ/cm^2, by probe loss associated with two-photon absorption for fluences between 1 and 10 mJ/cm^2, and by probe gain arising from transient energy transfer for fluences above ~10 mJ/cm^2.

In these studies, we have also isolated (experimentally and theoretically) the photorefractive effects from those of two-photon absorption and free-carrier transient energy transfer. To do this, we subtracted the *net* probe gain for the

crystal oriented for photorefractive probe gain from that for the crystal oriented for photorefractive probe loss. For small energy transfer and no pump depletion, the difference for the two orientations should be twice the photorefractive transfer alone. Results show that the measured photorefractive beam coupling gain of up to 4% in the fluence range 0.03 to 0.3 mJ/cm^2 is caused by a charge separation between photoionized electrons and positively ionized EL2 donors. For fluences between 3 and 10 mJ/cm^2, photorefractive gains of up to 12% can be attributed to charge separation between electron-hole pairs produced by two-photon ionization.

In the time elapsed since the AFOSR annual report was prepared, we have extended our calculations to allow for a time delay between the pump and probe. Using the same model as in previous calculations, we were able to account for the rich and varied structure in the probe gain as a function of delay. This structure is very sensitive to the relative strengths of photorefractive gain (loss), two-photon absorption, and free-carrier transient energy transfer. Comparison of our calculations to features in the data confirmed our choice of coupling coefficients (taken from the literature or independent measurements) and confirmed the accuracy of the model. During the coming months, we plan to extend our model and code to describe the three-pulse transient grating data. These measurements provide temporal information on grating decay as well as formation.

We have also investigated photorefractive index gratings in $BaTiO_3$ that were written, read and erased by single pulses 43-ps in duration at a wavelength of 0.532 μm and fluences in the range of 1 to 15 mJ/cm^2. One of the geometries used in our experiments was a three-pulse transient grating configuration in which two pulses interfered in the crystal to form a grating of period 0.48 μm. The grating formation and decay was interrogated by a third, time-delayed probe pulse that was counter-propagating to one of the writing pulses. The writing pulses were p-polarized, and the probe was s-polarized. Initially, the crystal was oriented such that the grating k-

vector was parallel to the c-axis so that any internal space charge field produced by the pump beams would produce a refractive index variation through the r_{13} electro-optic coefficient that could be sensed by the probe pulse. Subsequently, the crystal was rotated such that the k-vector was perpendicular to the c-axis so that no photorefractive effects would be sensed by the s-polarized probe.

Two distinct features are evident in the data. The first is a strong peak centered about zero delay with a width of approximately 65 ps, roughly the intensity autocorrelation time of our pulses. The strength and duration of this peak are independent of crystal orientation, and its properties are consistent with diffraction from a free-carrier index grating generated by one- and two-photon absorption. The short lifetime and symmetry of this peak suggest that the carrier lifetime is less than 50 ps. The second feature, a relatively constant, long lasting diffraction efficiency, is roughly 50 times weaker than the peak and is present only when the grating k-vector is perpendicular to the c-axis--a definitive signature of the photorefractive effect. The flat nature of this signal for tens of nanoseconds following excitation suggests that grating formation is complete shortly after the end of the excitation pulses.

In summary, we have observed the formation of photorefractive gratings in $BaTiO_3$ in less than 100 ps by using 43-ps pulses at 0.532 μm. This suggests a recombination time in our sample of less than 100 ps. The magnitude of the grating diffraction efficiency ($\sim 3 \times 10^{-6}$) indicates a less efficient photorefractive effect than found in GaAs with picosecond pulses or in $BaTiO_3$ with cw or nanosecond illumination.

Photoreflectance Characterization of MBE-Grown Structures

The objective of the task is to apply reflectance modulation techniques (photoreflectance) to characterize various MBE-grown multiple quantum wells and

heterostructures in order to determine their quality prior to embarking on tedious and time consuming nonlinear optical measurements.

Previously, the photoreflectance measurements and analysis were exclusively conducted in collaboration with Dr. Fred Pollak and his students at Brooklyn College of the City University of New York. This collaboration led to the first observation of forbidden transitions at room temperature in a compositional MQW's and the first photoreflectance study of GaAs doping superlattices. We have also developed, in collaboration with Dr. C. A. Lee and his students at Cornell University, a successful etching procedure for removing selected regions of the MQW's substrate to render semitransparent regions. Such regions have been investigated (in collaboration with co-workers at North Texas State University) using picosecond pump-and-continuum-probe techniques. The observed saturation of the exciton transition by the electron-hole plasma is consistent with observations by others.

More recently, our efforts have been concentrated on developing photo-reflectance facilities and curve fit routines for in-house (at North Texas State University) analysis of MQW's. This work is now complete, and we are routinely measuring and analyzing spectra from MQW's with this facility. This system has been used to analyze two MQW's with similar growth parameters produced at separate facilities. One sample consisted of 100 periods of 100A GaAs wells alternating with 150A AlGaAs ($x \simeq 0.3$) barriers, the other 100A GaAs wells alternating with 100A AlGaAs ($x=0.28$) barriers. Room temperature photoreflectance (PR) and absorbance spectra of each multiple quantum well structure were obtained. The spectral positions of all bound symmetry-allowed and several symmetry-forbidden transitions were visible in both the PR and the absorbance measurements. The energies for these transitions as determined by absorption techniques agreed to within 2 meV for the two samples. When these same transition energies were determined by PR, they agreed with those extracted from the absorbance

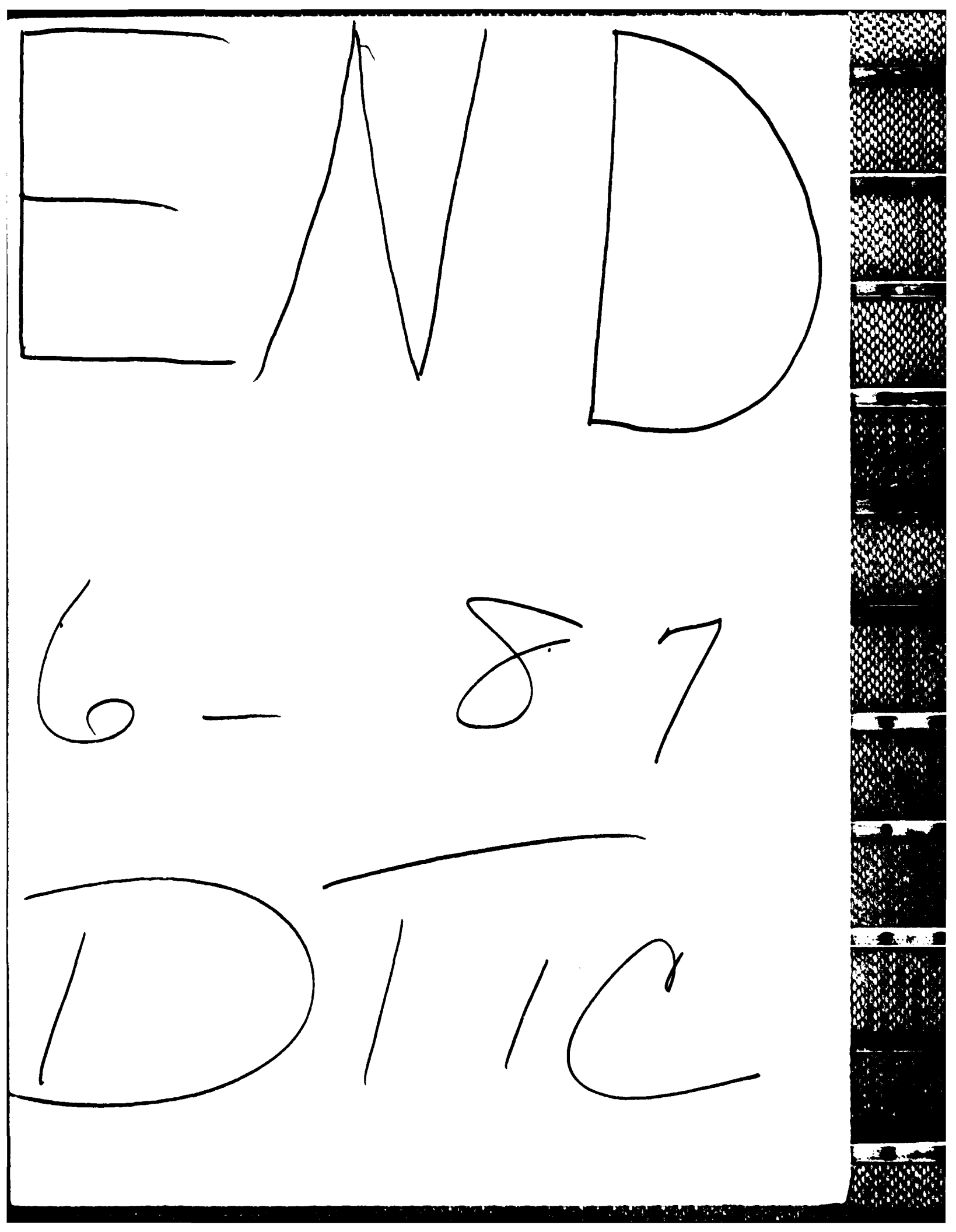
measurements to within a few meV for the lowest levels and agreed qualitatively for the higher confined levels.

We also used photoreflectance to analyze radiation damaged and annealed MQW's to determine whether successful uniform alloying could be achieved. Such a process has potential application to the development of MQWS distributed feedback lasers. Spectra were obtained for three samples that were implanted with Si dosages of 10^{14} cm^2, $2x10^{14}$ cm^2, and 10^{15}/cm^2. All samples were annealed following implantation. Spectra were also obtained for a control sample that was annealed but not implanted. Qualitatively, derivative-like features associated with charge confinement in the wells were observed to broaden, shift, change shape, and diminish in magnitude with increasing dosage. At these dosages, however, the features associated with confinement never completely disappeared, indicating that complete uniform alloying was never achieved. Nothing of a quantitative nature can be said, however, without a more detailed investigation of additional samples having larger and smaller implant dosages.

References

1. P. Vashishta and R. K. Kalia, Phys. Rev. **B25**, 6492 (1982).

2. P. J. Pearah, W. T. Masselink, J. Klem, T. Henderson, H. Morloc, C. W. Litton, and D. C. Reynolds, Phys. Rev. **B32**, 3857 (1985).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