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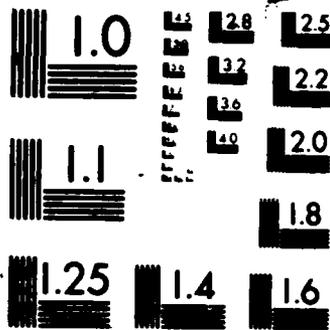
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NONLINEAR OPTICAL PROPERTIES AND SUBPICOSECOND DYNAMICS OF EXCITONS AND ELECTRON-HOLE PLASMAS IN MULTIPLE QUANTUM WELL STRUCTURES

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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. Detailed studies are reported of a marginally indirect sample displaying stimulated emission from both direct and indirect bandgaps.

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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 January-February 1987. We also describe progress on internal IR&D projects on which we have agreed to provide AFOSR unrestricted access. During this brief two month period, our progress has been primarily in three areas: Task 1 of the AFOSR descoped work statement (Time-Resolved Photoluminescence and Transient-Absorption Spectroscopy of MBE-Grown Ternary Alloys) and two tasks associated with Hughes Research Labs (HRL) IR&D projects: Photoreflective Characterization of MBE-Grown Structures and Ultrafast Laser Sources and Applications.

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

The purpose of this task is to apply the techniques of picosecond time-resolved photoluminescence and ultrafast transient absorption spectroscopy to investigate band-gap renormalization, alloy disorder, nonlinear diffusion, and bulk and surface recombination in MBE-grown heterostructures. We have previously reported the results of time resolved photoluminescence measurements on $Al_xGa_{1-x}As$ samples with x-values of 0.23, 0.38, 0.41, 0.49, and 0.52. Analysis of the luminescence data provides us with the carrier density and temperature as well as the renormalized band gap, all as a function of time. Our studies of the band gap renormalization have provided the first experimental evidence of the separate contribution of electron exchange to the total self-energy of a dense electron-hole plasma. These investigations also provide strong evidence of conduction-band mixing as a consequence of alloy disorder. Time resolved transient absorption spectroscopy of the x=0.52 sample showed clear indications of band filling, band-gap renormalization, and nonlinear refraction.

During the past two months, we have continued these studies using additional samples. A series of MBE $Al_xGa_{1-x}As$ samples with x-values of 0.0, 0.2, 0.34, 0.38, 0.40, 0.44, 0.48, 0.50, and 0.54 was grown at HRL for this purpose. Many of these samples proved unsuitable for our

purposes, displaying weak luminescence and short lifetimes (as short as 30 psec). A strong luminescence with a rapid rise time from the GaAs cap layer indicates that perhaps carriers are rapidly diffusing into the cap before they can recombine in the $Al_xGa_{1-x}As$. We are currently growing several samples without cap layers to investigate this phenomena.

One of these samples that does display good luminescence is the x=0.44 sample which is a marginally indirect-gap material but very near the direct-to-indirect gap crossover composition, $x_c \simeq 0.45$. This sample is particularly interesting since it displays simultaneous stimulated emission from both the direct and indirect band gaps. Stimulated emission from the indirect gap is a direct result of alloy disorder. In indirect gap AlGaAs $(x > x_c)$, the strong coupling between side valleys and the central valley of the conduction band leads to quasi-direct transitions out of the indirect bands. At sufficiently high excitation fluences (i.e., carrier densities), one can even surpass the threshold for stimulated emission. Although the transition probability for quasi-direct recombination falls off like $1/\Delta^2$ (Δ = energy difference between Γ- and X-band), we are able to detect stimulated emission out of the X-band for x-values up to 0.52, where Δ=100 meV. The stimulated luminescence always occurs at the low energy edge of the no-phonon line, which strongly supports our interpretation of the X-band luminescence as a superposition of a quasi-direct and an indirect luminescence band. For x=0.44 (just above the crossover composition) the threshold for stimulated emission is significantly reduced compared to x=0.52. Here it is possible, due to the small energy separation of indirect and direct bands, to get degenerate carrier distributions for X- and Γ-bands simultaneously. The stimulated emission turns into a broad luminescence band of up to 55 meV FWHM. The contributions of Γ- and X-band, however, are still distinguishable due to their different temporal evolution. Both the rise and decay time for the X-band stimulated emission are longer compared to the Γ-band contribution. Thus, for short decay times, t, after the excitation pulse, the spectrum is governed by direct recombination. The quasi-direct luminescence develops as a shoulder on the low energy side of the Γ-band emission and reaches its maximum at t=80 ps. After t=200 ps only this quasi-direct stimulated emission remains and is visible for up to 400 ps after excitation. These measurements directly confirm the strength of alloy disorder effects in AlGaAs close to the crossover composition.

Photoreflective Characterization of MBE-Grown Structures

The principal objective of this task is to demonstrate that the linear, modulation spectroscopy technique of photoreflectance can be used routinely for the room temperature determination of the composition, dimensionality, and spatial uniformity of MBE, LPE, and MOCVD-grown heterostructures and multiple quantum wells. Initially, we are attempting to establish the feasibility and utility of photoreflectance in two ways. First, we compare the energy levels extracted from photoreflectance spectra to those predicted by theory. Secondly, we compare the information obtained from photoreflectance with that obtained from other linear techniques such as low temperature photoluminescence and transmission spectroscopy. Once established, we intend to use our facilities and this technique in support of the other tasks in this project by routinely characterizing as-grown samples before they are further processed and used in tedious and time consuming nonlinear optical studies.

Initial studies, in which measured photoreflectance spectra were fit by nonlinear derivative fit-function routines and the extracted energy levels were carefully compared to theory, were principally performed in collaboration with Dr. Fred Pollak and co-workers at Brooklyn College of the City University of New York. As part of this collaboration, we have obtained quantitative fits to the derivative-like features observed in the photoreflectance spectra of both compositional MQWs and GaAs doping (NIPI) superlattices. In studies on one in-house grown compositional MQW structure that consisted of 100 periods of 100Å GaAs wells alternating with 150Å $Al_{0.18}Ga_{0.82}As$ barriers, we observed and identified all allowed quantum-confined transitions and several forbidden transitions. This was the first observation of forbidden transitions in an MQW at room temperature. All transitions were sharper and more clearly resolved in similar spectra obtained at lower temperatures. Energy levels extracted from the photoreflectance spectra obtained at both temperatures are in remarkable agreement with the theoretical values calculated using a tight-binding model.

As stated, this collaboration also led to the first photoreflectance studies of so-called NIPI doping superlattices. Here, spectra were obtained on two samples: one with a small built-in potential (estimated to be ~85 meV) and one with a large built-in potential (~1200 meV). For the

sample with the small built-in potential, the spectral features could be fit by a derivative functional form, and we were able to tentatively identify a number of the features. The sample with the large built-in potential, however, exhibited Franz-Keldysh oscillations. Since the period of these oscillations can be directly related to the built-in dc field, this technique provides, for the first time, a method for directly measuring the built-in field. Details of these results were provided in Appendices A-C of our last annual report (July 1986), and we won't dwell on them further here.

A key result of these studies is that they demonstrate that photoreflectance can provide detailed quantitative information about energy levels, linewidths, barrier heights, and transition strengths in layered materials. From this information, for example, one can readily deduce the layer thickness and aluminum concentration and judge layer uniformity. Although these studies demonstrated good agreement between theory and experiment for selected samples, it must be emphasized that profound questions remain concerning the correct functional form to be used in fitting the photoreflectance spectra. For example, originally we used a third-derivative of a two-dimensional critical point to fit the room temperature photoreflectance spectra and a first derivative of a Lorentzian-shaped excitonic feature to fit the low temperature (77 K) data. Recent work, however, now indicates that a first derivative of a broadened Gaussian-shaped excitonic feature should probably be used for both temperatures--although the Gaussian-broadening should be more pronounced at room temperature. We are presently modifying our fit routines to investigate and include such refinements.

In a second set of experiments designed to establish the credibility of the photoreflectance technique, we are carefully and quantitatively comparing the transitions extracted from photoreflectance line fits to the positions of excitonic features in the absorbance spectra of partially-transmitting MQW samples. This study has been performed exclusively in-house using photoreflectance facilities established at North Texas State University and curve-fit software developed by personnel at Hughes Research Laboratories. Our experimental apparatus is generically the same as the one described in our annual report, so we will not repeat that description here. Our recent comparisons of photoreflectance and absorbance spectra have tended to focus on two MQW samples with similar growth parameters produced at separate facilities. One sample consisted of 100

periods of 100Å GaAs wells alternating with 150Å AlGaAs (x ≃ 0.3) barriers, and the other, 100Å GaAs wells alternating with 100Å AlGaAs (x ≃ 0.28) barriers. Each sample was grown with an etch-stop barrier between the GaAs substrate and the MQW structure. Separate selective-etching procedures were subsequently used to remove selected regions of the GaAs substrate from the MQW structure to produce thin, semitransparent regions on each sample. In our January report, we described initial results of measuring the sample absorbance and photoreflectance of these two samples at room temperature. During January and February, we have continued these studies, primarily extending our measurements to lower temperatures (90 K) and including a sample with narrower well widths (50Å). We now summarize our observations and conclusions to this point.

We begin by emphasizing that all three samples were of excellent "optical quality." By that, we mean that the low temperature photoluminescence from each sample was strong and had a narrow width (< 5 meV); that several excitonic features (e.g., n=1, light hole and heavy hole and the n=2 heavy hole) were clearly visible in the room temperature transmission spectra of each sample; and that all allowed and several forbidden transitions were clearly visible in the low-temperature transmission spectra. By comparison, in typical photoreflectance spectra, the positions of the two lowest lying (n=1 heavy-hole and light-hole) transitions were always visible and readily extracted from the spectra. Moreover, the positions of these features agreed quantitatively with the absorbance peaks corresponding to the same excitonic features. In addition, several strong derivative-like features were always visible near the bandgap energy of the barrier layers. Notice that even if no other features were extracted, the technique has already provided valuable information: the well thickness can be deduced from the positions of the lowest lying levels, the aluminum concentration from the features near the barrier, and the uniformity of the well widths from the linewidths. Other allowed and forbidden transitions are visible; however, for reasons we do not as yet understand, which transitions are visible and their relative strengths vary dramatically from point-to-point on a given sample and from sample-to-sample. Moreover, which transitions are present seems to be influenced by sample processing and surface treatment following growth. Once understood, such variations of features in the photoreflectance spectra may provide valuable information about local field strengths and strain not available in absorbance spectra.

We have already used our photoreflectance apparatus for several routine characterization chores. In our last semimonthly report, for example, we described our use of photoreflectance to analyze radiation damaged and annealed MQW's to determine whether uniform alloying could be successfully achieved. Such a process has potential application to the development of distributed feedback MQW lasers. The reader is directed to our January report for a summary of that study. We have also routinely used photoreflectance to determine the bandgap (and, therefore, the composition) of the MBE and LPE-grown thin layers used in the task on <u>Time-Resolved Photoluminescence and Transient Absorption Spectroscopy of MBE-Grown Ternary Alloys</u> described earlier. As an example of such an application, during January, we characterized five samples grown by LPE in-house (at HRL) by Bob Loo. These samples were later used in the time-resolved studies mentioned above. Initially, however, their compositions were only roughly known. From the photoreflectance spectra, the x-values were determined to be 0.21, 0.25, 0.34, 0.4, and 0.45. We also measured the transmission and photoreflectance spectra of the first MQW grown by MOCVD at HRL. The sample was meant to consist of 100Å GaAs wells separated by 150Å AlGaAs ($x \simeq 0.3$) barriers. The spectra exhibited peaks characteristic of carrier confinement, but the features were very broad suggesting nonuniform well widths. Moreover, the positions of the features suggested a well width of roughly 250Å This latter result was confirmed by SIMS profiles.

As a final comment, we emphasize that useful photoreflectance spectra could not be obtained on many as-grown samples. Specifically, many spectra were dominated by semi-periodic oscillations in the modulated spectra so strong that they obscured all derivative-like features. We have seen these oscillations in both MQW's and MBE-grown thin films. We do not as yet understand the origin of these oscillations, but we remark that we have had limited success in suppressing them by specific surface and bulk processing.

Ultrafast Laser Sources and Applications

The principle objective of this task is to develop reliable sources of ultrashort optical pulses to be used in support of both HRL IR&D projects and externally funded contracts. Ultimately, we will amplify such pulses to energies sufficient for reliable broad-band continuum generation.

During the past year (in collaboration with colleagues at North Texas State University), we have developed a hybridly-modelocked (i.e., synchronously-pumped and passively modelocked) dye laser that has generated pulses as short as 55 fsec and has produced subpicosecond pulses from 560-975 nm by incorporating various gain and absorber dye combinations. The details of the cavity design are discussed in Appendices F-H in the 1986 AFOSR annual report, and they will not be repeated here. What we will describe here is the most recent results concerning the infrared operation of this laser using the gain dye Styryl 14.

Pure Styryl 14 is a photochemically stable infrared dye that is tunable from 940-1120 nm. It can be pumped with green light, which makes it compatible with synchronous pumping with the second harmonic of a cw modelocked Nd:YAG laser. The dye is also highly efficient (~10%) and thus offers the potential for relatively high output powers. There are numerous potential applications of this dye. Since it can be synchronously pumped with the frequency-doubled Nd:YAG, there exists the possibility for hybrid modelocking resulting in femtosecond pulse production near 1 μm. Since the dye can be tuned to 1.054 μm, there is the potential for using a Styryl 14 laser to seed high-gain Nd:glass amplifiers for high-power, subpicosecond, 1.054 μm pulse production. Also, since the operating wavelengths are well below the band gap of GaAs, there are potential applications in the areas of ultrafast photorefractive effects and electro-optic sampling. Finally, one would expect the peak powers produced by a synch-pumped Styryl 14 dye laser to be comparable to the peak powers obtained from a cw modelocked Nd:YAG laser. This implies that KTP can be used for efficient second harmonic generation of the dye laser output. Such a process would provide a source for tunable, picosecond, blue-green laser pulses.

Although there is clearly tremendous potential for this dye, unfortunately, it is not presently commercially available. Nevertheless, we have obtained experimental samples of the dye from Exciton, and we are working closely with them to encourage production of more Styryl 14. We have successfully synch-pumped this experimental sample of Styryl 14 resulting in the production of ~2 psec pulses over the tuning range 840-1040 nm. This tuning range overlaps with but does not reproduce the tuning curve of pure Styryl 14. Indeed, NMR analysis of our sample indicates that the dye is in fact a mixture of Styryl 14 and Styryl 9. Nevertheless, the tuning range of this

"contaminated" sample is useful in that it bridges the tuning gap between pure Styryl 9 and pure Styryl 14. These results demonstrate the first green-pumped cw modelocked dye laser to operate beyond 1 μm.

We have also hybridly modelocked this dye using three different polymethine cyanine dyes as saturable absorbers. Optimum results were obtained using 13-Acetoxy-1,1'-diethyl-2,2'-quinotetracarbocyanine iodide (DaQTeC). This hybrid laser produced pulses as short as 228 fsec at 975 nm. The saturable absorber 1,1'-diethyl-4,4'-quinotricarbocyanine iodide was used to produce 300 fsec pulses from 897-905 nm. Finally, 6,6'-dichloro-1,1'-diethyl-4,4'-quinotricarbocyanine was also used for hybrid modelocking and produced 480 fsec pulses at 972 nm. These are the longest wavelength femtosecond pulses ever to be produced directly from a hybridly modelocked dye laser.

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