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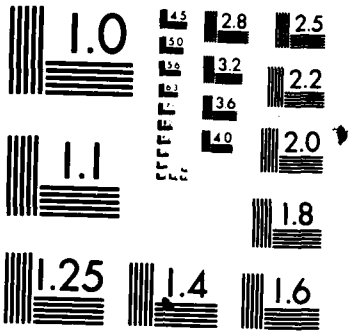

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This research was aimed at advancing understanding and utilization of selected optical properties of semiconductors containing magnetic elements. Emphasis was placed on the interaction of such materials with ultrashort pulses of laser radiation in order to study coupled electronic and magnetic excitations under selected nonequilibrium conditions. We hoped to generate novel results through experimental research for applications to fast optoelectronic devices. The mixed crystal semiconductors (Cd, Mn)Se and (Cd, Mn)Te were used. The contract work has generated a number of "firsts", e.g. we measured the formation of local, microscopic magnetically oriented "domains" through real-time spectroscopy with picosecond laser pulses.

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PICOSECOND NONLINEAR RESONANT INTERACTIONS IN SEMICONDUCTORS

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March 1986

1. Research Objectives and Summary

The objective of this phase of the research performed under AFOSR Contract F49620-82 C-0044 (1/1/84 to 12/31/84) was aimed at furthering our understanding and utilization of excitonic properties of semiconductors containing magnetic elements at a low temperature and under varying amount of impurity background. In particular, ultrashort pulses of laser radiation were employed to study localization phenomena of excitons under selected nonequilibrium conditions. The mixed crystal semiconductor (Cd,Mn)Te has provided the material basis for our work. In the short pulse experiments, photomodulated transient exciton spectra have been studied as a fucntion of the excitation photon energy in order to distinguish between localization by Coulomb centers (neutral and ionized impurities) and those originating from random potential fluctuations due to alloy compositional fluctuations. We have been succesfull in providing striking real-time evidence for the presence of localization by compositional fluctuations.

The research results derived from this AFOSR sponsored research have formed the basis of scientific publications, as enumerated below. In addition to regular scientific meetings, the principal investigator has been invited to present the research results in different scientific forums.

2. Research Accomplishments and Results

On the Question of Exciton Localization in (Cd,Mn)Te by Alloy Disorder

Many of the recent studies of optical properties in the II-VI diluted magnetic semiconductors (DMS) have involved near bandgap electronic excitations such as free and impurity bound excitons. At the same time there is an expected contribution to band edge broadening from alloy potential fluctuations which are inherent to any mixed crystal due to compositional disorder on microscopic scale. In particular, it has been realized some time ago that excitons may localize in such random potential wells at low temperatures (1). Experimentally, there have been reports in several mixed crystals of evidence for such localization, obtained through analysis of photoluminescence spectra (2), including recent transient spectroscopy (3).

In this segment of the accomplishments under the AFOSR supported research, we highlight results of time-resolved studies on subnanosecond timescale of excitonic spectra in $Cd_{1-x}Mn_xTe$ (x=.15) where picosecond laser techniques have been applied to look for evidence of exciton localization. The results are discussed below only in a qualitative way. Golnik and Lavallard have also performed similar work recently (4). Since $Cd_{1-x}Mn_xTe$ grows usually slightly p-type but with substantial compensation, one expects (neutral) acceptors and alloy fluctuations to compete with each other in the capture of free excitons injected to a sample. At higher impurity concentrations (but below the Mott transition) the formation of impurity bound excitons becomes the most likely event, as shown by us recently in n-$Cd_{1-x}Mn_xSe$ through time resolved spectroscopy

with the donor bound exciton (5). Localization by alloy potential fluctuations might take on additional significance in the DMS because of the possibility of a subsequent polaronic effect from the exchange interaction between the spins of the carriers and localized Mn-ion magnetic moments. Much of the recent experimental evidence for the 'bound magnetic polaron' (BMP) has come from studies of impurity bound exciton spectra (6). In that case, the electrostatic binding of a free carrier or a free exciton to an impurity complex provides the necessary initial confinement of the quasiparticle(s) in an effective Bohr volume.

Our experiments employed two picosecond dye lasers to excite and probe the $Cd_{1-x}Mn_xTe$ samples. We have used such photomodulation spectroscopy in several earlier occasions involving free and impurity bound excitons in semiconductors (7). One advantage of this approach over time resolved luminescence is the ability to distinguish spectral signatures between free and bound (localized) excitons. This follows from the distinct lineshape contribution which is made by exciton-exciton scattering to collisional broadening in such spectra for free excitons. Another benefit is the ability to apply resonant excitation without interference by scattered pump light in a noncollinear geometry with the help of suitable electro-optical modulation techniques (8).

In this work we concentrated selectively on $Cd_{1-x}Mn_xTe$ samples ($x=.15$) where cw-photoluminescence spectra did not show the strong emission characteristic of the neutral acceptor bound exciton (A^OX). (In samples where such A^OX emission dominates, we saw the large temperature dependent shifts in the emission energy, associated earlier with the BMP effect by Golnik et. al. (5).) Figure 1 shows luminescence spectra at three

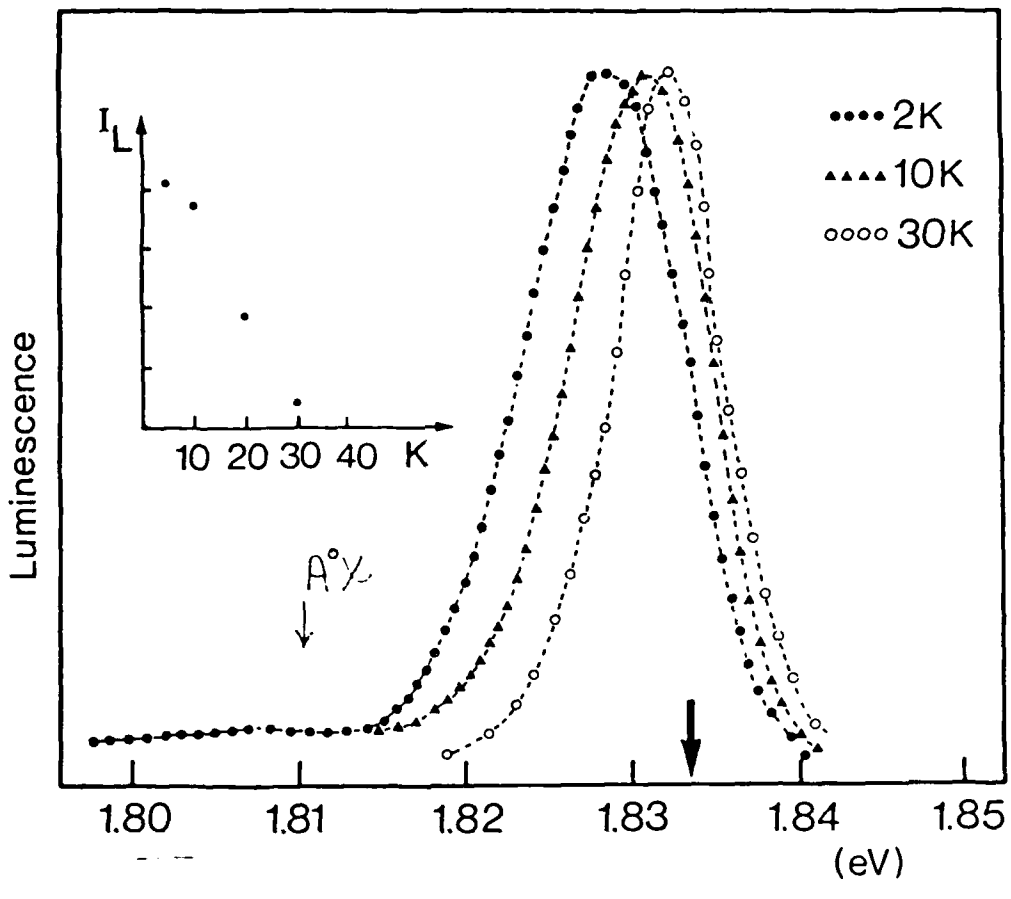

Figure 1: Photolumineacence in (Cd,Mn)Te (x= 0.15) at three different temperatures in the absence of a strong A^oX emission. The arrow indicates the position of the free exciton as determined from reflectance at T= 2K. The inset shows the actual temperature dependence of the luminescence amplitudes.

different temperatures, where the peaks are typically 10 meV and more above the known $A^{O}X$ energies. The free exciton energy as determined from reflectance at T=2K is also indicated, together with the temperature dependence of the amplitude. Note the blueshift of the peak energies with temperature and the narrowing of the spectra (the shift is, however, considerably less than that observed with the $A^{O}X$ emission). By T=30K the high energy side of the spectrum has a width comparable to kT. The spectra could suggest a (direct gap) material where close compensation has sufficiently reduced the density of neutral acceptor sites so that low temperature free exciton localization to near band edge states may have become possible. At the same time, nonexcitonic emission from other shallow impurity states is likely to add a contribution.

To test this possibility further, Figure 2 shows transient photomodulated spectra in this energy region, initiated by picosecond laser excitation in the same samples at T=2K. The absorbed photon density per pulse was maintained below 10^{15} cm^{-3}. The photon energy of excitation in Fig. 2a corresponds approximately that of the free exciton (from reflectance spectra) and that in Fig. 2b lies below it. Analysis of the photomodulation approach shows that the spectra are qualitatively those expected for a noninteracting exciton gas (in nonextended states), and can then be thought as analogous to time resolved photoluminescence. We see a pronounced difference between Figures 2a and 2b, with the rather narrow and distinct peak appearing at the photon energy of excitation in the latter. For photon energies of excitation varying over a range of nearly 10 meV below the free exciton energy such a peak was evident, tracking well the energy of excitation. At delay times t>0 a distinct low energy shoulder appeared, evolving while its spectral center of gravity moved to

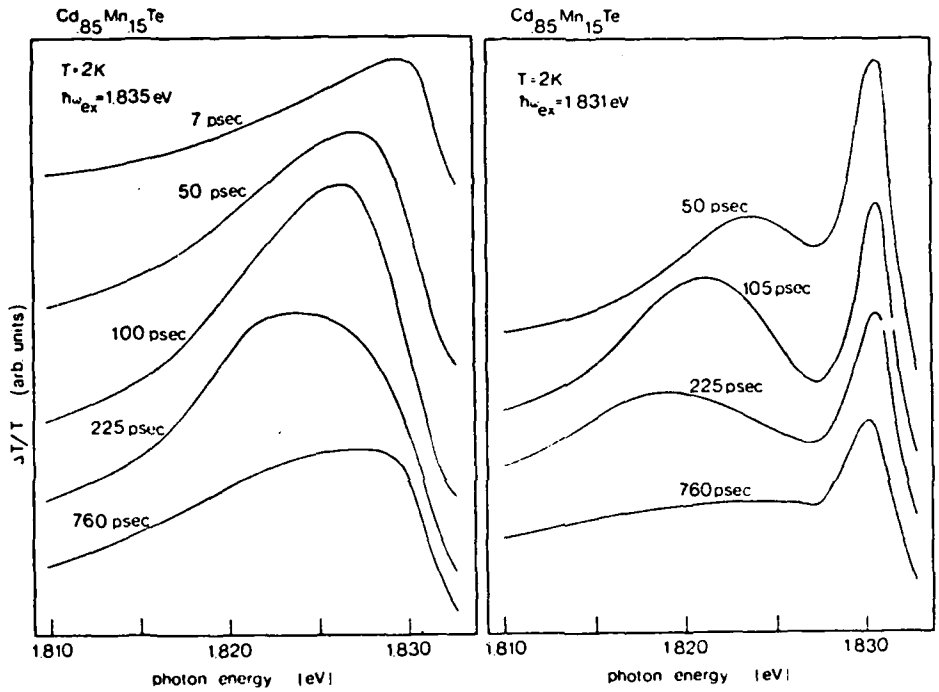

Figure 2: Transient spectra in x= 0.15 material at T=2 K below free exciton energy for two different energies of excitation: (i) at the free exciton energy (2a; left), and below the free exciton (2b; right).

lower energies. A time dependent shift is also seen in the broader nonresonantly excited features of Fig. 2a, following the free exciton injection at t=0. In all cases we also saw a long lived spectrally stationary 'background ' contribution which is apparent at the longest graphed delay times in Fig. 2. The lifetime associated with this background was typically many nanoseconds.

With increasing temperature, the peaks and their associated shoulders such as in Fig. 2b lost their distinctive shapes as well as decreased in amplitude when sample temperatures reached T=10 K. This change was also clearly seen through an increasingly rapid decay of the 'resonance' peaks (e.g. about 100 psec at T=5 K for the peak in Fig. 2b). At the same time, the long lived 'background' signals became relatively more dominant in this energy region, while at higher photon energies we saw spectral features which were suggestive of free exciton like behavior. The free exciton photomodulation spectra is usually distinguishable from the appearance of a sign change in the experimentally measured quantity dT/T originating from collision broadening of the exciton line (9). In our samples we also measured a substantial redshift in the tail of the absorption edge with increasing temperature. (For reference, at a value 100 cm^{-1} for the absorption coefficient, the redshift of the edge had an average value of 1.5 meV/K in the range of 2 to 20 K).

We interpret these results in the following way, while using qualitatively many of the ideas of Cohen and Sturge in Ref. 2. At the low temperatures, data such as in Fig. 2 is taken to imply the direct observation of localization of free excitons in alloy potential fluctuations. We can crudely estimate (10) the range of energies for such

localized states below the free exciton edge to be approximately 10 meV, if the Mn-ion distribution is statistical (there is uncertainty at present about the degree of clustering which may take place in this alloy). The relatively sharp structure which can be resonantly excited (Fig. 2b), is central to this argument, together with its disappearance at higher temperatures. The absence of a lower energy shoulder at t=0 and its appearance only at t>0 is interpreted as originating from acoustic phonon assisted tunneling to lower energy states in the continuum distribution of localized states (the details of such density of states are difficult to address from our data). An energy dependent tunneling cross-section together with a particular density of states can give rise to the appearance of a distinct shoulder as calculated in more detail by Cohen and Sturge (2).

In this interpretation, nonresonant excitation to the free exciton states above a 'mobility edge' (Fig. 2a) is followed by an initial capture to most of the localized states and subsequent energy relaxation within these states. From the time dependent data we infer capture rates typically on the order of 10^{11} sec^{-1} at T=2 K, slower than those seen earlier with impurity bound exciton formation (5). The low lattice temperature is unlikely to permit a significant thermal ionization and thus a multiple trapping like relaxation process should be less probable than the phonon assisted tunneling. From the observed spectral diffusion we extract tunneling rates which are on the order of 10^{9} sec^{-1} at T=2 K, i.e. comparable to the exciton lifetime (among the approximations made in this estimate is that of a constant optical matrix element within the localized states). Thus, a complete thermalization would not be achieved. At higher temperatures, the role of multiple trapping is expected to

increase the thermal equilibriation rate within the localized states as is qualitatively seen in our data. The amplitude of the cw-photoluminescence signal decreases rapidly at temperatures above 10 K suggesting a nonradiative trapping mechanism for free excitons. This behavior has also been confirmed earlier by us in time resolved experiments for the free exciton in $Cd_{1-x}Mn_xTe$ (11).

The evidence that free exciton localization is being directly seen in these experiments cannot be completely separated from the unavoidable effects of impuritites and other defects in the presently available $Cd_{1-x}Mn_xTe$. In particular, assuming a nearly compensated p-type (high resistivity) material implies that the position of the equilibrium Fermi level lies in the vicinity of the valence band edge at low temperatures. In fact, we suggest that E_F overlaps the valence bandtails which originate from the compositional disorder (the usual impurity/vacancy acceptor energy in CdTe is somewhat larger than the expected band edge smearing). With increasing temperature some thermal ionization of electrons into the unoccupied localized states takes place thus shifting E_F deeper into the gap and giving raise to the observed shift in the tail of the absorption edge. This also makes possible single electron (nonexcitonic) contributions in the photomodulated spectra, which follow the excitation of an electron from a defect state into the conduction band tail states. Any possible subsequent single carrier localization would be expected to result in longer lifetimes and is likely to be associated with the 'background' signal spectra observed by us.

In summary, we have presented experimental results and arguments that free exciton localization in alloy potential fluctuations can take place

in $Cd_{1-x}Mn_xTe$ when the competition by impurity bound exciton formation is not dominant. However, we have not yet seen any magnetic polaron-like effects under conditions of direct excitation to localized states although additional tests using an external magnetic field are under way. We also note that, in general, the real time spectral diffusion observed by us can also be expected from an inhomogeneously broadened impurity band. Finally, we would like to contrast these results with our recent work in n-$Cd_{1-x}Mn_xSe$, where transient spectra associated with neutral donor bound exciton was used to support arguments for the formation of an BMP (7).

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3. Scientific Publications and Presentations Resulting from the AFOSR Supported Research

1. "Kinetics of Free and Bound Excitons in Semiconductors", X.-C. Zhang, Y. Hefetz, and A. V. Nurmikko, in Ultrafast Phenomena IV, Springer Series in Chemical Physics, vol. 38, p. 176 (1984).
2. "On the Question of Exciton Localization in (Cd,Mn)Te by Alloy Disorder", X.-C. Zhang and A. V. Nurmikko", Proc. 17 Int. Conf. Physics of Semiconductors, North-Holland, p. 1443 (1985).
3. "Bound Exciton Formation and Excitonic Localization in Semimagnetic Semiconductors", A. V. Nurmikko, J. of Luminescence 30, 355 (1985).

In addition to these scientific publications, the AFOSR supported work has been presented in numerous scientific conferences.

1. Northeastern University, Physics Colloquium, May 1984.
2. Purdue University, Electrical Engineering and Physics Seminar, May 1984.
3. Symposium on Semimagnetic Semiconductors, Bad Honnef, Germany, June 1984.
4. Conference on High Excitation and Ultrafast Phenomena in Semiconductors, Trieste, Italy, July 1984.

4. Personnel

The following have had direct support from this AFORSR grant:

Professor A. V. Nurmikko, Principal Investigator.

William C. Goltsos, Ph.D. candidate.

5. Patents

No patents have been filed in connection with this AFOSR sponsored research.

6. Remaining Funds

No remaining funds exist.

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