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PICOSECOND NONLINEAR RESONANT INTERACTIONS IN
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ENGINEERING A V NURMIKKO MAR 86 AFOSR-TR-86-0528

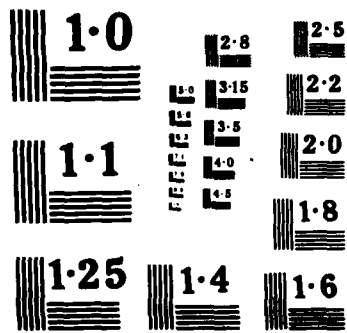
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PICOSECOND NONLINEAR RESONANT INTERACTIONS IN SEMICONDUCTORS

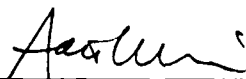
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Arto V. Nurmikko
Division of Engineering
Brown University
Providence, RI 02912

Report prepared by:


Arto V. Nurmikko
Professor of Engineering
Principal Investigator


Carl Cometta
Executive Officer
Division of Engineering

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<p>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.</p>			
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1. Research Objectives and Summary

The objective of the research performed under AFOSR Contract F49620-82 C-0044 (from 1/1/82 to 12/31/82) was aimed at advancing the understanding and utilization of selected optical properties of semiconductors containing magnetic elements. In particular, emphasis was placed on the interaction of such materials with intense laser radiation in order to study coupled electronic and magnetic excitations under selected nonequilibrium conditions. The mixed crystal semiconductors (Cd,Mn)Se and (Cd,Mn)Te have provided the material basis for our work. In this annual report we show how luminescence spectroscopy has yielded suggestions about the formation of local, microscopic magnetically oriented 'domains', the so-called bound magnetic polaron effects.

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



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2. Research Accomplishments and Results

The Exciton-Magnetic Polaron Effect in (Cd,Mn)Se

In this phase of the AFOSR supported research we have focussed on one of the more interesting effects encountered in the II-VI 'semimagnetic' semiconductors, namely the possibility of a finite magneto-optical contribution even at zero external magnetic field. Influence by the Mn⁺⁺-ion 3d electrons via an effective field on shallow impurities has been observed for donors in CdMnSe in spin-flip Raman scattering (1) and far-infrared spectroscopy (2), and for acceptors in CdMnTe in photoluminescence (3), (4). Theoretically, the inclusion of the exchange interaction of the impurity carrier with the Mn-ion spins within its orbit has been shown to lead to energy shifts and spin-splitting of the one-electron optical transition probability involving the impurity (5),(6),(1). These can originate from a net nonzero spin magnetization due to a trend toward local magnetic ordering within the Bohr volume of the carrier ('bound magnetic polaron') or a finite volume size effect which leaves a finite instantaneous magnetic moment even when thermal disorder dominates.

In the case of the wurtzite n-CdMnSe, recent work of Heiman, Wolff, and Warnock has clearly shown how the relatively large size of the donor orbit (>50Å) leads to a dominance of its zero field optical properties by the thermodynamic fluctuations even at a low temperature (6). For a photoluminescence event involving an exciton bound to the impurity, however, a question arises about the modification of the total exchange

interaction by the additional electron-hole pair. This is decidedly a difficult theoretical problem, but there is some experimental evidence to suggest that such effects may indeed be significant (7), (6). We have examined photoluminescence associated with the donor bound exciton in n-CdMnSe ($x=.10$ and $x=.05$) at zero external magnetic field and compared it with the same excitation in CdSe. In particular, we find that the presence of the Mn⁺⁺-ions affects a contribution to the binding energy and the linewidth of the bound exciton which is significantly larger than those observed for the 'bare' donor case (6). This is proposed to occur mainly as a consequence of a fairly temperature insensitive effective internal field within the relatively small exciton volume. We also find that the bound exciton luminescence spectrum for the $x=.10$ concentration shows a dependence on temperature which appears to be associated with a phase transition from an induced bound magnetic polaron regime. In our interpretation this is accompanied by large, inhomogeneously broadened spin-splittings which are reflected in anomalous spectral lineshapes and widths.

The experimental arrangement employed a cw frequency doubled Nd:YAG as a source of excitation at $h\nu=2.34$ eV. Photoluminescence spectra were obtained in a conventional way with samples mounted in a liquid helium immersion dewar. Figure 1 shows the dominant portion of the spectra in the near bandedge region, obtained for $x=.10$ samples at different temperatures. Somewhat unusually, however, strong reshaping and line narrowing occurs in the temperature range of 1.8 to 10 K. For comparison spectra for $x=.05$ and pure CdSe are shown in the inset. Care was exercised to provide a sufficiently low excitation (1mW) to avoid intensity dependent effects otherwise clearly evident. Additional spectral features

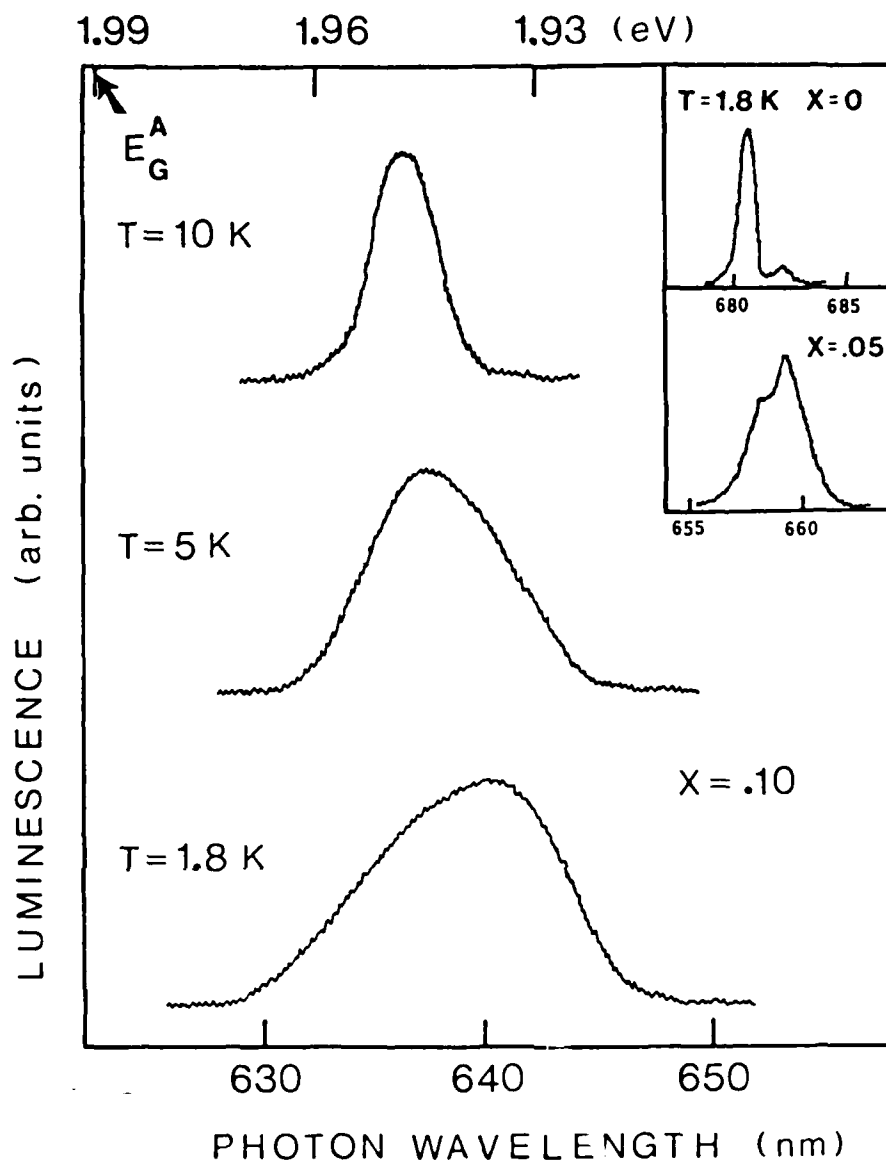


Figure 1: Low temperature luminescence spectra of (Cd,Mn)Se with $x=0.10$; E_G^A indicates the bandgap energy for this composition. In the inset, spectra taken for $x=0$ and $x=0.05$ at the lowest temperature are shown for comparison.

were observable at lower photon energies, attributed to donor-acceptor pair transitions. The spectral component shown in Figure 1 for $x=.10$ is identified by us as the zero phonon dominated line of a neutral donor bound exciton from the A-valence band for the samples containing approximately 3×10^{16} donors per cm^{-3} . It shows a clear connection with the similar dominant feature in the $x=.05$ and $x=0$ samples. In comparison with the $x=.10$ material, the feature in the $x=.05$ samples exhibited a more clearly evident two peak substructure where the relative amplitudes varied with temperature in a manner expected for thermal redistribution. Figure 2 summarizes the energetic position of the luminescence maxima as a function of temperature for the three compositions, referenced to the corresponding bandgap energy. Apart from lineshape asymmetries, the graphed quantity E_p is a measure of the average binding energy of the bound exciton in each case. As shown elsewhere, temperature variation of the bandgap and the free exciton energy in the range of interest here is rather small (8),(9). At higher temperatures ($T \geq 50\text{K}$) the bound exciton feature in CdMnSe showed increased broadening in a manner expected from acoustic phonon interactions. Throughout the low temperature range all the spectra remained fairly strongly linearly polarized (E p. c), in a manner seen for the A-exciton in CdSe.

These experimental observations show that the donor bound exciton luminescence in our CdMnSe samples implies a significant increase in both the binding energy of the exciton and its linewidth, when compared with CdSe. Furthermore, the $x=.10$ material shows anomalous temperature behavior where the lineshape strongly alters and narrows from approximately 2 to 10 K. Insofar as the spin-splitting and the linewidth of the neutral donor for CdMnSe ($x \leq .10$) in the same samples was measured to be only about 1

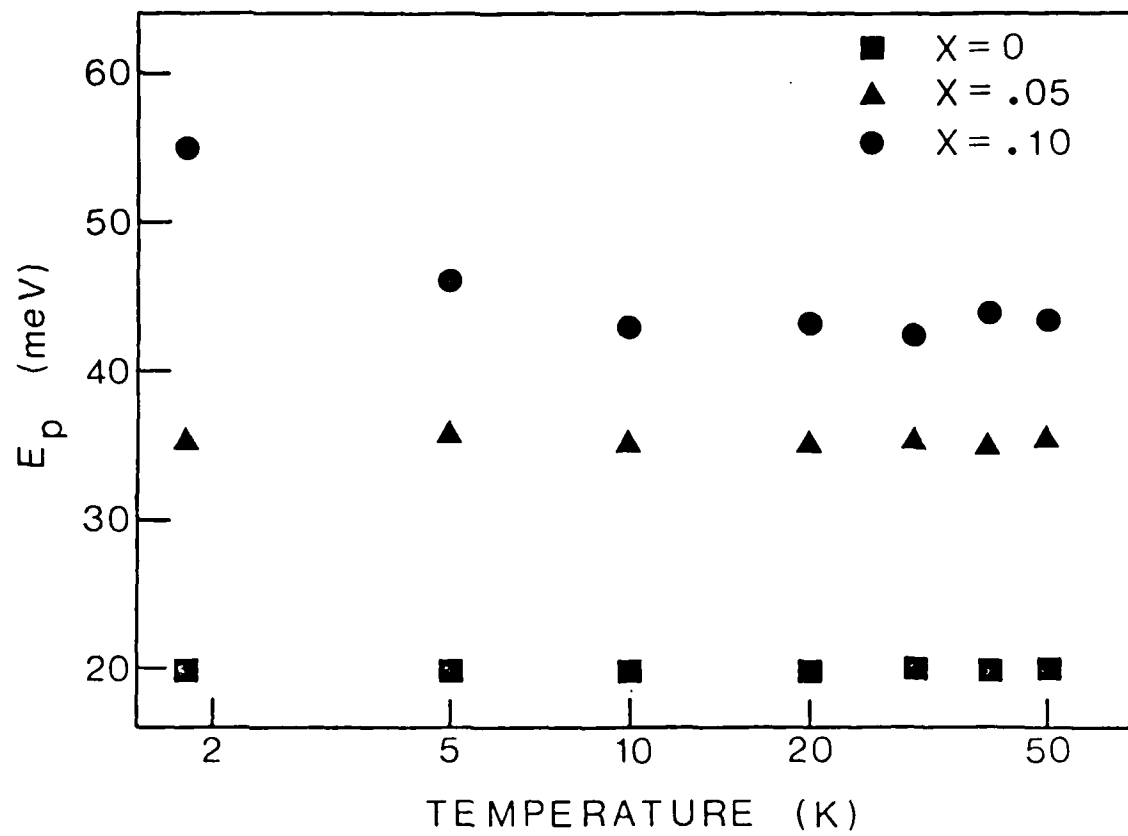


Figure 2: Energy position of the luminescence peaks as a function of temperature for three different compositions of (Cd,Mn)Se samples, reference to the corresponding bandgap energy in each case.

meV by spin-flip Raman scattering (6), we conclude that the initial exciton state is the dominant source of broadening in the luminescence event. It is well known that in nonmagnetic semiconductor alloys bound exciton lines acquire additional width from alloy potential (compositional) fluctuations. This follows from the relatively small effective radii of bound excitons. While such effects are likely to influence the bound exciton in CdMnSe as well, we believe that the major part of the observed energy shifts and broadening originates from the exchange interactions in the exciton-Mn⁺⁺ ion system. Analogous to the single particle impurity model which has wide experimental and theoretical support, both a bound magnetic polaron and a finite size magnetization effect need be considered within an effective Bohr radius of the exciton about the donor. In each case a substantial internal 'effective' magnetic field gives here rise to a spin splitting of the exciton. Alloy compositional fluctuations increase in importance for smaller radii, and lead to strong inhomogeneous broadening. Recent susceptibility measurements for CdMnSe indicate that for the compositional range considered here the magnetization remains paramagnetic at all temperatures (7),(8). The details of an inhomogeneous lineshape would, however, be strongly influenced by those remanent antiferromagnetic Mn-Mn interactions associated with elementary Mn-cluster formation.

These considerations lead us to suggest that for the $x=.10$ samples the initial strong temperature behavior indicates a phase transition from an exciton induced bound magnetic polaron regime to a fluctuation dominated one. The large linewidth (>30 meV) observed at 2K reflects the importance of alloy compositional fluctuations in the large internal effective field. In contrast, the $x=.05$ material remains in the fluctuation dominated

regime even at the lowest temperatures. In this regime, relatively temperature independent binding energy and linewidth are anticipated. Results similar to ours in magnitude have been recently reported by Nhung and Planel for a one-electron optical transition involving the neutral acceptor in CdMnTe (4). The expected spin-splitting into two components can be clearly seen in the $x=.05$ material while being somewhat obscured by a larger inhomogeneous broadening in the $x=.10$ samples. The origin of the broadening is due to the compositional fluctuations in the alloy and are therefore larger for smaller effective exciton radii. The inhomogeneous character of the lineshape has been also recently verified by studying energy diffusion in direct time-resolved experiments employing picosecond techniques (9).

An elementary estimate can be made about the expected size of the linewidth of the bound exciton in the fluctuation dominated regime, in the hydrogenic effective mass model. A sizable spin splitting with a broad linewidth is expected for an exciton radius of approximately 20 Å and a semi-quantitative agreement is obtained with the measurements. Such a zeroth order estimate provides a less satisfactory answer, however, with the transition to the bound magnetic polaron regime for the $x=.10$ material, evidently reflecting the fundamental complication associated with the multiparticle character of the exciton. The observation of polarized luminescence in accordance with selection rules for the A-exciton in CdSe implies that either these selection rules remain intact even in the rather high internal fields encountered (>100 kG) or there may be an 'easy' axis of magnetization for spontaneous alignment of the net local moments.

In summary, photoluminescence spectra associated with the neutral donor bound exciton in CdMnSe shows a significant enhancement in its binding energy and linewidth in comparison with the previously studied bare donor. This is attributed to increase in the exchange interaction with the Mn⁺⁺-ion spins within the exciton orbit. Apart from a general trend, significant questions remain concerning the details of the electron-hole configuration in the magnetic polaron regime.

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- (9) J. H. Harris and A. V. Nurmikko, Phys. Rev. B28, 1181 (1983)

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

1. "High Resolution Picosecond Modulation Spectroscopy of Near Interband Resonances in Semiconductors", S. Sugai, J. H. Harris, and A. V. Nurmikko, in Picosecond Phenomena III, Springer Series in Chem. Physics. vol. 23, p. 103 (1982).
2. "Transient Polarized Modulated Reflection Spectra of Near Bandedge Excitations in $Cd_{.70}Mn_{.30}Te$ ", A. V. Nurmikko, S. Sugai, and J. H. Harris, Proc. Int. Conf. Physics of Semic., Montpellier, Physica B117, 494 (1983).
3. "Magnetic Polaron Contribution to Donor Bound Exciton in $(Cd,Mn)Se$ ", C. A. Huber, A. V. Nurmikko, M. Gal, and A. Wold, Solid State Comm. 46, 41 (1983).

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

1. Conference on Progress in Quantum Electronics, Snowbird, Utah, Jan. 1982.
2. Conference on Mercury-Cadmium Telluride, Minneapolis, Feb. 1982.
3. General Motors Research Labs, Physics Department Colloquium, Detroit, March 1982.
4. Max Planck Institute for Solid State Physics. Colloquium, Stuttgart, Germany, June 1982.
5. American Physical Society, Southeastern Section, Lexington, KY, Oct. 1982.
6. IBM Research Laboratories, Physics Colloquium, Yorktown Heights, NY, Nov. 1982.

4. Personnel

The following have had direct support from this AFORSR grant:

Professor A. V. Nurmikko, Principal Investigator.

Bradley Schwartz, 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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