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Distinct exciton-polaron resonance in the infrared edge emission of semimagnetic $Hg_{1-x}Mn_xTe$

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Results of a study of the 10-μm wavelength edge emission from narrow-gap $Hg_{1-x}Mn_xTe$ in magnetic fields are presented. Under moderate magnetic fields ($B=1.0$–2.5 T) a strong resonance enhancement of the emission and a narrowing of the linewidth to less than 2 meV, smaller than we are aware has been reported for spontaneous emission from a narrow-gap material, were observed. These observations were interpreted based on a theoretical model suggested by Karpov and Tsidil'kovskii in which the appearance of small gaps within the tails of the density of electron states in diluted magnetic semiconductors due to magnetic polaron formation is predicted. The observed resonance, based on this model, is tentatively interpreted as the coincidence between the energetic positions of this gap and the free-exciton line position, which is widely tunable by applying a magnetic field in narrow-gap materials. This is experimental evidence for the formation of a free exciton-polaron complex and the influence of excitonic effects in narrow-gap materials.

The presence of "tail states" within the energy gap E_g of a semiconductor can arise from disorder effects and is usually empirically described by a broadening energy E_0, which gives the exponential slope of the so-called Urbach tail. Since a variety of microscopic mechanisms contribute to the broadening, this tailing can also be a function of external parameters such as temperature or magnetic field. Recently, theoretical investigations have been reported in which a peculiarity of the broadening mechanisms in semimagnetic semiconductors was predicted. Karpov and Tsidil'kovskii[1] found that under well-defined circumstances a polaron effect can produce an energy range with a reduced density of states (DOS) and thus open an additional gap within the tail of the DOS at energies slightly below E_g. This effect was predicted for the absence of a magnetic field. However, it can be expected in a modified form even in the presence of a weak magnetic field.

Generally excitonic effects are not often considered on the analysis of spectroscopic data of narrow-gap materials having E_g values of about 0.1 eV, such as $Hg_{0.8}Cd_{0.2}Te$ or $Hg_{0.9}Mn_{0.1}Te$. This is justified by the small oscillator strength for lower E_g values resulting from the scaling of the exciton binding energy E_x. However, it is known that a magnetic field significantly increases the energy of a hydrogenlike system, such as an exciton, by reducing the dimensions of the quasiparticle. The increasing E_x causes not only a direct enhancement of the oscillator strength of a free exciton but also results in a "tuning" of the line position of the excitonic ground-state emission to lower energies.

Under well-defined experimental conditions the interplay of both effects—the presence of a gap within the band tails as well as the exciton as a "tunable probe"—can cause a resonance. This paper reports the observation of such resonance in the 10-μm-wavelength photoluminescence (PL) emission of the semimagnetic $Hg_{1-x}Mn_xTe$. A full width at half maximum (FWHM) of the main emission line of about 2 meV was observed and is to the best of our knowledge the narrowest spontaneous emission from a narrow-gap semiconductor ever reported. It should be noted that experimental evidence on magnetic-field enhancement of PL and absorption structures in II-IV materials has been reported.[2–8] In narrow-gap material, Ivanov-Omskii *et al.*[2] assigned this effect to a free-exciton enhancement in $Hg_{0.7}Cd_{0.3}Te$. Galazka *et al.*[3] as well as Gelmont *et al.*[4] reported this effect in terms of acceptor-related processes. For wide-gap materials, Gaj[5] summarized data on magnetic-field-dependent excitonic luminescence measurements in $Cd_{1-x}Mn_xTe$ and mentioned—in addition to the enhancement—a destabilization of excitonic complexes in higher magnetic fields and consequently a decreasing PL signal of this particular process. Wolff[6] discussed changes in the magnetic-field-dependent excitonic luminescence in the same wide-gap material and discussed the conditions for the formation of free magnetic polarons.

None of the previous studies, however, report that ultranarrow structures such as those reported here have ever been observed in the 10-μm edge emission of a semiconductor.

A set of $Hg_{1-x}Mn_xTe$ wafers was cut from two crystals grown by the traveling heater method. The samples were as-grown n type with a carrier concentration and mobility of 3×10^{15} cm^{-3} and 5×10^{4} cm^2/(V s) at 77 K. In this paper we present the data for only two of the samples, which have nominal MnTe mole fractions of $x=0.095\pm0.003$ and 0.104 ± 0.003, as determined by x-ray microprobe analysis. The corresponding E_g values at $T=5$ K in the absence of a magnetic field are 105 ± 2 and 132 ± 2 meV, respectively. The mole fraction difference between the samples was confirmed by the obtained PL data. The sample surfaces were etched in standard bromine-methanol solution (3%) before measurements.

Infrared PL results were determined using an apparatus consisting of an Oxford Spectromag 4000-7T magnetocryostat and a Bruker IFS 88 infrared Fourier-transform spectrometer. Both systems were coupled using appropriate mir-

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ror optics. The optical excitation ($E=1.55$ eV, power density ~ 1 W cm^{-2}) was provided by an external diode laser module employing a double modulation technique with phase-sensitive excitation and detection. Reference measurements were also carried out with a Nd:YAG (yttrium aluminum garnet) laser [$E=1.7$ eV, excitation density ~ 10 W cm^{-2} (cw) and ~ 100 kW cm^{-2} (Q switched)].

Figure 1(a) depicts PL spectra of a $Hg_{0.896}Mn_{0.104}Te$ sample at a magnetic field of $B=2.25$ T for various temperatures. This particular B field was chosen because the PL signal magnitude was the largest at $T=5$ K, as demonstrated in Fig. 1(b), while having a minimum FWHM. Figure 1(c) depicts the temperature-dependent PL signal magnitude and illustrates the strong enhancement of the excitonic line at low temperatures if a magnetic field is present. Considering the spectral position of the emission peak ($h\nu$) versus temperature (not displayed here), we found for $T>20$ K that it follows roughly the $E_g(T)$ dependence, whereas for lower temperatures the slope increased by a factor of 4 to about $d(h\nu)/dT=1$ meV/K for $B=2.25$ T. For zero field this increase was significantly less pronounced. The second PL feature visible in Fig. 1(a), which is separated by about 15 meV towards lower energies, is still under investigation and is believed to be impurity related. The same investigations were made for the sample with $x=0.095$ and the data obtained confirmed the aforementioned tendencies. The magnetic field, where the PL shows the resonance narrowing at $T=5$ K, amounted to $B=1.1$ T. For this set of external parameters a FWHM of the PL line of 2.2 meV was observed.

The experimental results are now interpreted within the framework of a resonance between the exciton emission, the spectral position of which is tuned by the magnetic field, and the "tail gap" created by the exchange interaction.

All of these experimental data indicate that the observed narrow PL line is indeed edge emission, and not impurity related. Consistent with the assignment for excitation-power-dependent measurements towards higher-densities band filling and a blueshift interpreted as dynamic Burstein-Moss effect were observed. The observation of a saturation of the PL signal magnitude of the lower-energy line, on the other hand, confirmed the thesis that the line is impurity related. Furthermore, the magnetic-field dependence of the PL peak position was analyzed for both Faraday and Voigt geometry. Consistently, we found a remarkable fit within the framework of a modified Pidgeon-Brown model with free-exciton corrections under the assumption of realistic exchange ($N_0\alpha=0.9$ eV, $N_0\beta=-0.45$ eV) and clustering ($S_0\approx 1$, $T_0=9\pm 1$ K) parameters. These results confirm the interpretation of the line as edge emission and they are understandable in the framework of known theoretical concepts for narrow-gap semimagnetic zinc-blende semiconductors. This will be presented in detail later.[9] A third experimental indication for the nature of the narrow emission comes from the $h\nu(T)$ dependencies mentioned above. For wider-gap $Hg_{1-x}Cd_xTe$, Lusson, Fuchs, and Marfaing[10] interpreted comparable dependencies as thermal delocalization and dissociation of intrinsic (free) excitons trapped in potential fluctuations. Our findings are in agreement with their assignment but the microscopic mechanism seems to be different. Furthermore, the lowering of the slope $d(h\nu)/dT$ for zero field is in agreement with a reduced exciton binding energy expected for zero field.

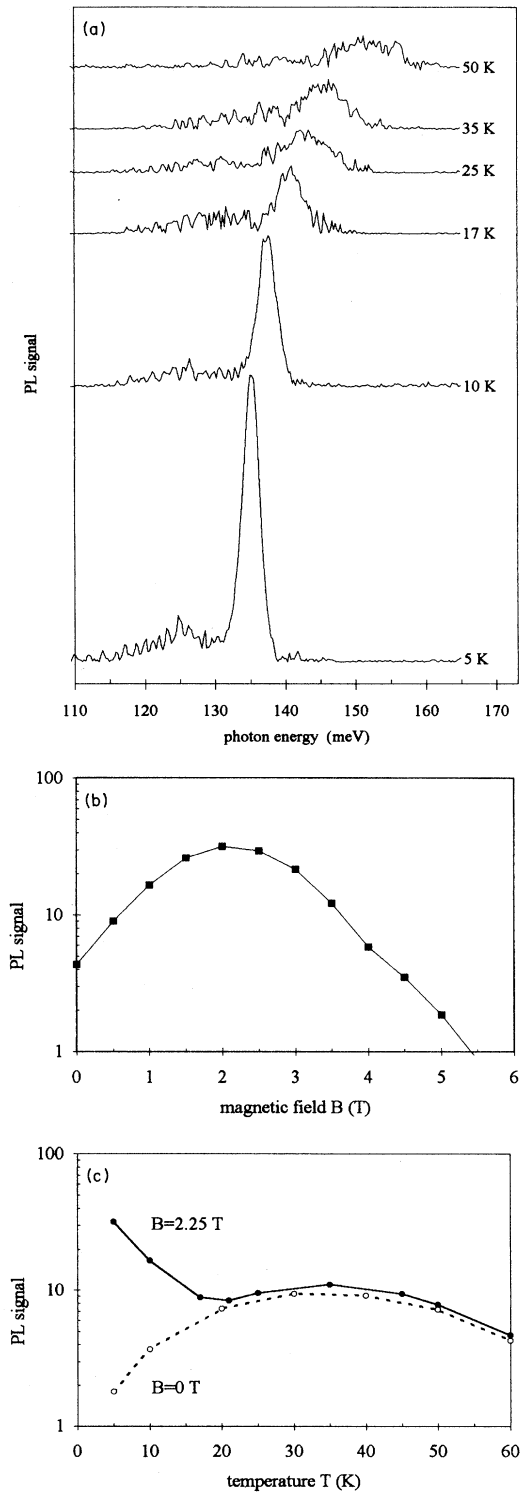

FIG. 1. PL of $Hg_{0.896}Mn_{0.104}Te$ sample. (a) PL spectra vs temperature for a fixed magnetic field of $B=2.25$ T for various temperatures. (b) PL signal vs B at $T=5$ K. (c) PL signal magnitude vs temperature for $B=0$ (empty circles) and $B=2.25$ T (full circles).

How relevant are exciton and polaron effects in narrow-

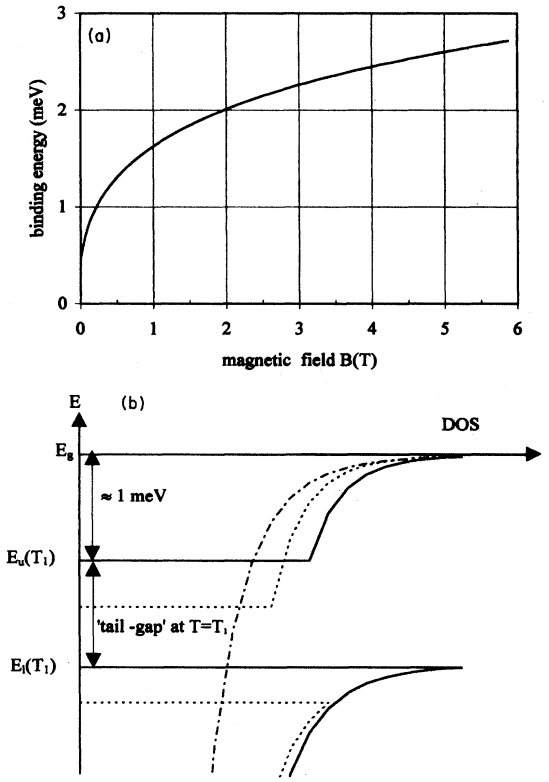

FIG. 2. (a) Free-exciton binding energy vs magnetic field for $Hg_{0.896}Mn_{0.104}Te$ at $T=5$ K. (b) Schematic diagram of the DOS in the energy region slightly below E_g in a diluted magnetic semiconductor according to Karpov and Tsidil'kovskii (Ref. 1). The dashed-dotted line depicts the usual Urbach tail, whereas the full (T_1) and the dotted line (T_2) illustrate the tail gap with $T_2>T_1$. The energetic distance between E_g and $E_u(T_1)$ was estimated to be on the order of 1 meV.

gap semimagnetic materials? Figure 2(a) depicts the exciton binding energy as a function of the magnetic field for the free exciton in $Hg_{0.896}Mn_{0.104}Te$ using relevant sample parameters (static dielectric constant $\varepsilon_0=17$, electron mass $m_e=0.01$, and effective heavy-hole mass $m_{hh}=0.55$) calculated according to Ekardt.[11] Note that the strongest B dependency occurs for $B<2$ T, and a value of nearly 3 meV is reached for $B=7$ T. This number is not very different from the known 4.3 meV in GaAs at $B=0$, and there excitonic effects dominate the edge emission at low temperatures. Taking into account only the binding-energy criterion, there are obviously good chances for the observation of excitonic effects in our samples.

Figure 2(b) qualitatively describes the tail gap in the DOS below E_g of the semimagnetic material, according to Karpov and Tsidil'kovskii,[1] for uncorrelated disorder. The dashed-dotted line represents the usual tail within a single band-state density, whereas the full and the dotted curves indicate the zero-field DOS modified by exchange interaction (magnetic polaron) for two different temperatures T_1 (full line) and T_2 (dotted line) with $T_2>T_1$. Note that for the given parameters the spacing between E_g and the upper edge of the tail gap $[E_u(T)]$ can be estimated[1] by

$$E_g-E_u(T)=\tfrac{1}{4}E_0(kT/E_0)^2 \quad (1)$$

with

$$E_0=[\alpha^4x^2(1-x)^2m^{*3}]/(178h^6N^2), \quad (2)$$

where m^* is the effective mass, N is the concentration of lattice sites containing either Hg or Mn atoms, and $\alpha=(dE/dx)$. Within the limits given by the accurate knowledge of the parameters we get a value for $E_g-E_u(T_1=5\text{ K})$ on the order of 1 meV [cf. Fig. 2(b)].

It must be noted that the magnetic field also influences the tail gap. It is expected that both edges merge to lower energies and finally the tail gap gets quenched since all spins are aligned in the external field and thus the polaron effect vanishes.

Considering $E_x(B)$ [cf. Fig. 2(a)] and the DOS scheme in Fig. 2(b), the resonance of the free-exciton emission at intermediate fields becomes clear. For zero field, the excitonic contribution is small since the binding energy is comparably small $[E_x(0)\approx 0.5$ meV] and the exciton is further broadened since its spectral position at E_g-E_x is within the tail. For intermediate fields, E_x grows at a faster rate with increasing field [cf. Fig. 2(a)] than the decreasing rate of E_u. Thus the excitonic line moves into the tail gap. Both increasing binding energy and the reduced broadening within the tail gap cause the resonance. For a weak magnetic field of about 1–2 T the probability for hole-related magnetic polaron formation is weaker than at 0 T. However, it is still possible[9] and thus it can occur together with the exciton. Hence the free exciton can be accompanied by a free-hole-related magnetic polaron, and these quasiparticles form a free-exciton-polaron complex.[7] Of course the formation of this new quasiparticle results in a further energetic lowering and thus enhances the chance to observe a PL resonance. For higher fields, E_x grows more slowly and the tail gap vanishes due to the quenching of the polaron effect in the external field.

The unusually sensitive temperature dependence of the PL in the presence of a B field illustrated by Figs. 1(a) and 1(c) now also becomes qualitatively clear: Besides the thermal dissociation of the free excitons the tail gap moves [cf. the dotted and full line in Fig. 2(b)] and reaches the exciton line. Note that the temperature dependence of the PL signal magnitude for zero fields agrees very well with the known behavior for it in $Hg_{1-x}Cd_xTe$ (cf. Ref. 12) and the strong excitonic enhancement occurs only in the presence of a B field.

In summary, we have reported the existence of a well-defined set of external (B,T) and material parameters (E_g, exchange interaction), which can cause a distinct excitonic resonance to occur in narrow-gap materials emitting in the 10-μm wavelength region. This paper gives experimental verification for this *region of existence* and also provides a qualitative microscopic explanation for the effect observed. This paper can also be understood as a report on the optical detection of a free-exciton-polaron complex in a narrow-gap semimagnetic material.

Besides the unique situation with respect to physics, a certain applicative relevance can also be expected since the critical magnetic fields for the appearance of the resonance

amount to 1.1 and 2.25 T for $Hg_{0.905}Mn_{0.095}Te$ and $Hg_{0.896}Mn_{0.104}Te$, respectively. These values can easily be reached with permanent magnets in very simple experimental configurations.

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