Giant effective *g*-factor in $Pb_xEu_{1-x}Te$ epitaxial films

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We investigated $Pb_xEu_{1-x}Te$ films with $x \leqslant 0.2$ by magneto-optical measurements. For $x \sim 0.01$, the optical emission is similar to high quality EuTe films with two narrow lines attributed to excitonic recombinations associated with magnetic polarons. For increasing x, the emission becomes dominated by a broader lower energy band, which is very efficient as compared to the binary emission. The magneto-optical properties of the ternary films show various similarities with EuTe results, such as quenchings at similar temperatures and magnetic fields. Most remarkably, they also present a giant effective *g*-factor that makes this material a strong candidate for spintronic applications. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2961018]

EuT is a magnetic semiconductor belonging to the europium chalcogenides family that has been extensively investigated since the 1960s.[1] At room temperature EuTe is paramagnetic, while below 10 K, it becomes antiferromagnetic ordered due to the exchange interaction between the magnetic moments of the Eu^{2+} ions with half-filled $4f$ levels. EuTe has band gap energy of 2.25 eV at 10 K.[2] Early studies of EuTe samples grown by traditional methods only presented a broad emission band at relatively low energy (~1.5 eV).[3] More recently, Heiss *et al.*[4–6] observed two additional narrow emission lines at higher energies on high quality EuTe epitaxial layers. The lines labeled MX_1 (1.92 eV) and MX_2 (1.88 eV) were attributed to excitonic recombinations associated with magnetic polarons (MPs).

The ternary compound $Pb_xEu_{1-x}Te$ is miscible over the whole alloy range. Its band gap energy can thus be varied in a rather wide range between the band gap energies of the binary semiconductors: 0.30 eV (PbTe) and 2.25 eV (EuTe) at 10 K.[7] Much attention has been devoted to ternary compounds with high Pb contents due to their applications for far-infrared devices.[8] Few works have been focused, however, on the low Pb limit despite its potential for spintronic devices. In this work we investigate the magneto-optical properties of $Pb_xEu_{1-x}Te$ alloys with $x \leqslant 0.20$. The films present a very efficient optical emission band with magneto-optical properties that are quite similar to high quality EuTe layers, whereas its peak can be varied between ~1.74 and ~1.92 eV. We present the results of photoluminescence (PL) measurements as a function of temperature and applied magnetic field. Based on those results, we discuss the origin of the optical emission that dominates the PL spectra from the low Pb content alloys.

The samples were grown by molecular beam epitaxy in a RIBER 32P system with Eu, Te, PbTe, and BaF_2 sources. Freshly cleaved BaF_2 (111) substrates were used. The substrates were heated up to 150 °C for 15 min in the preparation chamber and to 360 °C for 30 min in the main growth chamber. Two different substrate temperatures were used, 145 and 175 °C. $Pb_xEu_{1-x}Te$ films were usually covered with a BaF_2 layer to prevent oxidation. The film growth rate was ~1.5 Å/s in all cases, as estimated from scanning electron microscope micrographies. We investigate a series of $Pb_xEu_{1-x}Te$ films with Pb contents ranging from 0 to 0.20 and thicknesses varying between 100 and 400 nm. High resolution x-ray measurements revealed an increase in the full-width at half maximum (FWHM) from the rocking curves corresponding to the (222) Bragg peak as x was increased. For samples grown at 175 °C, the FWHM increased from 260 arc sec for EuTe to 670 arc sec for the sample with $x = 0.2$. All samples grown at 145 °C present a larger FWHM of ~700 arc sec The results show that both the Pb incorporation and the decrease in the growth temperature tend to degrade the crystalline quality of the epitaxial films.

Magneto-PL measurements were performed in the Faraday configuration for magnetic fields up to 12 T. The samples were photoexcited with the 488 nm line of an Ar^+ laser focused in a ~150 μm diameter spot. The optical emission was analyzed using a Jobin-Yvon monochromator and a Si charge coupled device detector.

Figure 1 shows the PL spectra at 2 K from a series of samples grown at 175 °C with different Pb compositions. The PL spectra were normalized by the intensity of the higher energy emission band from each spectrum. We observe the EuTe characteristic excitonic bands, MX_1 (1.92 eV) and MX_2 (1.88 eV), in the PL spectrum of our binary film, indicating a high quality crystal. The MX lines can still be observed for the sample with $x=0.01$, but they vanish for larger Pb contents. The films with low Pb compositions, including the binary sample, also present a broad emission band at ~1.5 eV, which is usually attributed to defects. This defect-related band abruptly disappears for samples with $x > 0.02$. Conversely, we observe the emergence of an additional emission band in between the excitonic and the defect-related bands, which develops to become the dominant emission for samples with $x \geqslant 0.02$. The additional emission, which we will label as PbRB (Pb related band) shifts to lower energies and broadens up with increasing x. The PbRB emission from the $Pb_{0.02}Eu_{0.98}Te$ film has a FWHM of ~130 meV centered at ~1.81 eV, while for the $Pb_{0.20}Eu_{0.80}Te$ film, the FWHM becomes ~230 meV, and its

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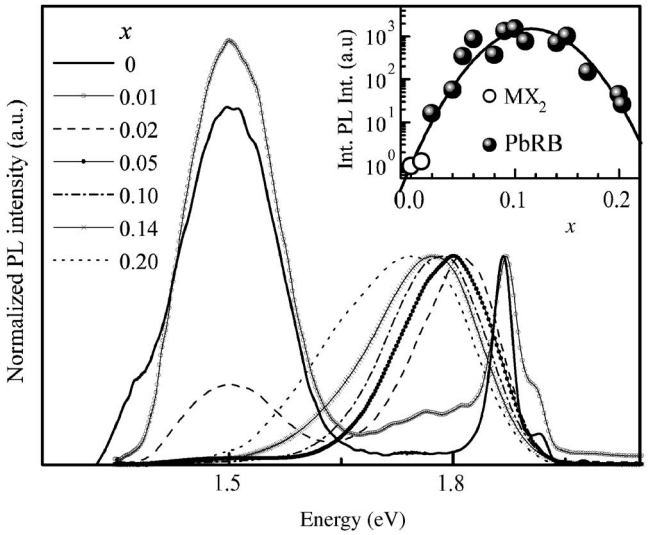

FIG. 1. PL spectra at 2 K from Pb_xEu_{1-x} Te epitaxial layers with different values of x normalized to the intensity of the higher energy emission band. The inset shows the integrated PL intensity of the PbRB and the MX_2 emission bands vs Pb content.

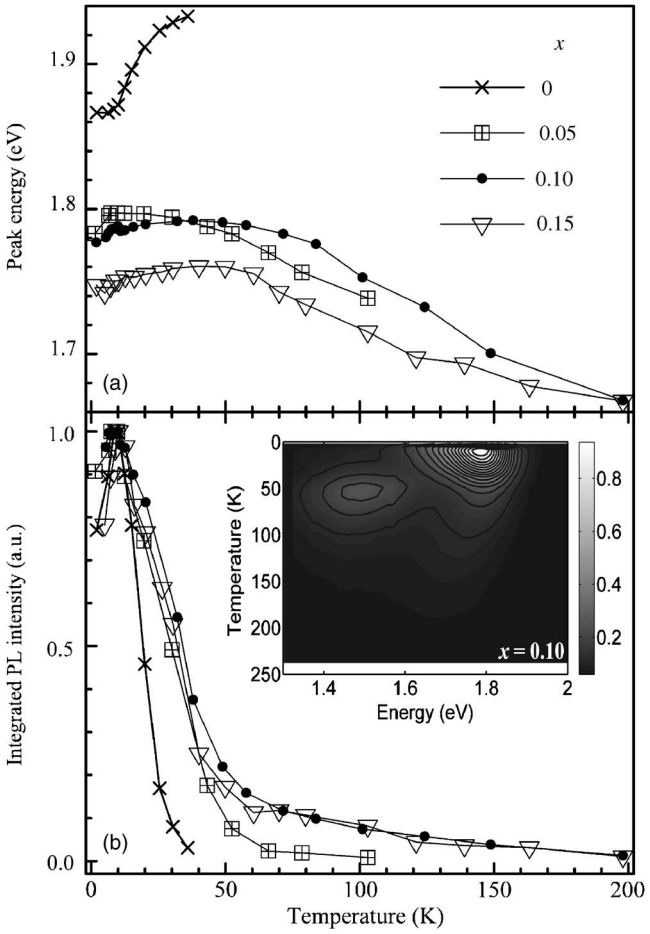

FIG. 2. (a) Energy of the PL peak and (b) integrated intensity as a function of the temperature for various films with different Pb contents. The inset shows a PL map of the emission from the $Pb_{0.10}Eu_{0.90}Te$ film vs energy emission and temperature.

peak appears at ~1.74 eV. The integrated PL intensities of the MX and PbRB bands are presented in the inset of Fig. 1 as a function of x. The integrated PL intensities of the PbRB emission are remarkably strong as compared to the integrated intensity of the MX lines measured under identical conditions. The PbRB emission shows a maximum at $x \sim 0.10$, where its integrated intensity becomes approximately a thousand times larger than the MX band from the EuTe film. $Pb_xEu_{1-x}Te$ films grown at 145 °C exhibit a similar behavior as that presented in Fig. 1. In this case, however, the MX lines are not observed for any film composition, which is in agreement with the poorer crystal quality as observed by x-ray measurements. This result indicates that the MX EuTe-like emission lines are more sensitive to the crystal quality of the film than the recombination associated to the PbRB.

The results concerning the temperature dependence of the PL emission are presented in Fig. 2. The inset shows the emission from the $Pb_{0.10}Eu_{0.90}Te$ film as an intensity map where the horizontal and vertical axes correspond, respectively, to energy and temperature. The PL peak position and integrated intensity from the MX/PbRB band for various films with different compositions are presented, respectively, in Figs. 2(a) and 2(b) as a function of temperature. The energy of the MX band from the EuTe film shows a characteristic kink at ~10 K followed by a sudden blueshift [Fig. 2(a)]. This behavior has been observed before, and it was attributed to the breaking of the MP due to the magnetic-phase transition of the film from the antiferromagnetic to a paramagnetic phase, where the blueshift corresponds to the MP binding energy.[4] The quenching of the PL intensity observed at the same temperature range was interpreted as an evidence that the MX recombination is optically forbidden but that it becomes allowed due to the MP interaction, which mixes states with different wave momenta.[4] The PbRB emission presents a very similar behavior concerning its integrated PL intensity, as shown in Fig. 2(b). The integrated intensities from the PbRB bands present a clear maximum at ~10 K, followed by an abrupt decrease. Contrary to the EuTe result, we observe a residual PL intensity from the ternary films for $T > 50$ K, which slowly decreases but is still observable up to ~200 K. As the intensity of the PbRB emission band is quenched with increasing temperatures, the defect-related emission band (~1.5 eV), which was not discerned at 2 K for films with $x > 0.02$, becomes observable (see inset in Fig. 2). Concerning the peak energy behavior, the PbRB band also presents a blue shift (~15 meV) between 10 and 40 K. This blueshift is smaller than the ~60 meV blueshift observed for the EuTe film at the same temperature range, which may be partially related to the relatively larger FWHM of the ternary bands, but it is still in contrast to the expected decrease in band gap energy with increasing temperatures. For temperatures above 50 K, the residual PbRB emission band does present a redshift following the band gap dependence.

Figure 3 shows a summary of the magneto-PL results at 2 K. The inset presents a typical intensity map of the $Pb_{0.11}Eu_{0.89}Te$ film as a function of energy and magnetic field. The peak energy and integrated intensity from the MX/PbRB emission band from various samples are presented, respectively, in Figs. 3(a) and 3(b). All samples present a similar and strong dependence on magnetic field. The intensities of both the MX_2 and the PbRB emission bands present a maximum at ~0.5 T, followed by a significant decrease with increasing fields up to ~7–8 T, above which we only observe a residually small and mainly constant PL intensity. In the case of the binary film, the decrease

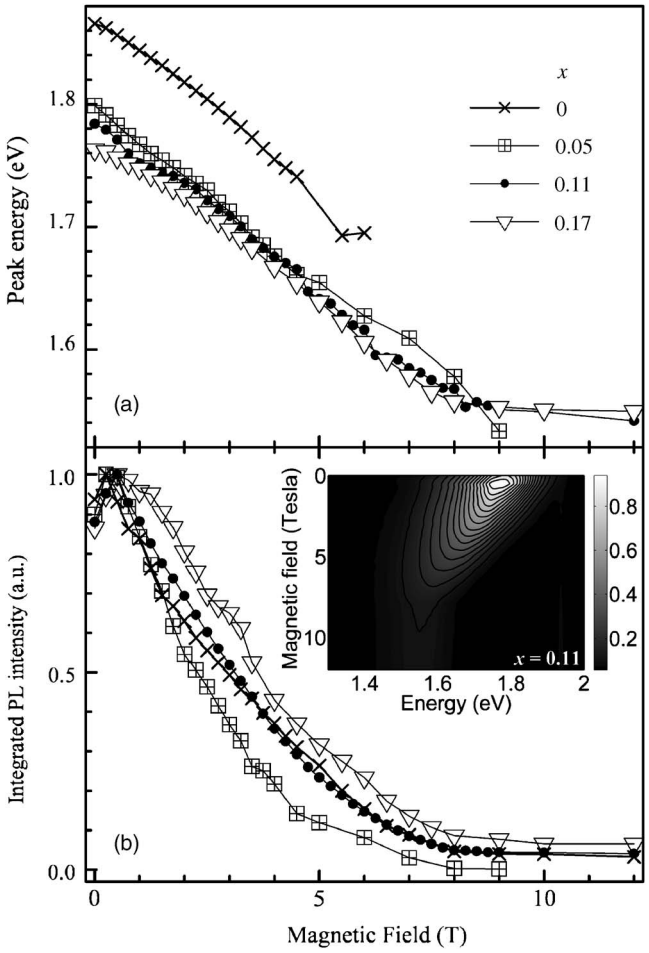

FIG. 3. (a) Energy of the PL peak and (b) integrated intensity as a function of the applied magnetic field for various films with different Pb contents. The inset shows a PL map of the emission from the $Pb_{0.11}Eu_{0.89}Te$ film vs energy emission and magnetic field.

in the PL intensity with magnetic field was attributed to the magnetic-phase transition from an antiferromagnetic to a ferromagnetic ordering induced by magnetic fields larger than the critical field of ~7.2 T, which also results in the quenching of the MP binding energy.[9] The most remarkable result is that not only the EuTe film but all the ternary films show an impressively large redshift with magnetic field. The total redshift amounts to values up to ~250 meV for 7 T. The dependence of the peak energies on magnetic field is approximately linear for fields up to ~7 T. The redshift rate decreases with increasing Pb content, from a maximum value of ~30 meV/T for $x=0.02$, to ~20 meV/T for $x=0.20$. The corresponding effective g-factors obtained for those limiting values using the relation $g_{\text{eff}}=2\Delta E/(\mu_B \Delta B)$ are, respectively, 1040 and 690. The values are comparable to the giant effective g-factor previously reported for high quality EuTe films (34 meV/T and $g\sim1140$).[6]

In summary, the PbRB emission bands from films of PbEuTe ternary alloys with low Pb contents show important similarities to the excitonic EuTe recombination. The ternary films as well as the binary sample show an abrupt intensity quenching for temperatures above the antiferromagnetic-paramagnetic Neel transition temperature (~10 K) and for magnetic fields larger than the critical field (~7–8 T) that generates a complete ferromagnetic alignment of the Eu^{+2} spins. Furthermore, the ternary films also show a giant effective g-factor, comparable to that reported for EuTe films. Based on those properties, it has been proposed that the MX recombination from EuTe films should be related to MPs. The formation of this complex breaks the selection rules, increasing the probability of the otherwise forbidden excitonic recombination, so that the unbinding of the MPs results in a drastic decrease in the PL emission.[4] Therefore, our results indicate that the PbRB emission from our ternary films may also be related to MPs. On the other hand, the ternary emission band is affected by the amount of Pb incorporated to the film. The PbRB band shifts to lower energies and broadens as the Pb content increases, while its intensity reaches a maximum at an ideal ~10% Pb concentration. This increase in intensity is also accompanied by the quenching of the low-energy defect-related emission band. This result indicates that the presence of Pb atoms increases the stability of the MP related to this emission, which may be attributed to localized-MPs by alloy fluctuations with larger binding energies.[10] The fact that we only observe a residual PbRB emission at large temperatures and magnetic fields for ternary films but not for the EuTe film may be interpreted as an indication that the localized MP from the ternary films are not completely forbidden, as the EuTe MX recombination.

Briefly, we investigated the magneto-optical properties of $Pb_xEu_{1-x}Te$ epitaxial films. The incorporation of low contents of Pb presents the advantage of band gap tuning, maintaining a very efficient optical emission and a giant g-factor, comparable to that observed for EuTe. Those properties make those ternary films especially interesting for spintronic applications. Based on our results, we propose that the main PL emission from ternary films is associated with localized-MPs.

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