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Effects of Pb doping on the magneto-optical properties of EuPbTe epitaxial films

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ABSTRACT

We investigate the magneto-optical properties of magnetic-semiconductor $Eu_{1-x}Pb_xTe$ epitaxial layers with Pb contents up to 5%. We show that the inclusion of a small amount of Pb atoms in EuTe affects the optical and magnetic properties of the resulting alloy. The incorporation of Pb gives rise to a reduction of the Néel temperature and of the slope of the giant magneto-red-shift of the magnetic polaron optical emission. All those effects can be understood in terms of the magnetic dilution effect due to the reduced Eu concentration. The introduction of Pb also reveals a splitting of the high emission energy side-band under applied magnetic field, presenting a more complex feature of the band structure of the alloys. Our results cannot be fully explained on the basis of the current theoretical knowledge of the EuTe band structure and, therefore, we expect that they can stimulate future theoretical investigations and encourage applied investigations of spintronic devices based on these materials.

1. Introduction

Magnetic-semiconductors show singular magnetic and semiconductor properties that can be combined to design spintronic devices [1]. An advantage of such systems compared to diluted-magnetic-semiconductors is that the exchange interaction effect is significantly stronger in the former due to the higher density of magnetic ions. In this context, EuTe with an optical emission in the visible spectral range is a promising material for such applications. This material presents an anti-ferromagnetic phase below the Néel temperature (9.6 K) and a paramagnetic phase above it. The high quality of EuTe epitaxial layers grown by Molecular Beam Epitaxy (MBE) on BaF_2 substrates have lead to investigations of the magnetic and the optical properties of this material [2–5]. Even though some works have been reported on $Eu_{1-x}Pb_xTe$ alloys [6–8], there is still little information concerning the details of the magnetic and optical properties, as well as the band structure of these compounds.

The optical emission from EuTe layers is usually dominated by the magnetic polaron effect involving the transition of electrons localized at the X-point (minimum of the conduction band) and 4f localized states. This emission band, usually denominated MX, is ~350 meV below the observed optical absorption edge from this material [2,4]. Recently, Heredia et al. [9] have observed a higher energy emission band (~ 60 meV above the MX band) when EuTe samples were excited with high excitation intensities (> 1 kW/cm^2). This emission band, named HE, was attributed to another conduction band valley with slightly higher energy than the X-point. Therefore, high excitation intensities are required in order to observe this optical emission. In contrast to the MX emission, the HE band is less sensitive to the magnetic phase transition and persists at higher temperatures as compared to the MX band [9].

In this work, we investigate the optical and the magneto-optical properties of $Eu_{1-x}Pb_xTe$ epitaxial layers with x up to 5%. We observe that besides the alloys maintain the MX and HE lines, even a rather small Pb content, it gives rise to a reduction of the Néel temperature as a consequence of decreasing of the magnetic Eu content. In contrast to pure EuTe, the HE line from the alloys shows a clear splitting with increasing magnetic fields. Furthermore, the saturation field to order Eu dipoles and the slopes of the typical MX and HE red-shifts with the magnetic field are reduced with increasing Pb content.

2. Experimental details

The $Eu_{1-x}Pb_xTe$ epitaxial layers were grown by MBE on BaF_2 (111) substrates with x varying from 0% to 5%. The Pb contents correspond to the nominal growth values. More details on their

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epitaxial growth and structural properties are presented in Ref. [8]. Photoluminescence (PL) measurements were performed in a magneto-cryostat using a 488 nm laser beam focused by a 50 × objective for excitation. The emission was analyzed by a single monochromator coupled to a Si-CCD. The magnetic field was applied parallel to the growth direction and varied up to 10 T. The sample temperature was varied from 4 to 300 K.

3. Results and discussion

Fig. 1 shows typical PL spectra for our set of samples under high excitation intensities (8 kW/cm^2). The HE (~1.98 eV) and the MX (~1.91 eV) lines are observed for all samples, except the $x=5\%$ sample, where the MX is hidden by a low energy broad emission band. This broad band is attributed to Pb localized states. It arises with increasing Pb content and becomes dominant for $x\sim5\%$ [8]. The HE band is only observed at high excitation intensities (larger than ~1 kW/cm^2) [9], while the MX band is observed for any excitation intensity [2–5]. The MX band typically dominates the emission from high quality EuTe epitaxial layers grown by MBE, as reported previously by several authors [2–5], and it is attributed to the recombination of magnetic polarons involving the transition of an electron at the X-point minimum of the conduction band (see Fig. 4 of the Ref. [4]) and a 4f localized state [4]. Since the HE band corresponds to a higher energy transition, the electron is at a higher energy valley of the conduction band. Consequently, the electron associated to this band may have shorter lifetime, for instance, 10 times smaller, as compared to the MX band [8], due to rapid relaxation of the electrons to the conduction band minimum at the X-point. This explains why the HE band is only observed under a high excitation regime.

The temperature evolution of the peak energy of the MX and HE bands is very distinct, as shown in Fig. 2. With increasing temperatures, the MX band shows a strong and abrupt blue-shift around the EuTe Néel temperature ($T_N\sim10$ K) while the HE band presents a rather smooth U-like temperature dependence.

Concerning the MX band, all samples show a very similar behavior. The large blue-shift can be explained by the temperature annihilation of the magnetic polarons [10] that dominate the PL emission bellow T_N. These complexes are a result of the exchange interaction between the conduction band and 4f Eu states. We notice that the blue-shift onset moves to smaller temperatures with increasing values of x. For $x=5\%$, this tendency cannot be confirmed due to the broad emission band that hides the MX band. Since the MX blue-shift was previously associated to T_N [2–4], we can use our results to estimate T_N for the alloys. Based on the well established value of T_N for EuTe (9.6 K) when the MX line shifts to 1.924 eV, we consider that this energy also presents the threshold of the magnetic transition for the other samples. We

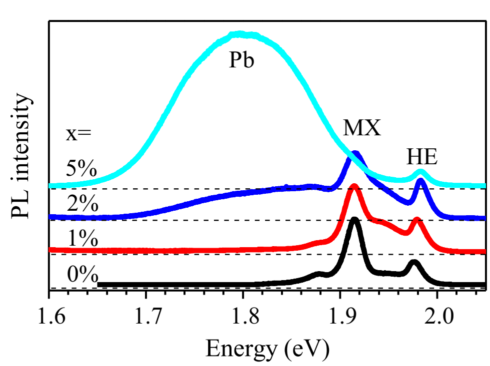

Fig. 1. PL spectra of $Eu_{1-x}Pb_xTe$ epitaxial layers (x= 0–5%) under high excitation intensity (8 kW/cm^2) at 5 K.

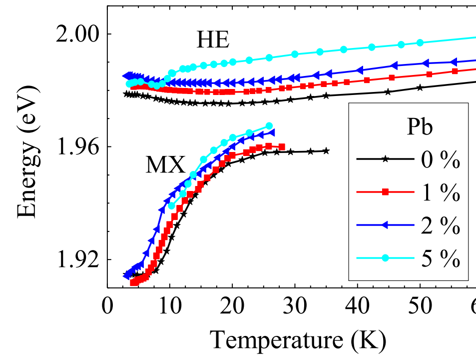

Fig. 2. Temperature dependence of PL peak energy of the MX and HE emission bands.

thus estimate T_N values of 8.5 K for $x=1\%$ and 6.6 K for $x=2\%$. The result reveals a reduction of the Néel temperature with increasing Pb contents, which is consistent with the reduced density of Eu magnetic ions. Similar magnetic dilution effects were observed by introducing other nonmagnetic atom, such as Sr in EuTe [11,12].

On the other hand, the HE band only shows a small red-shift around T_N, which is rather surprising due to the energy proximity of the conduction band valleys associated with the HE and the MX bands [4,13]. Possible explanations are that the carriers involved in such transition do not give rise to stable magnetic polarons [9], which may be associated to rather shorter emission decay time than that for the MX band. Another explanation is the compensation effect, where the blue-shift is canceled by a red-shift of the conduction band valley with increasing temperature as observed in absorption measurements of the optical band-gap edge [4]. Once more, the 5% sample presents a distinct behavior. In fact, this sample shows a clear and rather abrupt blue-shift of the HE line around T_N, surprisingly similar to the MX band with a smaller energy scale. The different behavior observed in the HE band for temperature around T_N is under current investigation.

Fig. 3 shows the evolution of the PL spectra with applied magnetic field (B) for samples with $x=0$ (Fig. 3(a) and (b)) and 5% (Fig. 3(c) and (d)). Both the MX and the HE emission bands present large red-shifts with B as observed in Figs. 3(a) and (c), respectively, which is also another signature of magnetic polaron effects [4,5,9]. The spectra are normalized but the intensities of the bands decrease with B, as previously observed for EuTe layers [4,5,9]. We also observe an unexpected splitting of the HE emission in two lines (HE_{low} and HE_{high} in Fig. 3(c)), which is more evident for the alloy samples. In order to further investigate this point, we also measured the circular-polarized components of the PL emission (Fig. 3(b) and (d)) using a linearly polarized light for excitation. For the 5% sample, the doublet is clearly observed even at ~0.5 T (Fig. 3(d)). We point out that the relative intensity between the HE_{high} and HE_{low} peaks on Fig. 3(d) is different than that of Fig. 3 (c). This is due to the different excitation intensity used in both experiments. The different intensities of the two circular-polarized components reveal a small spin-polarization for both states of the doublet. On the other hand, for pure EuTe, the doublet is not resolved (Fig. 3(b)). Nevertheless, when a magnetic field is applied to the 0% sample the spin polarization is smaller but we observe a slight asymmetry and broadening of the HE band that is consistent with the splitting of the 5% sample. The result suggests that the HE emission encompasses more than one conduction band valleys and this degeneracy can be broken by a magnetic field. The effect is stronger by the presence of Pb ions.

We also point out that the effective spin splitting at 1 T for both lines of the HE doublet from the 5% sample are negligibly small (see Fig. 3(d)). However, the origin of the non-zero spin