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Magnetic Field Induced Absorption in Pb$_x$Eu$_{1-x}$Te Magnetic Semiconductors

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We report an investigation of the optical absorption spectrum, using non-polarized light, in Pb$_x$Eu$_{1-x}$Te, x=0 and x=0.095, epitaxial thick layers grown by molecular beam epitaxy (MBE). The absorption edge is described by a broad band, due to the electronic transitions from the $4f^7$ of Eu^{2+} to the states in $4f^6 5d$ configuration, as seen previously in bulk Eu chalcogenides. When a magnetic field is applied, a narrow absorption band (full width $\sim$50 meV) emerges from the broad one. The energy of this absorption peak red shifts when the magnetic field increases, and reaches saturation when the Eu^{2+} attain ferromagnetic arrangement. This behaviour can be described by a localized excitation model with $d - f$ exchange interaction.

1 Introduction

Europium telluride is a classical MnO-type antiferromagnet, whose magneto-optical properties have been intensely investigated in the past [1–4]. As in other europium chalcogenides, the optical absorption edge is described by a broad band, which is associated with transitions from the $4f^7(^8S_{7/2})$ ground state of Eu^{2+} to the $4f^6(^7F_J)5d(t_{2g})$ configuration. In the excited state, the electron and the hole left at the Eu-site form a bound system that extends over several lattice parameters [5]. The excitation thus formed is denominated *magnetic exciton*, due to the characteristic exchange interaction (the $d - f$ interaction) between the electron and the Eu^{2+} spins located inside the exciton sphere. More recently, the interest in EuTe and other magnetic semiconductors has been renewed, because of their potential for optical devices controlled by a magnetic field [6], and the interest in the science and technology of spin-dependent phenomena in semiconductors [7]. Also, the fabrication processes of europium chalcogenides were improved over the last years, which has allowed investigators to obtain samples of unprecedented purity. Recently, Heiss *et al* [8] reported the observation, for the first time in EuTe, of sharp photoluminescence lines at energies above 1.9 eV. This photoluminescence is very sensitive to applied magnetic field, resulting in a giant effective g-factor of 1140 [8]. The samples studied in Ref.[8] were grown by molecular beam epitaxy (MBE), and the new observations were attributed to the high purity of the samples.

In this work we report on an investigation of the absorption spectrum in a magnetic field of EuTe and Pb$_x$Eu$_{1-x}$Te grown by MBE on the [111] crystal direction over a BaF$_2$ substrate. For a magnetic field applied in the Faraday configuration we observe that a sharp line emerges from the $4f^7 \rightarrow 4f^6 5d$ absorption band in the low energy side of the spectrum. This line shows a red shift as the magnetic field increases and the antiferromagnet enters the canted phase. The red shift is saturated when complete spin alignment is achieved. The red shift of the absorption line is in good agreement with a simple theoretical model of an excitonic transition with $d - f$ exchange interaction. Early investigations of the reflectivity of single crystals of EuTe grown from the melt have detected a splitting of the spectrum in a magnetic field, which was attributed to the ferromagnetic exchange splitting of the $5d(t_{2g})$ final state [1], although no quantitative theoretical modeling of the red shift was presented by the authors.

2 Results and discussion

We studied thick layers (thickness 1-2 μm) of Pb$_x$Eu$_{1-x}$Te on BaF$_2$, with nominal values of x = 0 and x = 0.095, grown by MBE. The samples were grown on the [111] crystalline direction. The optical transmission through the samples was measured at 1.5 K under magnetic fields up to 9.6 T (Figs. 1 and 2). The monochromatic light source was a 48 W tungsten lamp connected to a SPEX270M spectrometer. The light was conveyed to the sample, and collected from the sample, using optical fibers, and the intensity of the transmitted signal was measured using a photomultiplier. The direction of the incident light and the direction of the magnetic field were both perpendicular to the epitaxial layers.

At B=0 the absorption spectra of the samples revealed a broad band due to the $4f^7 \rightarrow 4f^6 5d$ electronic transitions localized at the Eu ions [1–4]. When Pb is introduced into the sample in small concentrations, the absorption peak is detected at approximately the same energy (Fig. 2).

When a magnetic field is applied, the absorption edge red shifts, as observed in other works [1, 2, 8]. At B$\sim$ 6T
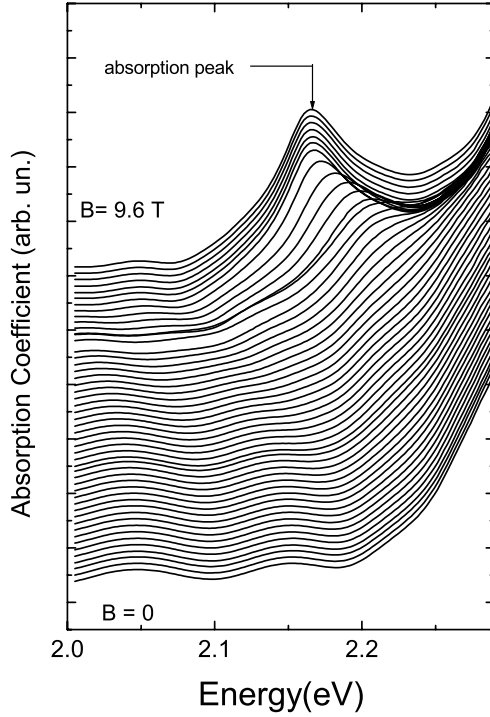
Figure 1. Absorption coefficients of EuTe sample (sample 790) measured applying magnetic field at temperature of 1.5 K.
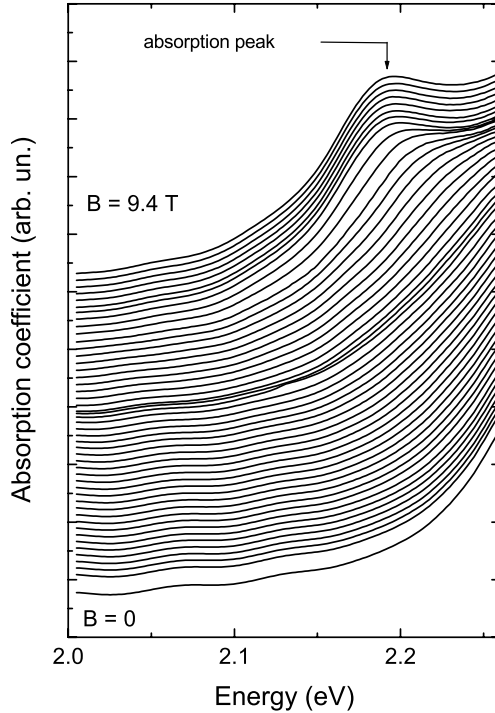

Figure 2. Absorption coefficients of $Pb_{0.095}Eu_{0.905}Te$ sample (sample 853) measured applying magnetic field at temperature of 1.5 K.

and above a sharp absorption peak emerges from the broad absorption band in EuTe and $Pb_{0.095}Eu_{0.905}Te$ (Figs. 1 and

2). Between 6 and 8 T the emerging peak shifts to lower energies, reaching a maximum displacement of 50 meV at the saturation field of about 8 T when the material becomes ferromagnetic [9]. At fields above 8.3 T no further changes are observed in the position or magnitude of the emerging absorption peak. For the geometry of our experiment (a thin layer under perpendicular magnetic field) an applied field of 8.3 T corresponds to an internal field of 7.2 T, which is the well known critical field intensity for EuTe [9]. Figs. 1 and 2 show that the net shift of the emerging peak is smaller in the $Pb_{0.095}Eu_{0.905}Te$ sample, in comparison to the EuTe one.

Next we show that the newly observed absorption can be explained in the framework of the magnetic polaron model, used in Ref. [5] to describe the photoluminescence spectrum of EuTe. In this model, the energy of the excitonic emission is described using the effective mass approximation. For the absorption spectrum, according to the Franck-Condon principle, the energy position of the absorption lines can be obtained from the same model, but excluding the lattice relaxation that characterizes the emission process. In this case, the energy shift in a magnetic field of the absorption line will be mainly determined by the $d - f$ interaction:

$$\mathcal{H}_{d-f} = -(1 - x)\, J_{df} \sum_{n,\mu,\nu} a_{n\mu}^{\dagger} a_{n\nu}\ \vec{S}_n \cdot \vec{\sigma}$$

where $a_{n\nu}^{\dagger}$ is the creation operator for a Wannier function located at site $\vec{R}_n$ with spin ν, $\vec{\sigma}$ is a Pauli spin operator, $\vec{S}_n$ is the spin of the n−th europium site and J_{df}=40 meV [10] is the $d - f$ exchange interaction constant.

We treat H_{df} as a perturbation over a photoexcited electron in a d−band with definite spin, whose wave function is given by $\Psi = \sum_n c_n a_{n\chi}^{\dagger}|0>$. Assuming that the excitation extends over several lattice parameters, a first-order perturbational treatment of $\mathcal{H}_{d-f}$ leads to the following magnetic field dependence for the peak position of the line:

$$E = E_0 - (1 - x)\, J_{df}\, |\vec{S}| \left\{ \begin{array}{ll} \left(\frac{B}{B_c}\right)^2 & B < B_c \\ 1 & B \geq B_c \end{array} \right. \quad (1)$$

where B_c is the saturation field.

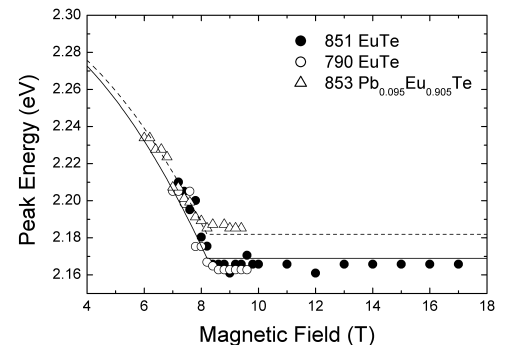Figure 3. Absorption peak positions taken from Fig. 1 and Fig. 2. Solid and dashed lines were obtained using Eq. (1) for x=0 and x=0.095, respectively.

Figure 3 shows the peak positions of the absorption line, and the theoretical curve (1). The theoretical curves shown in Fig. 3 were obtained assuming $|\vec{S}| = \frac{7}{2}$, $E_0 = 2.305$ eV, i.e. the absorption peak position at $B = 0$ (see Fig. 1), and $B_c = 8.2$ T, the saturation field value. The excellent agreement between theory and experiment, obtained without any fitting parameters, demonstrates that the newly observed absorption peak is associated with an electronic transition that splits from the $4f^7 \rightarrow 4f^6 5d$ band, red shifted due to the $d - f$ exchange interaction. The observation of a new absorption line in our samples is attributed to the growth procedure (MBE) used by us, which probably leads to samples of higher purity and homogeneity than in previous investigations.

Acknowledgments

References

[1] J. Feinleib, C. R. Pidgeon, Phys. Rev. Lett. **23** 1391 (1969).

[2] P. Wachter and P. Weber, Solid State Commun. **8**, 1133 (1970).

[3] J. Schoenes, J. of Magn. Magn. Mater. **11** 102 (1979).

[4] T. Kasuya, J. of Magn. Magn. Mater. **195**, 141 (1999).

[5] M. Umehara, Phys. Rev. B **52**, 8140 (1995); M. Umehara, J. of Magn. and Magn. Mater. **187**,177 (1998); M.Umehara, Phys. Rev. B **65**, 205208 (2002).

[6] Y. Ohno, D. K. Young, B. Beschoten, F. Matsukura, H. Ohno, and D. D. Awschalom, Nature **402**, 790 (1999); S. A. Crooker, d. A. Tulchinski, J. Levy, and D. D. Awschalom, Phys. Rev. Lett. **75**, 505 (1995).

[7] *Special issue on semiconductor spintronics*, edited by Hideo Ohno, Semicond. Sci. Technol. **17**, Number 4, (2002).

[8] W. Heiss, G. Prechtl, and G. Springholz, Appl. Phys. Lett. **78**, 3484 (2001); W. Heiss, G. Prechtl, G. Springholz, Phys. Rev. B **63**, 165323 (2001); W. Heiss, *Habilitationsschrift zur Erlangung der Lehrbefugnis für das Fach*, Univerität Linz, Linz (2002).

[9] N. F. Oliveira, Jr., S. Foner, Y. Shapira, and T. B. Reed, Phys. Rev. B **5**, 2634 (1972).

[10] T. Kasuya, A. Yanase, and T. Takeda, Solid State Commun. **8**, 1543 (1970).