TEMPERATURE DEPENDENCE OF ENERGY GAP OF HIGHLY CONCENTRATED $Cd_{1-x}Mn_xTe$ $(0.6 < x \leq 1.0)$ EPILAYERS

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The temperature dependence of the energy gap of MBE grown $Cd_{1-x}Mn_xTe$ $(0.6 < x \leq 1.0)$ was measured for $2\ \mathrm{K} \leq T \leq 200\ \mathrm{K}$ and $B \leq 5\ \mathrm{T}$. The results are interpreted in the frames of the model predicting that the exchange contribution to the band edge shift is proportional to the product of the magnetic susceptibility and the temperature.

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The energy gap variation with composition and temperature in diluted magnetic semiconductors (DMS) [1] is strongly affected by s, p–d exchange interaction, which leads to large shifts of the conduction and valence band edges even in the absence of a magnetic field. These effects were observed in bulk CdMnTe [2–4], ZnMnTe [5], ZnMnSe [6,7] and CdMnS [8]. The exchange contribution to the energy gap reflects a correlation between spins of the magnetic ions [9, 3, 7] and, therefore, it is particularly important in concentrated alloys possessing magnetically ordered (spin-glass or antiferromagnetic) phases [1]. However, bulk materials of single crystallographic phase are available only in a limited concentration range ($x < 0.75$ in the case of CdMnTe [1], and $x < 0.80$ for ZnMnTe [1]). This means that effects associated with AF–SG (antiferromagnetic–spin-glass) transition cannot be easily investigated in the bulk. Recent progress in growing techniques of diluted magnetic semiconductors (MBE) enabled fabrication of materials in the entire concentration range, i.e., up to $x = 1.0$ [4, 10]. In this communication we report on the energy gap variation with the temperature in $Cd_{1-x}Mn_xTe$ epilayers with Mn concentration ranging from $x = 0.6$ to $x = 1.0$ (i.e. in cubic MnTe).

The samples used for the present study were grown by MBE technique as a few microns ($< 5\ \mu$m) thick epilayers of cubic $Cd_{1-x}Mn_xTe$, on GaAs substrate. The manganese concentration was $x = 0.66, 0.73, 0.85$ and 1.0 (MnTe), as determined from the epilayer lattice constant. We measured the reflectance in the free exciton range for $2\ \mathrm{K} \leq T \leq 200\ \mathrm{K}$ and magnetic fields $B \leq 5\ \mathrm{T}$. In most cases the excitonic structures were not visible in a standard reflectance experiment, irrespective of the applied magnetic field, most probably due to large exciton line width

and small modulation of total reflectance. On the other hand, the degree of circular polarization of reflectance ("magnetic circular dichroism", MCD), defined as

$$P = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^+)} \tag{1}$$

(where $I(\sigma^+)$ and $I(\sigma^-)$ are intensities of the reflected light in σ^+, σ^- circular polarizations), showed pronounced, although broad minimum revealing the exciton splitting in magnetic field. The depth of the minimum was increasing with magnetic field. The typical spectra of different temperatures are shown in Fig. 1.
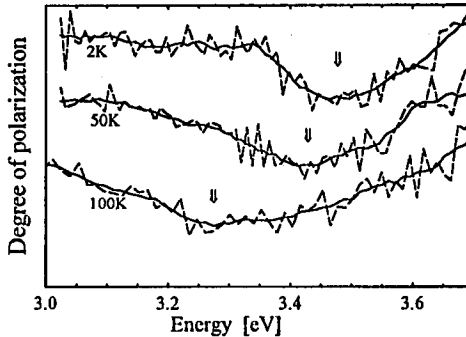

Fig. 1. Degree of polarization of reflectance of MnTe at various temperatures. Due to the large noise the solid lines were added to guide the eyes.

Assuming a symmetric exciton splitting [1, 12] we ascribed the energy of MCD minimum to the exciton (energy gap) position and used it as a means for evaluation of the energy gap. Examples of the energy gap variation with temperature are shown in Fig. 2a (for $Cd_{0.34}Mn_{0.66}Te$) and Fig. 2b (for MnTe). The red shift of the energy gap with temperature is clearly visible for all the samples, being the largest for MnTe (about 0.3 eV from 2 K to 150 K). Moreover for MnTe a kink-like anomaly may be observed around $T = 70$ K, it is well correlated with the antiferromagnetic–paramagnetic phase transition found in neutron scattering [13] and magnetic experiments [10]. On the other hand, for diluted systems ($x = 0.66$, 0.73, 0.85) no anomaly in $E_g(T)$ dependence can be noticed at temperature corresponding to the magnetic phase transition (clearly visible in susceptibility versus T plot [10]). We should mention that the systems with lower concentration of Mn are thought to form a spin-glass phase [1]. In this case, relatively high magnetic field employed by us (5 T) may cause a considerable broadening of the transition.

Variation with temperature of the energy gap in DMS (as well as in magnetic semiconductors [14]) results from two effects: a nonmagnetic effect, reflecting phonon-induced lattice constant variation with T, typical of nonmagnetic semiconductors [15] and the mentioned above exchange correction to the conduction and valence bands [9, 2, 3, 16]. The exchange interaction between the bottom of the conduction band and the upper conduction band states as well as interaction between the top of the valence band and the lower valence band states yields a red shift of the energy gap [9, 2, 3, 7]. This red shift is proportional to the spin correlation
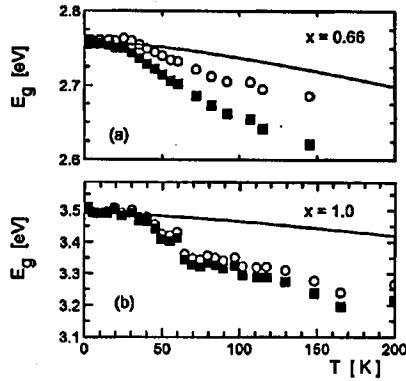
Fig. 2. Energy gap variation with temperature of $Cd_{0.34}Mn_{0.66}Te$ (a), and MnTe (b) — full points. The solid lines show predicted energy gap dependence for "nonmagnetic" $Cd_{1-x}Mn_xTe$ (Eq. (3)). The empty points depict experimental energy gap E_g after subtracting exchange contribution described by Eq. (2) (i.e. $E_g - \Delta E_{exch}$).

function and the strength of the exchange interaction. For disordered magnetic ion system the spin correlation function can be expressed by k-independent magnetic susceptibility and finally exchange contribution is proportional to the product of susceptibility and temperature [7, 3]:

$$\Delta E_{exch} = b\chi(x,T)T. \tag{2}$$

The proportionality factor b ($b < 0$) contains the square of exchange s, p–d integrals and the cut-off value of k-vector q_0, limiting the band states contributing to the correction [7]. The cut-off value q_0 is usually considered as an adjustable parameter [7, 3]. For the nonmagnetic part of the energy gap of $Cd_{1-x}Mn_xTe$ the phenomenological description was proposed [3]:

$$E_g^{non} = A(x) + (1 + ax)(E_g^{CdTe}(T) - E_g^{CdTe}(0)), \tag{3}$$

where $A(x)$ describes the energy gap variation with composition and the second term is the pure CdTe gap temperature dependence extrapolated for the mixed crystal. Finally, the energy gap of DMS is the sum of Eqs. (2) and (3) with three adjustable parameters: $A(x)$, a and q_0. The energy gap of bulk CdMnTe crystals was well described by Eqs. (2), (3) for $x \leq 0.4$, for which $a = 0.86$ and $q_0 = 6.6 \times 10^7$ cm^{-1} were found [3]. Using the same parameters and the experimental susceptibility of our epilayers [10] we calculated the exchange correction according to Eq. (2) and subtracted it from the measured energy gap (Figs. 2a, b). The resulting energy ($= E_g - \Delta E_{exch}$) should be described by Eq. (3), which apparently is not the case (Figs. 2a, b). The calculated exchange correction (Eq. (2)) recovers only a small fraction of the magnetic red shift. The deviation from the predicted behaviour is the smallest for $Cd_{0.34}Mn_{0.66}Te$ and the largest for MnTe. We notice that in order to reproduce the experimental magnetic contribution one should use concentration dependent q_0 parameter, which for MnTe should be several times larger than the one obtained in [3] for $x \leq 0.4$. We believe that the observed

behaviour reflects the crudeness of our approximation of the spin correlation function applied in Eq. (2). Apparently for highly concentrated magnetic systems the assumption of k-independent correlation function is much worse than for more diluted crystals. The development of more realistic spin correlation function seems very important since it should shed some light into magnetic order of highly concentrated DMS, in particular for the transition between antiferromagnetic and spin-glass phases. From this point of view the presented energy gap temperature (and concentration) dependence can serve as a convenient test of the theoretical models.

Acknowledgment

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