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Exchange Disorder Band Bending of the $\text{Cd}_{1-x}\text{Fe}_x\text{S}$ Diluted Magnetic Semiconductor

By

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The energy of excitons A, B, and C of $\text{Cd}_{1-x}\text{Fe}_x\text{S}$ ($x < 0.02$) is studied as a function of Fe concentration at $T = 10$ K in the absence of a magnetic field. The observed band bending effect (largest for exciton A) is attributed to the s, p–d exchange interaction correction resulting from the exchange disorder.

1. Introduction

In the case of a ternary mixed crystal $\text{A}_{1-x}\text{B}_x\text{C}$, grown of two different semiconductors AC and BC, with a random distribution of B cations in the AC lattice, the conventional treatment based on the periodicity of the crystal fails. The simplest approximation overcoming this difficulty is the so-called virtual crystal approximation (VCA [1 to 4]), which assumes a fictitious, periodic crystal with cation sites occupied by a fraction x of cations B and $1 - x$ of cations A. In effect the simplest VCA model, including only few electronic bands, predicts a linear variation of the fundamental energy gap with x . The more extended models, taking into account higher bands or the variation of the lattice constant with x , yield bowing effects in the $E_g(x)$ dependence. Such models provide a reasonable description of the experimental $E_g(x)$ data for III–V compounds, see e.g. [4, 5]. On the other hand, substantial deviations from VCA models were observed for II–VI materials [4, 6] and ascribed to the chemical disorder (i.e. the excess local potential reflecting the difference between cations A and B). For both III–V and II–VI compounds the maximum of the bowing effect was observed around $x = 0.5$ [4 to 6].

A particular class of mixed crystals is formed by the so-called diluted magnetic semiconductors (DMS, see e.g. [7]), in which nonmagnetic cations A are substituted by magnetic ions (typically transition metal ions, such as Mn, Fe, or Co). For these materials the s, p–d exchange interaction between band electrons and magnetic ions strongly modifies the random chemical potential and consequently the bowing effects. The bowing of the energy gap was observed for CdMnS [8], ZnMnSe [9, 10], and ZnMnTe [11]. However,

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differently from nonmagnetic compounds, the maximum bowing occurred for rather low x ($x < 0.1$). Moreover, an unusual dependence of the energy gap on temperature was observed [12], as well as an anomalous exchange band splitting for very low x values for CdMnS [13]. All these effects were attributed to the exchange disorder present in DMS [14 to 17]. We note that similar effects of the exchange interaction were also observed for magnetic semiconductors (such as EuO, MnS, or CoO [18]), however, in this case the problem of disorder was absent.

So far most of the experimental data concerning exchange disorder in DMS were obtained for Mn-based crystals. Among them the strongest effects were observed for hexagonal CdMnS [8, 13]. We therefore thought it worthwhile to study band bending effects for another CdS-based material, with a different magnetic ion, namely $\text{Cd}_{1-x}\text{Fe}_x\text{S}$.

2. Theoretical Background

The chemical and exchange disorders can substantially modify the band structure and therefore should be included in the full band Hamiltonian and diagonalized exactly. However, if the magnitude of the random potentials is small with respect to the relevant bandwidth, a perturbative approach can be used. The chemical and exchange disorder may be regarded separately in the standard perturbation theory. This way was followed by Rys et al. [19], who calculated the exchange-induced correction to the band energy for a magnetic semiconductor, up to the second order. Along the lines of this approach exchange correction was obtained for DMS [10, 20, 12]. Generally speaking, the exchange correction is proportional to the square of the exchange integral (reflecting the exchange interaction strength) and the magnetic ion spin correlation function (measured by the product of magnetic susceptibility and temperature) [10, 20]. Generalizing the results obtained for cubic materials [10], one obtains for hexagonal crystals the following exchange disorder corrections (at zero magnetic field):

for the conduction band,

$$E_c = m_c p_0 (\Omega_0 \alpha)^2 \frac{k_B T}{(gh\mu_B)^2} (\chi_{\parallel} + 2\chi_{\perp}), \quad (1)$$

for the heavy hole band (Γ_9),

$$E_A = m_A p_0 (\Omega_0 \beta)^2 \frac{k_B T}{(gh\mu_B)^2} \times \left(\chi_{\parallel} + 2(1 - q^2) \frac{m_B}{m_A} \chi_{\perp} \mathcal{L}(p_0, m_B, E_B - E_A) + 2q^2 \frac{m_C}{m_A} \chi_{\perp} \mathcal{L}(p_0, m_C, E_C - E_A) \right), \quad (2)$$

for the light hole band (Γ_7),

$$E_B = m_B p_0 (\Omega_0 \beta)^2 \frac{k_B T}{(gh\mu_B)^2} \left(-2(1 - q^2) \frac{m_A}{m_B} \chi_{\perp} \mathcal{L}(p_0, m_A, E_B - E_A) + (2q^2 - 1)^2 \chi_{\parallel} + 2q^4 \chi_{\perp} + 2q^2(1 - q^2) (2\chi_{\parallel} + \chi_{\perp}) \frac{m_C}{m_B} \mathcal{L}(p_0, m_C, E_C - E_B) \right), \quad (3)$$

and for the spin-orbit split band (Γ_7),

$$E_C = m_C p_0 (\Omega_0 \beta)^2 \frac{k_B T}{(g \hbar \mu_B)^2} \left(-2q^2 \frac{m_A}{m_C} \chi_\perp \mathcal{E}(p_0, m_A, E_C - E_A) \right. \\ \left. - 2q^2 (1 - q^2) (2\chi_\parallel + \chi_\perp) \frac{m_B}{m_C} \mathcal{E}(p_0, m_B, E_C - E_B) + (2q^2 - 1)^2 \chi_\parallel + 2(1 - q^2)^2 \chi_\perp \right), \quad (4)$$

where

$$\mathcal{L}(p, m, E) = 1 - \frac{\sqrt{2mE}}{\hbar p} \arctan \left(\frac{\hbar p}{\sqrt{2mE}} \right), \quad (5)$$

$$\mathcal{E}(p, m, E) = -1 + \frac{\sqrt{2mE}}{2\hbar p} \ln \left| \frac{\sqrt{2mE} + \hbar p}{\sqrt{2mE} - \hbar p} \right|. \quad (6)$$

m is the electron (hole)⁴⁾ mass, Ω_0 the unit cell volume, α the exchange integral for the conduction band, β the exchange integral for the valence band, p_0 denotes the range of the exchange interaction in k -space [10], k_B is the Boltzmann constant, $\hbar$ the Planck constant, μ_B the Bohr magneton, g the g -factor, $\chi_\parallel$ ($\chi_\perp$) the magnetic susceptibility for the magnetic field parallel (perpendicular) to the crystal hexagonal axis, q^2 the wave function mixing coefficient for hexagonal crystals and is given by [21]

$$q^2 = \frac{1}{2} \left(1 - \frac{\Delta_1 - \Delta_2}{\sqrt{(\Delta_1 - \Delta_2)^2 + 8\Delta_3^2}} \right). \quad (7)$$

The Δ_1 , Δ_2 , and Δ_3 are the conventional crystal field and spin-orbit parameters, see, e.g. [21]. The functions $\mathcal{L}(\cdot)$ and $\mathcal{E}(\cdot)$ in (2) to (4) result from the fact that the valence band is split by the crystal field and spin-orbit interaction. The positive factors in (2) to (4) describe the upward repelling of the band by the lower lying states (which eventually lead to the redshift of the energy gap), while negative factors characterize the downward pulldown by the higher lying states. We notice that results for lh and spin-orbit subbands are valid only if the cutoff value p_0 is sufficiently small compared with the corresponding valence band splittings: $\hbar^2 p_0^2 / 2m_A \ll E_B - E_A$ for the lh band and $\hbar^2 p_0^2 / 2m_A \ll E_C - E_A$, $\hbar^2 p_0^2 / 2m_B \ll E_C - E_B$ for the s-o subband. For $\Delta_2 = \Delta_3$ there is no hh-lh splitting ($E_B = E_A$) and (3) and (4) reduce to standard formulas for cubic crystals [10]. We stress that chemical disorder effects are not accounted for in (1) to (4).

For the sizable chemical disorder the electron or hole can be nearly localized in the nonmagnetic potential well, and exchange effects can be strongly enhanced and may no longer be considered as a small correction regarding the band width. In such a case the perturbative approach cannot be used and chemical and exchange disorder effects cannot be regarded separately. Instead, the alloy theory taking into account both disorder potentials simultaneously should be applied. This type of nonperturbative calculations has been performed for DMS by Scalbert and coworkers [14, 15] and most recently by Tworzydło

⁴⁾ The isotropic hole mass was assumed in deriving (2) to (4), which is not the case for CdS. The calculations taking into account valence band anisotropy would result in different values of the integral over the k -space, which could be translated into a sort of "average" mass. Effectively, this would scale p_0 , which is a fitting parameter anyway.

[16, 17]. Unfortunately these models provide no simple analytical results and are limited to a nondegenerate electron or hole band. The resulting energy correction is parametrized by the corresponding exchange integral, size of the nonmagnetic potential well (width and depth), and band carrier mass. The disorder effects for CdMnS (band bending in the absence of a magnetic field and Zeeman band splittings) were successfully described by the alloy theory model [16, 17].

3. Experimental Results

The $\text{Cd}_{1-x}\text{Fe}_x\text{S}$ crystals were grown by the modified Bridgman method at the Military Academy of Technology (Warsaw, Poland). Crystals of acceptable optical quality were obtained for $x < 0.02$. The iron content in the samples was determined from magnetization measurements in the following way. Previous magnetic experiments showed that for low Fe concentrations ($x < 0.02$), the magnetization of CdFeS results in practice from isolated (noninteracting) Fe ions [22]. The contribution of Fe–Fe pairs or larger clusters can be neglected at low temperatures and not very strong magnetic fields (< 20 T). Since the probability of finding an Fe ion with no nearest magnetic neighbors is $(1 - x)^{12}$ and the number of Fe ions in the crystal is xN (N is the number of the cation sites per unit mass), the magnetization of the crystal can be expressed as $M = (1 - x)^{12} xN\langle m \rangle$, where $\langle m \rangle$ is the magnetic moment of an isolated Fe ion. Therefore, the magnetization of two different crystals scales with the concentration in the following way:

$$\frac{M_1}{M_2} = \frac{(1 - x_1)^{12} x_1}{(1 - x_2)^{12} x_2}. \quad (8)$$

Using this equation and measuring the magnetization (at 5 T and 4.2 K) of our samples and comparing it with the magnetization of the standard crystals (for which the Fe concentration was determined by microprobe and atomic absorption [22]), we were able to determine the Fe content of all the samples. The absolute accuracy of this method is limited only by the accuracy of the x determination for the standard crystals (which was about 6%), since the accuracy of our magnetic measurements performed with the use of a SQUID magnetometer is much higher (about 0.5%). We should note that the concentration determined from magnetization of the sample reflects the volume average of x , while the optical experiment described below, probes only the surface of the sample. Therefore, the nonhomogeneity of the crystals may result in an additional inaccuracy of x .

Magnetization data obtained at a magnetic field $B = 0.1$ T parallel to the crystal hexagonal axis and at 4.2 K were used to evaluate the susceptibility: $\chi_{\parallel} = M/B$. We will return to this point in the discussion.

We studied excitonic transitions by measuring the reflectance. The samples were cleaved and mounted on the cold finger of a closed cycle refrigerator providing a temperature $T = 10$ K. Nonpolarized light was used. Typical reflectance spectra are presented in Fig. 1 for $\text{Cd}_{0.9946}\text{Fe}_{0.0054}\text{S}$ and $\text{Cd}_{0.988}\text{Fe}_{0.012}\text{S}$. The excitonic structures corresponding to the transitions from the heavy hole band (exciton A), the light hole band (exciton B), and the spin–orbit split band (exciton C) are clearly visible. The exciton energy was ascribed to the inflection point of the spectrum (marked in Fig. 1). The resulting variation of the energy of the excitons A, B, and C with x is shown in Fig. 2. The energy of the exciton A decreases with increasing Fe concentration, the energy of the exciton B is nearly

Fig. 1. The reflectance spectra (in arb. units) of $\text{Cd}_{0.9946}\text{Fe}_{0.0054}\text{S}$ and $\text{Cd}_{0.988}\text{Fe}_{0.012}\text{S}$ at $T = 10\text{ K}$. The markers denote the excitons A, B, and C

constant, while the energy of the exciton C slightly increases with x . The pronounced broadening of the exciton lines with increasing x results in a rather large uncertainty of the evaluated exciton energy. For $x > 0.02$ the exciton lines were too broad for a reliable evaluation of the exciton energy.

Fig. 2. Energy of excitons A, B, and C of $\text{Cd}_{1-x}\text{Fe}_x\text{S}$ vs. Fe concentration. The size of the data points corresponds to the experimental errors

4. Discussion

In general one should expect an opening of the energy gap (a blueshift) with increasing Fe content for CdFeS, since the energy gap of FeS is expected to be much higher (probably 3 to 4 eV) than that of pure CdS ($E_A = 2.553$ eV). The chemical (nonmagnetic) disorder should affect the $E(x)$ dependence, but maximal bending is expected for x around 0.50. This means that in our concentration range ($x < 0.02$) the exciton energy should in practice increase linearly with x . This conclusion is corroborated by the nonperturbative calculations described below, for which a vanishing exchange interaction was assumed (Fig. 3 and discussion below). In this respect the observed redshift of the exciton A is an evidence of the exchange disorder effects. The situation is not so clear for exciton B (very weak redshift) or exciton C (slight blueshift).

We will first describe the exchange disorder effects using the perturbative model (1) to (4). We assume that the energy of each of excitons A, B, and C is the simple sum,

$$E_i = E_{0i} + ax + (E_e + E_l), \quad (9)$$

where i denotes the exciton A, B, or C, E_{0i} is the pure CdS energy, ax characterizes the nonmagnetic correction to the exciton energy, and the last term describes the exchange disorder correction for the conduction and valence bands (1) to (4).

Since the energy gap of the hexagonal FeS is not known, we assumed the parameter $a \approx 1.0$ eV, which corresponds to 3.6 eV for the exciton A in FeS and is similar to the gap of MnTe (3.6 eV [23]). We also assumed that this value is common for all the three excitons. For the other perturbation theory parameters describing the band structure we adopted the pure CdS values: $\Omega_0 = 4.95 \times 10^{-23}$ cm³, $\Delta_1 = 28.4$ meV, $\Delta_2 = 20.9$ meV, and $\Delta_3 = 20.7$ meV [24] (which results in $q^2 = 0.436$). The electron mass is $m_e = 0.2m_0$ [25]. For the heavy hole mass we have taken the geometrical average of the masses parallel and perpendicular to the hexagonal axis $m_A = 1.35m_0$ [25]. For the light hole mass we assumed $m_B = 1.1m_0$ [25]. For the spin-orbit split valence band mass m_C no reliable data are available [25]. The exchange constant for the conduction band of CdFeS was evaluated from the spin-flip Raman scattering: $\alpha = 0.2$ eV [26]. On the other hand, the exchange integral for the valence band was not reported so far. To estimate the β value we used the results of the kinetic exchange theory for the Fe^{2+} ions in DMS [27]. The exchange constant β depends on the energy differences between the ground and the excited states of the transition metal ion. These differences can be inferred [28] from the acceptor and donor level positions measured in II-VI compounds with Fe^{2+} impurities [29]. We took the data for the β value determined in several Fe-based DMS, see e.g. [30], and we calculated the corresponding p-d hybridization parameter V_{pd} with the use of the estimated excitation energy of the Fe^+ ($S = 3/2$) and Fe^{3+} ($S = 3/2$) ion states. The scaling of the hybridization parameter according to the Harrison rule $V_{pd} \propto d^{-7/2}$ [31] (d being the magnetic ion-anion distance) was used to find the V_{pd} for CdFeS. From this we eventually got $\beta = -2.0$ eV. We note the large difference between the exchange constants and the effective masses for conduction and valence bands $(m_e\alpha^2)/(m_h\beta^2) = 0.002$, which means that the contribution from the conduction band can be neglected.

The values of the magnetic susceptibility $\chi_{\parallel}$, $\chi_{\perp}$ at $T = 10$ K were obtained from the measured $\chi_{\parallel}$ at $T = 4.2$ K, since $\chi_{\perp} = 0.7\chi_{\parallel}$ [22] and $\chi(10 \text{ K}) \approx 0.9\chi(4.2 \text{ K})$. The scaling of magnetization (and susceptibility) discussed above allows one to rewrite (9) in the following way:

$$E_i = E_{0i} + ax + (c_i\chi_{\parallel}T) = E_{0i} + ax + (d_ix(1-x)^{12}T), \quad (10)$$

where c_i and d_i are constants containing known parameters, except p_0 , which has to be regarded as an adjustable parameter.

Employing (10) we fit the data for exciton A and obtained $d_A = 3.07$ eV (Fig. 2), which corresponds to $p_0 = 7.1 \times 10^7 \text{ cm}^{-1}$. This value is slightly higher than those reported for CdMnTe ($p_0 = 6.6 \times 10^7 \text{ cm}^{-1}$ [20]) and ZnMnSe ($p_0 = 3.3 \times 10^7 \text{ cm}^{-1}$ [10]), but still within the same order of magnitude. The calculated $E_A(x)$ fits the experimental points quite satisfactorily, although the data points seem to bend slightly more than our prediction.

The obtained p_0 value corresponds to the band energy $\hbar^2 p_0^2 / 2m$ of about 2 eV, which is at least an order of magnitude larger than $E_B - E_A$ (about 15 meV) and $E_C - E_A$ (about 78 meV). Therefore the perturbative formulas for the lh and s-o subbands (3) and (4) cannot be used. However, some general remarks can be inferred from the perturbative theory results. The strongest exchange correction is predicted for the hh subband, since all the contributing states are below the top of the hh subband (only positive factors in (2)). For the lh subband there are contributing states both below and above the top of the band, which is pictured by two positive and one negative factors in (3). In general such a situation should result in a smaller exchange effect than for the hh subband. For the s-o subband this trend is still enhanced, since there is one positive and two negative factors in (4). This prediction is in agreement with the experimental observation (the largest redshift for exciton A and the smallest for exciton C).

For the A exciton one can apply the nonperturbative approach of [17, 32]. We assume a linear concentration dependence of the bottom of the conduction band with the same coefficient as in CdMnS [8, 14]. As we have noted above, the nonperturbative model applied in the description of the valence band is parametrized by: the exchange constant β , the depth of the nonmagnetic potential η , and the width of the potential well b . It has been shown for CdMnS [14] that the chemical substitutional potential originates mainly from the p-d hybridization. We can thus estimate the ratio η/β within the chemical trend scheme, which we have previously used for the determination of β . The value $\eta/\beta \approx 0.1$ calculated in this way corresponds to the virtual crystal result of 3.9 eV for the A exciton energy in the hypothetical FeS . This is about 10% more than the value used by us in (8) and reflects the uncertainty of the input parameters. Adjusting the parameter b we obtained the best fit (Fig. 3) for $b = 3.9 \text{ \AA}$, which is close to the value obtained for CdMnS [16, 17]. We notice that this model predicts stronger bending than the perturbative approach and therefore recovers the experimental behaviour slightly better. The deviation between both models increases with x .

Neglecting the exchange interaction one can calculate the energy gap variation of "nonmagnetic" CdFeS . The result is very close to the VCA result and is shown in Fig. 3. The observed band bending can be thus attributed to the exchange disorder contribution in spite of the fact that the nonperturbative theory incorporates both the magnetic and nonmagnetic impurity potentials.

Finally we would like to comment on the reported variation of the crystal field parameters with concentration of the magnetic ions for CdS based DMS [33]. We recall that this effect was anticipated from the observed increase of the energy differences $E_B - E_A$ and $E_C - E_A$ with concentration x . In view of the above results it is clear that the observed behaviour of $E_B - E_A$ and $E_C - E_A$ reflects a strong bowing effect for exciton A and rather weak bowing for excitons B and C. We note that for CdSe based DMS, for which the exchange disorder effects are much less pronounced (cf. energy gap variation with x for CdFeSe [34] and CdMnSe [35]), the energy differences $E_B - E_A$ and $E_C - E_A$ seem to be rather concentration independent [33].

Fig. 3. Energy of the excitons A and B of $\text{Cd}_{1-x}\text{Fe}_x\text{S}$ vs. Fe concentration. The solid line shows the results of the perturbative model described in the text (10). The dashed lines represent the alloy theory model for the valence band exchange parameter $\beta = 2$ eV (lower line) and for the "nonmagnetic" CdFeS ($\beta = 0$, upper line)

5. Conclusions

We discussed the energy variation of the excitons A, B, and C in hexagonal mixed crystals $\text{Cd}_{1-x}\text{Fe}_x\text{S}$ ($x < 0.02$) with the concentration of Fe ions, taking into account the s, p-d exchange disorder effects. We used the perturbative and nonperturbative (the latter is valid only for exciton A) approaches. Both provide a satisfactory description of the experimental data within the investigated concentration range. The alloy theory (nonperturbative) predicts stronger bending and seems to describe the data slightly better. Experimental data in a broader concentration range would be very helpful in the verification of the theoretical models. This would, however, require substantial progress in CdFeS growth technology. The additional test of the model should include a study of the exciton splitting in a magnetic field. In particular a comparison of such data for CdFeS and CdMnS would be crucial for understanding the role of the chemical (exchange independent) disorder potential of different magnetic ions.

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