

Magnetic ion statistics and thermodynamics in dilute magnetic semiconductor quantum structures

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We have extended our generalized pair approximation applicable, in principle, to diluted magnetic semiconductors, in which the spatial distribution of the magnetic ions may be assumed to be random and homogeneous, to systems with a variable magnetic ion concentration. Using our model, which may be called the extended generalized pair approximation, we have developed a quantitative description of the magnetization in dilute magnetic semiconductor quantum structures with moderate magnetic ion concentration ($x < 10\%$). The latest models describing the radial dependence of the exchange interaction, clustering, and interface mixing are also discussed. [S0163-1829(97)04923-0]

I. INTRODUCTION

A dilute magnetic semiconductor (DMS) is formed by replacing a fraction of the cations in a host semiconductor (SC) alloy with transition-metal ions such as manganese (Mn^{2+}). The presence of a magnetic component introduces specific properties related to the strong coupling of the spins of band electrons or holes of a semiconductor to the localized magnetic moments due to the d electrons of the transition-metal ions. This, in turn, greatly modifies the spin splitting of the band carriers and makes it uniquely temperature and magnetic field dependent. The magnetic properties of bulk DMS containing moderate magnetic ion concentrations ($x < 10\%$) have been satisfactorily described by the pair approximation (for a review see Ref. 1) or generalized pair approximation² (GPA), which express the total free energy of DMS as a sum of interacting pairs multiplied by appropriate probabilities.

The molecular beam epitaxy (MBE) technique gives the possibility to manufacture DMS quantum structures (QS), such as thin films, quantum-well structures, and superlattices, with the energy gap depending on the magnetic ion concentration, which can be controlled during the growth process. As a consequence, the shape of the quantum potential of the DMS QS depends on the magnetic ion distribution along the growth direction. Recent magneto-optical studies of the sharpness of the interfaces between the DMS and SC regions revealed the existence of small-scale disorder, which is also called interface mixing (see Ref. 3). This nonintentional dilution at the interfaces leads to a giant Zeeman splitting in $\text{CdTe/Cd}_{1-x}\text{Mn}_x\text{Te}$ quantum wells³ (QW) and is responsible, in our opinion, for the increase in the number of uncompensated paramagnetic spins, recently observed ex-

perimentally in digital magnetic heterostructures.⁴ In studies of the magneto-optical properties of DMS QS the determination of the average spin of the magnetic ions and knowledge of the exact magnetic ion concentration is of utmost importance as the magnetic-field-induced band potential is proportional to the product of the average spin (the mean spin projection) of the magnetic ions S_z and the magnetic ion concentration x . The mean spin projection of DMS QS has been approximated until now by $S_x = S_0(x)B_J(B, T_{\text{eff}}(x))$,⁵ where B is the external magnetic field, and B_J is a Brillouin function for the spin value J with two adjustable parameters, i.e., the saturation value $S_0(x)$ and effective temperature $T_{\text{eff}}(x)$. It has been proposed recently⁶ that in the limit of high magnetic field, a saturation value has to be replaced by $S_0(x) + \delta S_0(x)$ with $\delta S_0(x) = [Jg\mu_B - S_0(x)]B_J(B, T'_{\text{eff}}(x))$, where g is the Landé factor, μ_B is the Bohr magneton, and $T'_{\text{eff}}(x)$ an additional effective temperature that describes the field-dependent increase in the saturation value. However, such an approximation, which satisfactorily reproduces the experimental data, is justified only for bulk materials in which the spatial distribution of the magnetic ions may be assumed to be random and homogeneous.⁷ Moreover, this approach is not very useful when studying the exchange interaction between the magnetic ions and is additionally invalid at low temperature and high fields, where a steplike behavior of S_z occurs and values of the adjustable parameters depend on the number of layers.

Motivated by the difficulties mentioned above with the description of the mean spin in DMS QS, we have undertaken a theoretical effort to extend the generalized pair approximation of DMS systems with variable magnetic ion concentrations. We have subsequently used the resulting

model to test the recently proposed independent path model of the radial dependence of superexchange, to check the randomness of the magnetic ion distribution in MBE-grown DMS epilayers, and to calculate the effect of the interface mixing in extremely thin DMS epilayers.

II. THEORY

The pair approximation or its extended version with open triplets called ENNPA (Ref. 8) may be applied to DMS's with moderate concentrations ($x < 10\%$) of magnetic ions randomly distributed over a cation sublattice and where each cation site has the same arrangement of other cations. This results in the same number of equidistant nearest neighbors (NN's) and applies to most of the known II-VI DMS's crystallizing in the zinc-blende structure. As follows from our previous work,² where we generalized the pair approximation for the case of an arbitrary crystal structure possessing t kinds of inequivalent sites in the host lattice, the total free energy of the system of magnetic ions can be written as

$$F(x) = \sum_{\alpha=1}^t N_{\alpha} F^{\alpha}(x), \quad (1)$$

where $F^{\alpha}(x)$ is the mean free energy per spin, N_{α} is the number of spins occupying the lattice sites of the α th kind, and the summation runs over all inequivalent cation sites. Treating the sites of the host lattice as being arranged in spheres ν at distances R_{ν}^{α} around the reference site of the α th kind, $F^{\alpha}(x)$ can be defined as²

$$F^{\alpha}(x) = \sum_{\nu}^{\infty} P_{\nu}^{\alpha}(x) F_{\nu}^{\alpha}, \quad (2)$$

with $\sum_{\nu}^{\infty} P_{\nu}^{\alpha}(x) = 1$, and where

$$F_{\nu}^{\alpha} = -\frac{k_B T}{2} \ln \text{Tr} \exp \beta [g \mu_B (\mathbf{S}_{\nu} + \mathbf{S}^{\alpha}) \cdot \mathbf{B} + 2J_{\nu}^{\alpha} \mathbf{S}_{\nu} \cdot \mathbf{S}^{\alpha}] \quad (3)$$

is the free energy of a pair of interacting spins, $J_{\nu}^{\alpha} = J(R_{\nu}^{\alpha})$ is the corresponding exchange constant, \mathbf{S}_{ν} and \mathbf{S}^{α} denote the spin operators of the magnetic ions, $\beta = (k_B T)^{-1}$ (with k_B

the Boltzmann constant), and the probability $P_{\nu}^{\alpha}(x)$ of finding such a pair is given by

$$P_{\nu}^{\alpha}(x) = (1-x)^{n_{\nu}^{\alpha}-1} - (1-x)^{n_{\nu}^{\alpha}}, \quad (4)$$

where $n_{\nu}^{\alpha} = \sum_{\tau} N_{\tau}$ with N_{τ} being the number of sites in π th sphere. Based on Eqs. (1)–(3), other thermodynamic quantities can be derived such as the magnetization ($M = -[\partial F / \partial B]_T$), the magnetic specific heat ($C_m = -T[\partial^2 F / \partial T^2]_B$), and the magnetic susceptibility ($\chi = -[\partial^2 F / \partial B^2]_T$).

The application of the GPA to DMS QS becomes possible because of the following facts:

(i) From the point of view of the GPA, DMS QS are solids with nonequivalent planes perpendicular to the growth direction. This means that all the ions belonging to a given plane have the same numbers of first (1N), second (2N), third (3N), etc. neighbors, so they are all equivalent, but the ion sites belonging to different planes may be inequivalent. For example, all the ions placed in a DMS quantum well (QW) in the plane adjacent to a nonmagnetic SC have eight 1N, five 2N, twelve 3N, etc., while the ions in the adjacent DMS plane have 12, 5, 20, etc., neighbors, respectively.

(ii) Due to the technological process, the magnetic ion concentration in DMS QS may be varied intentionally or nonintentionally (the interface mixing) when passing from one inequivalent plane to another.

(iii) In II-V DMS, the number of NN's is not greater than 5 (i.e., there may be 3, 4, and 5 NN's), while in II-VI DMS this number is 12 and this essential difference requires one to take into account also the contributions of larger clusters, first of all the open triplets. Therefore, such an approximation may be called the extended generalized pair approximation (EGPA). To simplify the resulting equations, we assume that, as far as the distances between ions are concerned, all the cation sites in the DMS are equivalent. Formally this means that the exchange constant is independent of the index α .

Taking into account the above considerations, the mean free energy of a magnetic ion placed in the α plane can be written as

$$F^{\alpha}(x_1, x_2, \dots, x_t) = P_1^{\alpha}(x_{\alpha-1}, x_{\alpha}, x_{\alpha+1}) F_1^{\alpha} + T^{\alpha}(x_{\alpha-1}, x_{\alpha}, x_{\alpha+1}) F_T^{\alpha} + \sum_{\tau=2}^{\bar{\nu}} P_{\tau}^{\alpha}(x_{\alpha-\tau}, \dots, x_{\alpha-1}, x_{\alpha}, x_{\alpha+1}, \dots, x_{\alpha+\tau}) F_{\tau}^{\alpha}, \quad (5)$$

where F_T^{α} is the free energy of the open triplet, which is given by

$$F_T^{\alpha} = -\frac{k_B}{3} \ln \text{Tr} \exp \{ \beta [g \mu_B (\mathbf{S}_1 + \mathbf{S}^{\alpha} + \mathbf{S}_{1'}) \cdot \mathbf{B} + 2(2J_1 \mathbf{S}_1 \mathbf{S}^{\alpha} + J(R_4) \mathbf{S}_1 \mathbf{S}_{1'})] \}.$$

The probabilities P_{ν}^{α} and T^{α} , which determine whether a given magnetic ion placed in plane α with concentration x_{α} has a nearest magnetic neighbor at distance R_{ν} , now reads as

$$P_1^{\alpha}(x_{\alpha-1}, x_{\alpha}, x_{\alpha+1}) = \{ m_1^{\alpha} x_{\alpha} (1-x_{\alpha})^{m_1^{\alpha}} (1-x_{\alpha+1})^{m_1^{\alpha+1}} (1-x_{\alpha-1})^{m_1^{\alpha-1}} \} + \{ \text{CPI} \}, \quad (6)$$

$$T^\alpha(x_{\alpha-1}, x_\alpha, x_{\alpha+1}) = \left(\frac{m_1^\alpha}{2}\right) x_\alpha^2 (1-x_\alpha)^{m_1^\alpha-2} (1-x_{\alpha+1})^{m_1^{\alpha+1}} (1-x_{\alpha-1})^{m_1^{\alpha-1}} \\ + m_1^\alpha m_1^{\alpha+1} x_\alpha x_{\alpha+1} (1-x_\alpha)^{m_1^\alpha-1} (1-x_{\alpha+1})^{m_1^{\alpha+1}-1} (1-x_{\alpha-1})^{m_1^{\alpha-1}} + \{\text{CPI}\}, \quad (7)$$

$$P_v^\alpha(x_{\alpha-\tau}, \dots, x_{\alpha-1}, x_\alpha, x_{\alpha+1}, \dots, x_{\alpha+\tau}) = \prod_{\sigma=-\tau}^{\tau} (1-x_{\alpha+\sigma})^{m_v^{\alpha+\sigma}} - \prod_{\sigma=-\tau}^{\tau} (1-x_{\alpha+\sigma})^{m_v^{\alpha+\sigma}}, \quad (8)$$

where m_v^ε is the number of sites in the circle defined by the intersection of the v th sphere centered on the α plane with a plane ε , such that $\alpha - \tau \leq \varepsilon \leq \alpha + \tau$ with τ being the number of planes corresponding to the length R_v , and $\{\text{CPI}\}$ denotes the sum of two similar terms obtained by cyclic permutation of indices $\{\alpha, \alpha+1, \alpha-1\}$.

III. APPLICATION OF THE EGPA TO II-VI DMS

The total free energy F derived within EGPA from Eqs. (1) and (5) is the sum of the free energies of pairs and triplets of interacting spins multiplied by appropriate probabilities, which can easily be calculated for a given x provided the crystal structure and the parameters of the QW in question are known. Thus, in order to calculate F , one needs to specify an exchange constant for each pair. Recently, when discussing the radial dependence of the d - d exchange interaction in DMS, some new facts have appeared requiring additional treatment. Namely, Shen, Luo, and Furdyna⁹ have studied the exchange striction effects (i.e., the temperature-dependent change of the lattice constants parallel and normal to $\text{Zn}_{0.062}\text{Mn}_{0.938}\text{Te}$ (ZMT) [001] epilayers) and have found failures in the existing models. They compared the experi-

mentally determined sum of the spatial gradients $\nabla J(r)$ with those calculated from the phenomenological model following the power law¹⁰ $J(r) \sim (r)^{-n}$, $n=7-14$ and also the short-range superexchange in the Gaussian form¹² $\exp(-\mu r^2)$, where μ is a constant factor. As a solution, they have proposed a new, independent-exchange-path model in which the spatial dependence of the superexchange interaction, expressed by the consecutive exchange constants J_ν , is described by the product of two parameters J_0 and γ as $J_1 = J_0 \gamma$, $J_2 = 4J_0 \gamma^2$, $J_3 = 2J_0 \gamma^2$, $J_4 = J_0 \gamma^2$, and $J_5 = 4J_0 \gamma^3$ with $\gamma=0.044$ for ZMT. The arguments used by the authors of Ref. 9 to show the failures in the existing description are based on several simplified assumptions; the most important of these is that the analytical expressions written above describe fully the radial dependence of the exchange interaction. In reality, as is known from experimental studies of the magnetization under high pressure in DMS (Ref. 11) and from theoretical considerations,¹² the proportionality constant in these expressions strongly depends on the Mn-Te bond length d , giving an additional strong radial dependence as d^{-16} . A compression of the lattice constants in the x - y plane accompanied by an expansion along the z direction (for so-called ‘‘Z’’ domains, see Ref. 9) shortens the bond

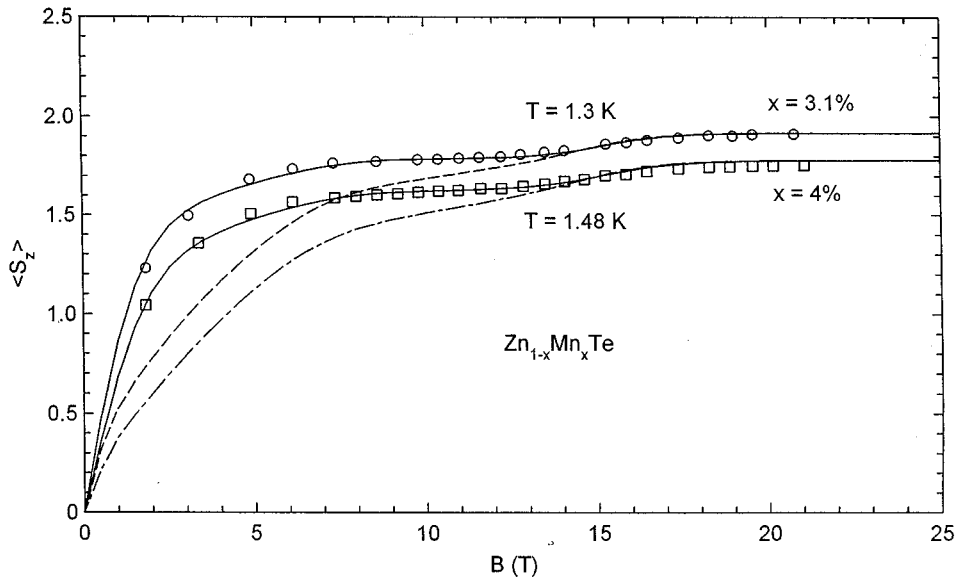


FIG. 1. A comparison of our calculation (lines) of the mean spin projection $\langle S_z \rangle$ in $\text{Zn}_{1-x}\text{Mn}_x\text{Te}$ with the experimental data (points) taken from Ref. 14. The lines are calculated within the extended generalized pair approximation. The solid lines correspond to the phenomenological power-law dependence of the exchange interaction $J(R_\nu)/k_B = J_1 R_\nu^{-6.8}$. The dashed lines are calculated within the independent-exchange-path model (Ref. 9) in which the first five exchange interactions are the following: J_1 , $J_2 = 4J_1 \gamma$, $J_3 = 2J_1 \gamma$, $J_4 = J_1 \gamma$, $J_5 = 4J_1 \gamma^2$ with $\gamma=0.044$. In both cases the experimentally determined (see Ref. 14) value of -10 K is taken for J_1/k_B .

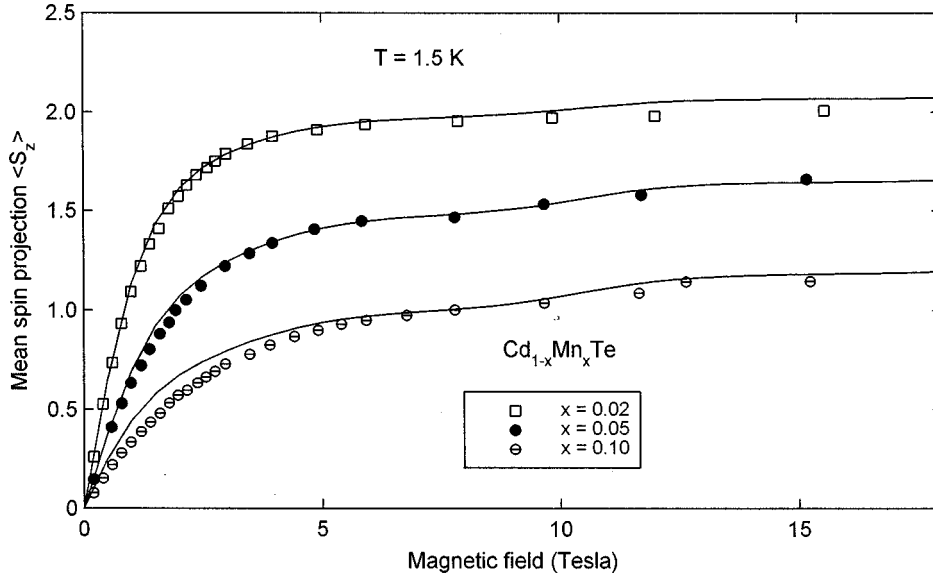


FIG. 2. The mean spin projection $\langle S_z \rangle$ for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ at $T=1.5$ K as a function of magnetic field. The solid lines are calculated within the extended generalized pair approximation and the phenomenological power-law dependence of the exchange interaction $J(R_\nu)/k_B = J_1 R_\nu^{-6.8}$ with $J_1/k_B = -7$ K (see Ref. 1). The experimental points are taken from Ref. 5.

length d . In contrast, the cation-cation distance r , in the z direction, becomes larger. The result is that the final radial dependence of the superexchange interaction observed in the magnetostriction experiments should be a competition between these two opposite mechanisms, which leads to slower spatial dependence. For this reason we will not discard the phenomenological description, which has a simple form. However, since the radial dependence of the exchange interaction given by the new model is also simple, we will test both models. Furthermore neither model requires fitting parameters if for J_1 the experimentally determined value is taken. The magnetization data for $\text{Zn}_{1-x}\text{Mn}_x\text{Te}$ (Ref. 14) together with the theoretical curves calculated with EGPA for the power-law radial dependence (solid line) and with the independent-exchange-path model (broken line) are shown in Fig. 1. The calculations were performed with the value of the nearest-neighbor constant equal to -10 K, as experimentally determined from the position of the magnetization step in ZMT.¹⁴ It can be seen that the EGPA describes very well the experimental data with the phenomenological radial dependence of the exchange interaction in the power-law form, while the independent-exchange-path model gives the wrong description. For the latter consideration we need also the exchange parameters for another DMS; in Figs. 2 and 3 we show the results of similar calculations for bulk $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ (CMT) and $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ (ZMS) with experimentally determined nearest-neighbor exchange constant $J_1 = -7$ K for CMT and $J_1 = -12.2$ K for ZMS (see Ref. 1) and with distant neighbor interaction in the power-law $J_\nu = J_1 r^{-6.8}$ and $J_\nu = J_0 r^{-6.8}$, $\nu \geq 2$ and $J_0 = -10$ K, respectively, for CMT and ZMS. It is remarkable that the description with the same exponent in the power law appears to be equally good for these wide-gap Mn alloyed DMS. In order to show the limits of applicability of the presented model in higher magnetic fields, we present in Fig. 4 the experimental data of Ref. 16 obtained from Faraday rotation measurements up to 150 T. As can be seen, our model works well even in fields up to 50 T for $x=0.1$. On the other hand, the fact that the model reproduces very well experimental data with $x \leq 0.01$ provides a very good proof of the randomness of the magnetic ion distribution in bulk DMS. We note here

also that recently as a better approximation of the short-range superexchange the power-law radial dependence $J(r) \sim r^{-8.5}$ has been proposed¹³ than a Gaussian form. In light of our analysis the acceptable value of the exponent n for the DMS considered is $n = 6.8 \pm 0.2$.

We concentrate now on the discussion of clustering effects in DMS QS grown by MBE. The concept of a nonrandom distribution of magnetic ions in these structures (clustering) was introduced to explain the quantum size effect observed in DMS QS.¹⁷ In order to clarify the situation, we have applied our model to describe the Zeeman splitting measured in thick $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ and $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ epilayers

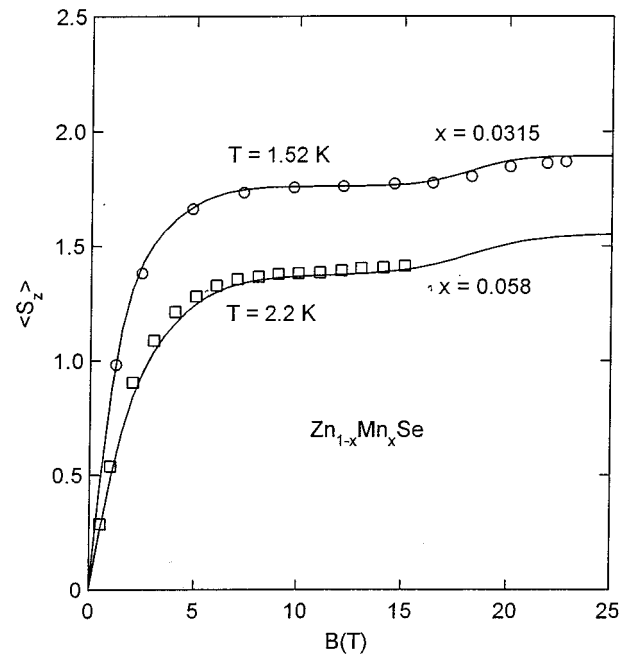


FIG. 3. The mean spin projection $\langle S_z \rangle$ for $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ as a function of magnetic field, at $T=1.52$ K (circles Ref. 15) and at $T=2.2$ K (boxes Ref. 10). The lines are calculated in the same way as in Fig. 2 with experimentally determined $J_1/k_B = -12.2$ K (see Ref. 1) and $J_\nu/k_B = J_0 R_\nu^{-6.8}$, $\nu \geq 2$ and $J_0/k_B = -10$ K.

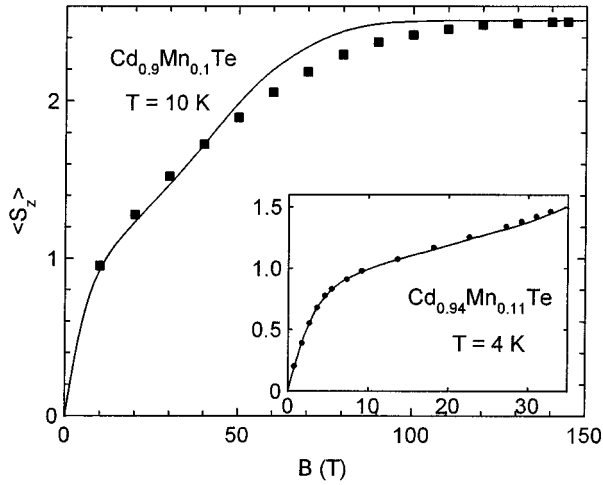


FIG. 4. The high-magnetic-field mean spin projection $\langle S_z \rangle$ of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$. The solid lines are calculated in the same way as in Fig. 2. The experimental data of Refs. 16 and 17 are shown in the inset.

grown by MBE, i.e., bulklike samples grown under nonequilibrium conditions. Figures 5 and 6 show recently measured Zeeman splittings in pulsed high magnetic fields (up to 45 T) by Nicholas *et al.*⁶ in a series of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ epilayers, grown by MBE on [001] InSb substrates as well as the experimental data obtained by Yu *et al.*¹⁸ on a series of $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ epilayers grown by MBE on semi-insulating GaAs[100] substrates. From magneto-optical measurements we have access to the energies of the Zeeman splittings, which for the σ^\pm excitations are given by $\pm \frac{1}{2}(N_0\alpha/N_0\beta)x\langle S_z \rangle$, where $N_0\alpha$ and $N_0\beta$ are the normal-

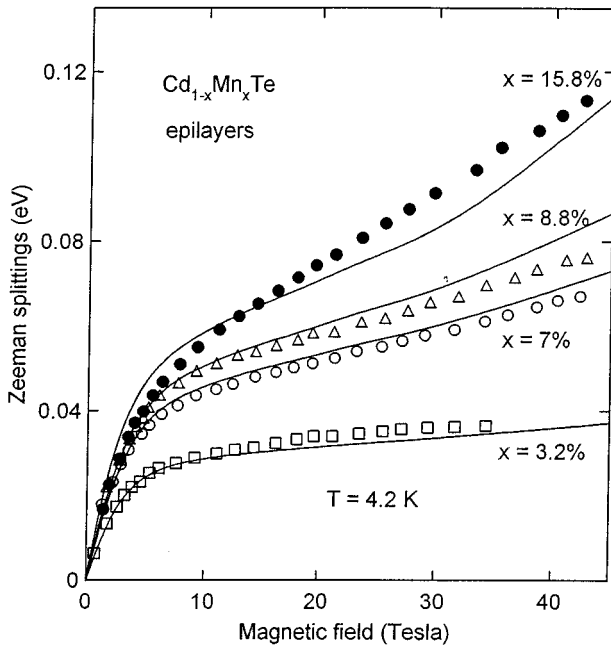


FIG. 5. A comparison of the calculated Zeeman splittings (solid lines) at $T = 4.2$ K with experiments for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$. The solid lines are calculated with the same values of parameters as in Fig. 2. Experimental data are taken from Ref. 6.

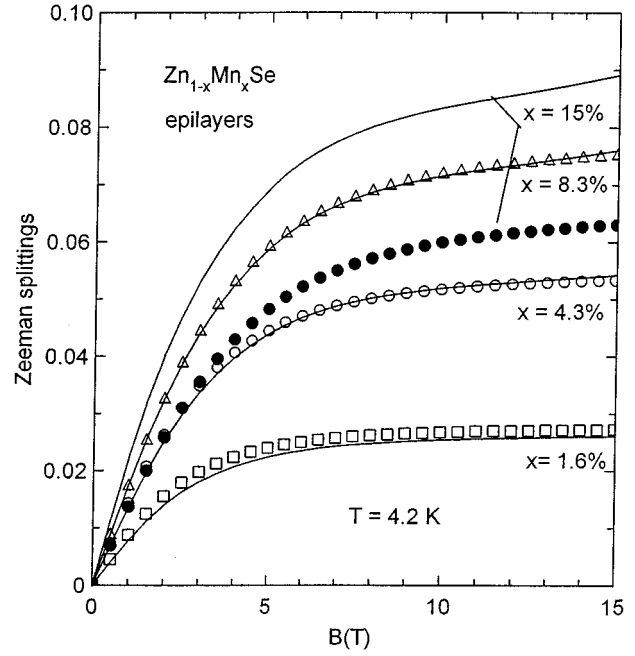


FIG. 6. A comparison of the calculated Zeeman splittings (solid lines) with experiment for $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ as a function of magnetic field at 4.2 K. The solid lines are calculated in the same way as in Fig. 3. The experimental data are taken from Ref. 18.

ized conduction- and valence-band integrals, respectively. The comparison of the experimental data with the results of calculations, performed with the same set of parameters as for the bulk, leads us to the important conclusion that the magnetic ion distribution in these samples is random. Some deviation observed in the case of the CMT epilayers in high magnetic fields is most probably due to an incorrect diamagnetic correction and/or pulsed high magnetic field measurement calibration.

Another application of the EGPA model is the possibility to take into account the effects connected with the dilution of the SC/DMS interface. To simulate the real distribution of the Mn ions near the interfaces, we use (according to Ref. 19) 20% interface disorder over two monolayers (MN), which means that 2 MN in the barrier region adjacent to the interface have magnetic ion concentration $0.8x$, and 2 MN in the well region adjacent to the interface have $0.2x$. We have chosen a nominal concentration $x = 5\%$ and a series of DMS thin epilayers buried between SC layers consisting, in the perfect case, of 2, 3, 5, and 7 ML. In Fig. 7 we show the influence of interface mixing on the averaged spin $\langle S_z \rangle$ by comparing these results of calculations for perfect DMS epilayers with those obtained when the local disorder is included. The main difference in the magnetic properties of such quantum structures has only a statistical source and the appropriate probabilities must be calculated for each specific epilayer separately. Generally, the average spin in these thin DMS structures increases systematically as the number of layers decreases and this is very pronounced in low magnetic fields. Additionally, the dilution effects lead to an enhancement of the paramagnetic behavior. At higher fields $\langle S_z \rangle$ exhibits a temperature-broadened, steplike increase due to the magnetic-field-induced alignments of the antiferromagnetically coupled 1N pairs and for this reason the calculation

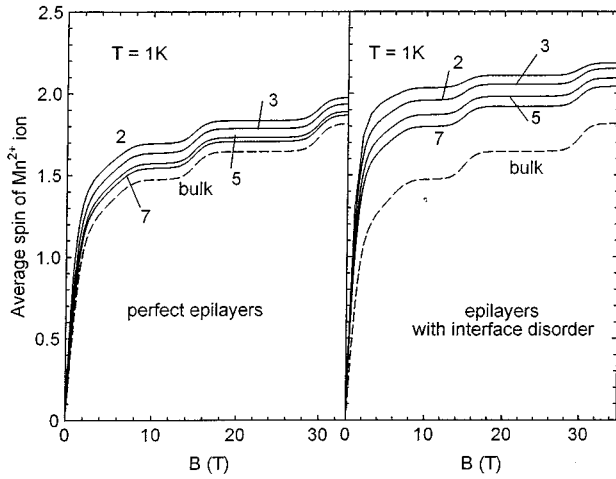


FIG. 7. The calculated magnetic field dependence of the averaged spin of Mn^{2+} ions in $\text{Zn}_{0.95}\text{Mn}_{0.05}\text{Te}$ at $T = 1$ K: (a) thin perfect epilayers consisting of with 2, 3, 5, and 7 layers are compared with bulk, (b) the same as in (a) but with interface disorder included.

has been performed with a temperature of 1 K. A comparison of the step magnitude ($\Delta\langle S_z \rangle_{\text{step}}$) of these curves shows that the dilution in thin epilayers decreases the step height, while in thin perfect layers the step height is the same as in the bulk.

IV. SUMMARY AND FINAL REMARKS

We have extended the generalized pair approximation which, in principle, is applicable to DMS in which the spatial distribution of the magnetic ions may be assumed to be ran-

dom and homogeneous, for use with the systems with variable magnetic ion concentration. Using our model, which may be called the extended generalized pair approximation, we have shown that the recently proposed, independent-exchange-path model of the spatial dependence of the superexchange interaction appears to be of limited use for the description of the magnetic properties of DMS. We have also shown that the EGPA, together with the phenomenological radial dependence of the exchange interaction in the power-law form, which has no fitting parameters, describes the experimental data very well for moderate magnetic ion concentrations ($x \leq 10\%$). This description is sufficiently good to be used as a method for determining the magnetic ion concentrations in DMS. Application of the EGPA to the measured Zeeman splittings obtained for thick epilayers of $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ and $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, grown under nonequilibrium conditions by the MBE technique, shows that the distribution of magnetic ions in these structures can be assumed to be random. This is an important conclusion that demonstrates that DMS QS with a random distribution of magnetic ions can in principle be fabricated. This indicates that the presented method would have applications in the determination of growth parameters and cluster formation in DMS QS. Another important feature of the EGPA is the possibility to take correctly into account dilution effects at the SC/DMS interface, making it a useful tool for studies of interface sharpness and the potential profiles of DMS QS.

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