Bound-magnetic-polaron molecule in diluted magnetic semiconductors

Henryk Bednarski\textsuperscript{1} and Jozef Spałek\textsuperscript{2}

\textsuperscript{1} Centre of Polymer and Carbon Materials, Polish Academy of Sciences, M Curie-Skłodowska 34, 41-819 Zabrze, Poland
\textsuperscript{2} Marian Smoluchowski Institute of Physics, Jagiellonian University, Reymonta 4, 30-059 Kraków, Poland

E-mail: hbednarski@cmpw-pan.edu.pl and ufspalek@if.uj.edu.pl

Received 9 December 2011, in final form 10 April 2012
Published 10 May 2012
Online at stacks.iop.org/JPhysCM/24/235801

Abstract

We formulate a complete microscopic theory of a coupled pair of bound magnetic polarons, the bound-magnetic-polaron molecule (BMPM) in a diluted magnetic semiconductor, by taking into account both the proper two-body nature of the impurity-electron wavefunction and within the general spin-rotation-invariant approach to the electronic states. Also, the model takes into account both the Heisenberg and the antiferromagnetic kinetic-exchange interactions, as well as the ferromagnetic coupling within the common spin BMPM cloud. In this manner, we correct, unify and extend the weakly interacting BMP pair models of Wolff–Bhatt–Durst (2002 Phys. Rev. B 65 235205) and the model of nonoverlapping polarons considered by Angelescu and Bhatt (2002 Phys. Rev. B 65 75211). The resulting BMPM Hamiltonian is solved within the continuum-medium and the effective-mass approximations for the donor case and the thermodynamics is derived. In our approach the thermodynamic fluctuations of magnetization of the spins within BMPM are taken as Gaussian. It appears that the fluctuations can strongly stabilize the spin-triplet state, which may constitute a precursor effect of a ferromagnetic ordering in a many-impurity system.

(Some figures may appear in colour only in the online journal)

1. Introduction

The bound magnetic polaron (BMP) represents an impurity charge carrier quantum bound state formed when its spin is coupled to spins of localized magnetic moments in a diluted magnetic semiconductor (DMS). A bound-magnetic-polaron molecule (BMPM) is realized when two such impurities are placed in a vicinity (within the sum of their effective Bohr radii) such that they share a common spin cloud. The thermodynamic fluctuations of magnetization of the spins influence their binding energy and lead to a nontrivial thermal behavior. The importance of these quantum systems has been discussed for novel materials encompassing diluted magnetic semiconductors \cite{1}, ferromagnetic perovskites \cite{2}, and dilute ferromagnetic oxides \cite{3}. For example, the origin of ferromagnetism in diluted magnetic semiconductor such as Ga\textsubscript{1-x}Mn\textsubscript{x}As, has been intensively discussed in recent years \cite{1, 4-8} and various mechanisms have been considered. Also, those objects being of nanoscale size have stimulated an interest in studies of magnetic quantum dots \cite{9-12}. Moreover, the origin of ferromagnetism is a basic question in view of the dominant role of antiferromagnetic superexchange in all of those materials when the carrier concentration is very low \cite{13}. Therefore, it is crucial to describe these interactions accurately in a physically tractable situation. Here we propose a solvable model of two interacting impurity electrons forming a BMPM. The magnetic interaction between a single electron located on a shallow impurity and the localized magnetic moments in DMS has been studied intensively for quite some time \cite{14-20}. In the first period, the influence of classical fluctuations of magnetization on quantum states of the impurity electron was analyzed \cite{14-20}, leading to the concept of BMP. The main result obtained was to demonstrate that thermodynamic fluctuations suffice to produce a spontaneous spin splitting of the donor states in the paramagnetic spin system. The renewed interest in the present decade was stimulated by the possibility of ferromagnetic...
interpolaron interactions [21–23] which would contribute in a fundamental manner to the origin and properties of ferromagnetic DMS systems and other materials [2, 3]. In [21] the authors extended the theory of a single BMP [14–20] to the weakly interacting polaron pair case. The mutual interaction is approximated by the antiferromagnetic Heisenberg exchange interaction, induced by virtual hopping of carriers between the two separated impurities. In a later version of the approach [22] the authors approximate the two-impurity wavefunction by piecewise constant values. They consider also the detailed thermodynamics of the resultant spin model and optimize the coupling parameters. As another approach, in the complementary model of a weakly interacting and nonoverlapping BMP pair [23], the mutual interpolaron interaction is approximated by the Hubbard Hamiltonian. The polaron’s magnetic cloud is represented by an effective magnetic field that is oriented arbitrarily, but whose amplitude is a parameter of the approach [23]. It follows, thus, that in both these models the polaron–polaron interaction is accounted for approximately.

In this paper we correct the indicated deficiencies of both models by accounting for the interpolaron interaction accurately. We formulate a fairly complete microscopic theory of a coupled pair of bound magnetic polarons, the BMPM, in a diluted magnetic semiconductor by taking into account both the proper two-body nature of the impurity-electron wavefunction and within the spin-rotation-invariant approach to the electronic states. The model also takes into account both the Heisenberg ferromagnetic and the antiferromagnetic kinetic-exchange interactions, as well as the ferromagnetic coupling within the common spin BMPM cloud. In this manner our model corrects and unifies existing approaches to an interacting polaron pair: The Wolff–Bhatt–Durst (thereafter referred to as WBD) model [22] of an overlapping polaron pair and the multiple-level generalized Hubbard model of Angelescu and Bhatt (thereafter referred to as AB) with random fields [23]. Additionally, the WBD model is completed by providing a microscopic justification of their Hamiltonian parameters. While the AB model of nonoverlapping, equal-magnitude and large polarons is generalized to the case of an overlapping and arbitrary-magnitude interacting polaron pair, the extension is limited here only to the regular Hubbard model (i.e. within one lowest energy level on each polaron site only). We solve the resulting BMPM Hamiltonian within the continuum-medium [19] and the effective-mass approximations for the donor case and derive the thermodynamics. In our approach the thermodynamic fluctuations of magnetization of the spins within BMPM are taken as Gaussian. Importantly, our approach allows us to go beyond the limit of weak interpolaron interaction assumed in the WBD and the AB approaches.

Recently, we have introduced the BMPM model for the case of two interacting donors and solved it within the simplified Heitler–London approximation in a reduced spin-state space [24]. In this paper we solve the BMPM model accurately in an extended spin-state space which includes also the ionic (doubly occupied) impurity configurations.

We would like to emphasize that we provide here a natural extension of the single BMP theory developed by Dietl and Spałek (DS) [14] to the polaron molecule situation by including the interaction between two such polarons. All details connected with the derivation of the single-polaron Hamiltonian here and the related basic assumptions constitute the starting point of the present paper.

The structure of the paper is as follows. In section 2 we formulate our model. In section 3 we diagonalize the effective Hamiltonian for the BMP molecule. In section 4 the thermodynamical properties of BMPM are considered, whereas section 5 contains concluding remarks. The appendices A and B provide details of analytic calculations.

2. The BMP molecule model

2.1. General formulation

We start by considering two interacting hydrogen-like impurities (donors) in DMS. The electrostatic attraction to the impurity centers provides the primary force of the localization of the impurity electrons. Namely, we assume that the two electrons (of charge $-e$ each) are attracted to two impurities, each of charge $e$, located at $R_a$ and $R_b$, respectively. Additionally, the impurity electrons interact with localized spins of magnetic ions by the contact Fermi (s–d) interaction. In other words, we consider two interacting BMPs in DMS with a random distribution of the spins. Within the continuum-medium and the effective-mass approximations, one can write the starting Hamiltonian in the form:

$$H = H_1 + H_{\Pi}$$

$$\equiv H_1 - \frac{\hbar^2}{2m^*} \left( \nabla_1^2 + \nabla_2^2 \right)$$

$$- e^2 \left[ \frac{1}{r_{a1}} + \frac{1}{r_{a2}} + \frac{1}{r_{b1}} + \frac{1}{r_{b2}} - \frac{1}{r_{12}} \right]$$

$$\equiv - J_e \left[ \hat{S}(\textbf{r}_1) \cdot \hat{\Sigma}_1 + \hat{S}(\textbf{r}_2) \cdot \hat{\Sigma}_2 \right] + H_{\Pi},$$

(1)

where $m^*$ is the impurity-electron effective mass, $a$ and $b$ label the two impurity sites, $l = 1, 2$ label the two electrons, $r_{a1}, r_{a2}, r_{b1}, r_{b2}, r_{12}$ and $R_{ab} = |R_a - R_b|$ are the corresponding relative distances appearing in the problem, $\varepsilon$ is the static dielectric constant, $J_e$ is the exchange integral of the contact s–d interaction between spins $\{\hat{\Sigma}_1\}$ and those of impurity carriers, $\{\hat{\Sigma}_2\}$. Also, $\hat{S}(\textbf{r}_i) = \sum \hat{S}_i (\textbf{r}_i - \textbf{R}_i)$ is the spin density operator with the sum running over the sites occupied by magnetic ions (Mn$^{2+}$). In the above expression consecutive equations define $H_1, H_{\Pi}$, and $H_{\Pi}$, respectively. $H_{\Pi}$ is the hydrogenic-like molecule part, which is supplemented with $H_1$, the s–d exchange coupling between the spins of impurities and the localized spins. Note also that the continuum-medium approximation is realistic, since this assumption was checked out explicitly [19] by comparing the results with those coming from exact diagonalization. Thus the only simplifying assumptions are the choice of the spin density as classical and of their fluctuations as Gaussian.

The solution of the BMP pair problem is complex. On one hand, a systematic approach to the correlated electrons
on the lattice and, in particular, the hydrogen molecule can be achieved within the second quantization formalism [25]. On the other hand, the spin part \( H_4 \) is dependent on the positions of the localized-moment spins. This fact makes the problem complex to solve. However, it becomes tractable when one tries to diagonalize \( H \) as consisting of the hydrogenic-like molecule Hamiltonian \( H_B \) influenced additionally by the s–d coupling \( H_s \). Such a division suggests approaching the solution of the BMP molecule by selecting as trial states the eigenstates of the set of mutually compatible observables \( \{ \hat{s}_{\text{tot}}, \hat{s}_{\text{tot}} \} \), where \( s_{\text{tot}} \) is the total spin of impurity electrons. In fact, as carriers interact via an isotropic Coulomb repulsion, only the rotations of the whole molecule conserve their relative distance, thus leaving their energy unchanged. Moreover, we know that in the Heitler–London approximation (cf [24]) the Hamiltonian \( H_\text{II} \) commutes with both \( \hat{s}_{\text{tot}}^{z} \) and \( \hat{s}_{\text{tot}}^{z} \), and is thus diagonal for the four singlet–triplet states. Such an approach greatly simplifies the problem, as in practice we have solutions for \( H_2 \) within the eigenstates of \( \{ \hat{s}_{\text{tot}}^{z}, \hat{s}_{\text{tot}}^{z} \} \). Here we consider the general solution, i.e. beyond the Heitler–London approximation of the hydrogen molecule, by including also the ionic (doubly occupied impurity) configurations. In this case, \( H_2 \) is not diagonal in the basis formed from eigenstates of \( \hat{s}_{\text{tot}}^{z} \) and \( \hat{s}_{\text{tot}}^{z} \). Fortunately, for the off-diagonal elements, the hopping Couples only different spin-singlet states.

2.2. Coupling of impurity electrons to spins

Next, we express \( H_1 \) in terms of the creation and annihilation operators. Our methodology is based on an explicit demonstration that the postulated occupation-number representation of the s–d coupling part of the BMPM Hamiltonian, denoted by \( \hat{H}_1 \), leads to the same matrix representation as that obtained within the first quantization scheme. Details of the corresponding calculations of the matrix representation of \( H_1 \) within the first quantization formalism are presented in appendix A. Thus, we assume \( \hat{H}_1 \) in the following form:

\[
\hat{H}_1 = \frac{1}{2} \sum_{s,s'} c_{s,s'}^{\dagger} (\Delta_{ss'} (\Delta_{ss'} - \Delta_{ss'})) a_{s,s}' + a_{s,s}^{\dagger} (\Delta_{ss'} - \Delta_{ss'}),
\]

where \( a_{s,s}^{\dagger} (a_{s,s}) \) is the creation (annihilation) operator for the state on impurity \( c = a, b \) with the spin \( s(s = \uparrow, \downarrow) \). The polaron exchange fields \( \Delta_{cc'} \) are defined as:

\[
\Delta_{cc'} = \frac{\alpha}{g \mu_B} \int w_{c}(r) M(r) w_{c'}(r) d^{3}r, \quad c', c = a, b,
\]

where \( M(r) = -g \mu_B N_0 x(\chi_S) \hat{S}(r) |\chi_S \rangle \) is the local magnetization per unit volume, \( \alpha \equiv J_{\text{FNN}}/m_B \) is the Landé factor of spins, \( \mu_B \) is the Bohr magneton, \( N_0 = n_0/e_0 \) is the number of atoms per unit volume, \( x \) is the fraction of magnetic atoms, and \( \chi_S \) is the wavefunction of the spins, \( w_{a}(r) \) and \( w_{b}(r) \) are the orthogonal molecular wavefunctions:

\[
w_{a,b}(r) = \rho[\psi_{\alpha,b}(r) - \gamma \psi_{\alpha,a}(r)],
\]

which are built from the single-particle atomic wavefunctions \( \psi_{\alpha} \) and \( \psi_{\beta} \):

\[
\psi_{\alpha}(r) = (\pi a_B^3)^{-1/2} \exp(-|r - R_c|/a_B),
\]

with \( c = a, b \), being the solution of the corresponding single-particle hydrogenic-like Schrödinger equation with the effective Bohr (donor) radius \( a_B \):

\[
a_B = \frac{\hbar e}{m_e e^2},
\]

where \( \hbar \) is the Planck constant divided by \( 2\pi \). In equation (4) \( \rho \) is the normalization factor and \( \gamma \) is the mixing coefficient; they are defined as:

\[
\rho = \frac{1}{\sqrt{2}} \left[ \frac{1 - \Omega^2}{1 - \Omega^2} + \frac{1 - \Omega^2}{1 - \Omega^2} \right]^{1/2},
\]

\[
\Omega = \frac{1 + \sqrt{1 - \Omega^2}}{1 - \Omega^2},
\]

determined by imposing the orthogonality and the normalization conditions:

\[
\langle w_{c}(r) | w_{c'}(r) \rangle = \int w_{c}(r) w_{c'}(r) d^{3}r = \delta_{cc'},
\]

\[
c, c' = a, b,
\]

whereas the overlap integral \( \Omega \) is defined as follows:

\[
\Omega = \Omega(R_{ab}) = \langle \psi_{\alpha}(r) | \psi_{\beta}(r) \rangle = \int \psi_{\alpha}^{\dagger}(r) \psi_{\beta}(r) d^{3}r
\]

\[
= \exp(-R_{ab}/a_B) \left( 1 + \frac{R_{ab}}{a_B} + \frac{R_{ab}^2}{3 a_B^2} \right).
\]

Hence, \( \rho \) and \( \gamma \) are also dependent on the interimpurity distance \( R_{ab} \). It is important to note here that, when calculating \( \Delta_{cc'} \), we replaced the spin density operator \( \hat{S}(r) \) by its quantum mechanical average, thereby introducing the adiabatic and mean field approximations. This is acceptable, because the same approximations were assumed in the single BMP theory and provided good results [14]. The exchange fields of the BMP, \( \Delta_{cc'} \), \( c' = a, b \), may be oriented in an arbitrary direction. In our approach these polaronic exchange fields are overlapping through the presence of the mixing coefficient in the definition of \( \Delta_{c} = \Delta_{a}, \Delta_{b} \equiv \Delta_{bb}, \text{ and } \Delta_{cc'} \).

Next, we introduce the six eigenstates of the carriers total spin operator \( \hat{S} = \hat{S}_z = \hat{S}_z \); three triplet \( (s = 1) \) and three singlet \( (s = 0) \) states [25]. The triplet states are:

\[
|1\rangle = \frac{1}{\sqrt{2}} (a_{\uparrow}^{\dagger} a_{\downarrow}^{\dagger} |0\rangle + a_{\downarrow}^{\dagger} a_{\uparrow}^{\dagger} |0\rangle),
\]

\[
|2\rangle = a_{\uparrow}^{\dagger} a_{\downarrow}^{\dagger} |0\rangle,
\]

\[
|3\rangle = a_{\downarrow}^{\dagger} a_{\uparrow}^{\dagger} |0\rangle,
\]

and the corresponding three singlet states are:

\[
|4\rangle = \frac{1}{\sqrt{2}} (a_{\uparrow}^{\dagger} a_{\downarrow}^{\dagger} - a_{\downarrow}^{\dagger} a_{\uparrow}^{\dagger} |0\rangle),
\]

\[
|5\rangle = \frac{1}{\sqrt{2}} (a_{\uparrow}^{\dagger} a_{\downarrow}^{\dagger} + a_{\downarrow}^{\dagger} a_{\uparrow}^{\dagger} |0\rangle),
\]

\[
|6\rangle = \frac{1}{\sqrt{2}} (a_{\uparrow}^{\dagger} a_{\downarrow}^{\dagger} - a_{\downarrow}^{\dagger} a_{\uparrow}^{\dagger} |0\rangle).
\]
and

\[ |6⟩ = \frac{1}{\sqrt{2}} (a^+_a |a⟩ - a^+_b |b⟩) |0⟩. \tag{14} \]

A detailed insight reveals that the polaronic exchange fields \( Δ_a \) and \( Δ_b \), originating from the single BMPs fields, induce coupling with impurity electrons within the subspace of molecular states \([1], \ldots, [4] \). Furthermore, the fields \( Δ_{ab} \) and \( Δ_{ba} \) originate solely from the overlapping BMP electron clouds, as can be seen by writing down the explicit form of the exchange field \( Δ_{ab} \), namely:

\[
Δ_{ab} = \frac{α}{gμ_B} \int w_a^* w_b M d^3r
= (1 + γ^2)α^2 \frac{α}{gμ_B} \int ψ_a^* ψ_b M d^3r
- γρ^2 \frac{α}{gμ_B} \int (|ψ_a|^2 + |ψ_b|^2) M d^3r. \tag{15}\]

They induce a coupling between the ionic state \([6]\) and the triplet states, \([1]–[3]\) (cf equation (A.25) in appendix A). Note also, that the \( Δ_{ab} \) \( (Δ_{ba}) \) is composed of the difference of two terms, each approximately proportional to \( γ \). In the case of the antiparallel alignment of the BMP exchange fields, the total magnetization of magnetic ions in the overlapping region is small, thus leading to a small magnitude of \( Δ_{ab} \). For the case of the parallel alignment of the BMP exchange fields, a small magnitude of \( Δ_{ab} \) may be deduced from the orthogonality condition equation (8). This analysis clearly shows that couplings originating from the polaronic exchange fields \( Δ_{ab} \) and \( Δ_{ba} \) between the ionic state \([6]\) and the triplet states are weak. At the same time, the presence of \( Δ_{ab} \) and \( Δ_{ba} \) in \( \tilde{H}_1 \) make the solution of the BMPM problem significantly more complex. For these reasons, we assume here that \( Δ_{ab} = Δ_{ba} \approx 0 \) and call the resulting model Hamiltonian an effective BMPM Hamiltonian. In our numerical calculations, apart from this simplifying assumption, we do not make any a priori restriction on the magnitude and orientation of the polaronic exchange fields \( Δ_a \) and \( Δ_b \). Moreover, we provide later a further convincing quantitative argument that the thermodynamic fluctuations of \( Δ_{ab} \) are negligible for interimpurity distances greater than 3ab.

To proceed further, it is also more convenient to transform the BMP exchange fields \( Δ_a \) and \( Δ_b \) to \( Δ^+ \) and \( Δ^- \) according to the prescriptions:

\[
Δ^+ = \frac{1}{2} (Δ_a + Δ_b) \quad \text{and} \quad Δ^- = \frac{1}{2} (Δ_a - Δ_b), \tag{16}\]

which enable us to write the polaron fields in the form:

\[
Δ^±[M] = \frac{α}{gμ_B} \int \left[ |w_a|^2 ± |w_b|^2 \right] |M(r)⟩ d^3r. \tag{17}\]

This, in turn, leads to the effective \( \tilde{H}_1 \) in the form:

\[
\tilde{H}_1 \approx \frac{1}{2} \sum_{ss'} \left[ a^+_a (Δ^+ • \sigma)_{ss'} a_{ss'} + a^+_b (Δ^- • \sigma)_{ss'} a_{ss'} + a^+_a (Δ^- • \sigma)_{ss'} a_{ss'} - a^+_b (Δ^+ • \sigma)_{ss'} a_{ss'} \right]. \tag{18}\]

Now, we select the direction of the global spin quantization axis as aligned with \( Δ^- \). This choice leads to the following 6 × 6 singlet–triplet matrix representation of \( H_1 = \langle m | H_1 | n⟩ \):

\[
H_1 = \begin{pmatrix}
0 & Δ^+ \sinθ e^{−iφ} & Δ^+ \sinθ e^{−iφ} & Δ^- 0 0 \\
Δ^+ \sinθ e^{−iφ} & 0 & 0 & 0 0 0 \\
Δ^+ \sinθ e^{−iφ} & 0 & 0 & 0 0 0 \\
Δ^- & 0 & 0 & 0 0 0 \\
0 & 0 & 0 & 0 0 0 \\
0 & 0 & 0 & 0 0 0 \\
\end{pmatrix}, \tag{19}\]

where \( θ \) and \( φ \) are respectively, the polar and azimuthal angles between the exchange fields \( Δ^+ \) and \( Δ^- \).

2.3. Comment: effective spin–spin coupling

To demonstrate the generality of our approach more clearly, we map the effective spin Hamiltonian \( \tilde{H}_1 \) onto the corresponding parts of the WBD model Hamiltonian (the whole WBD model contains also the antiferromagnetic Heisenberg exchange interaction between carrier spins, which we do not specify explicitly). Namely, we can rewrite the \( \tilde{H}_1 \) in the spin operator form:

\[
H_{s−d} ≡ I_1 + Δ_1 + Δ_2, \tag{20}\]

and take into account the explicit form of the exchange fields \( Δ_a \) and \( Δ_b \), which yields:

\[
H_{s−d} = \frac{αρ^2}{gμ_B} \left[ \tilde{S}_1 \cdot \int |ψ_a|^2 M d^3r + \tilde{S}_2 \cdot \int |ψ_b|^2 M d^3r \right]
- \frac{αρ^2γ}{gμ_B} \int (ψ_a^† ψ_b + ψ_b^† ψ_a) M d^3r
+ \frac{αρ^2γ^2}{gμ_B} \int \tilde{S}_1 \cdot \int |ψ_b|^2 M d^3r + \tilde{S}_2^2
\times \int |ψ_a|^2 M d^3r \right]. \tag{21}\]

This expression should be compared with the following s–d coupling part of the WBD model Hamiltonian [21, 22]:

\[
H_{WBD} = K[\tilde{S}_1 \cdot \tilde{S}_1 + \tilde{S}_2 \cdot \tilde{S}_2] + K' (\tilde{S}_1 + \tilde{S}_2) \cdot \tilde{S}_3, \tag{22}\]

where now \( K \) and \( K' \) are the WBD model parameters, and \( \tilde{S}_3 \) is the total spin of magnetic ions in the overlapping region. A direct mapping can be established by introducing the following definitions:

\[
K ≡ αρ^2, \quad K' ≡ −γ K, \quad S_{1(2)} ≡ \int |ψ(a(b))^2 |M| d^3r, \tag{23}\]

and

\[
S_3 = \int (ψ_a^† ψ_b + ψ_b^† ψ_a) |M| d^3r \]

which allow us to rewrite \( H_{s−d} \) in the final form:

\[
H_{s−d} = K[\tilde{S}_1 \cdot \tilde{S}_1 + \tilde{S}_2 \cdot \tilde{S}_2] + K' (\tilde{S}_1 + \tilde{S}_2) \cdot \tilde{S}_3
+ γ^2 K[\tilde{S}_1 \cdot \tilde{S}_2 + \tilde{S}_2 \cdot \tilde{S}_1]. \tag{24}\]
For the solution of the BMPM problem, 2.4. Impurity-electron states

0. objects, despite the simplifying assumption that
model. Moreover, it now appears clearly that the interacting
$\boldsymbol{R}_a$ that their magnitude depends on the interpolaron distance
orthogonalized single-particle wavefunctions shows explicitly
in the second quantization form. Such a representation
with the hydrogenic-like molecular part

$\Delta_{\text{ab}}$ relation of BMP exchange fields
the weak-interaction approximation. Furthermore, the derived
$\gamma$ proportional to
the meaning of its parameters. Also, the inclusion of the term
provided a microscopic derivation of the WBD model and

3 terms in equation (24) describe the contributions
γ
is different from zero at large distances, and in this limit
is equal to that for the two isolated BMPs. In

In figure 1 we have plotted the mixing coefficient $\gamma$ (and $\gamma^2$) as a function of
interimpurity distance $R_{ab}$, whereas in
figure 2 we display the same dependence for $K$, $\gamma K$, and $\gamma^2 K$. As can be seen from these plots, only the parameter $K$ is different from zero at large distances, and in this limit it is equal to that for the two isolated BMPs. In $H_{\text{wbd}}$, the parameters $\gamma$ and $\gamma^2$ appear as interaction couplings and

The three terms in equation (24) describe the contributions
γ
as interaction couplings and
the three terms in equation (24) describe the contributions
coming from the zero-, first- and second-order processes,
respectively. One can also see that, at large interpolaron
distances, when $\gamma^2 \to 0$, $H_{\text{wbd}}$ reduces to the s–d coupling
part of WBD model Hamiltonian. In this manner, we have
provided a microscopic derivation of the WBD model and
the meaning of its parameters. Also, the inclusion of the term
proportional to $\gamma^2$ into the WBD model extends it beyond
the weak-interaction approximation. Furthermore, the derived
relation of BMP exchange fields $\Delta_a$ and $\Delta_b$ to the carrier
orthogonalized single-particle wavefunctions shows explicitly
that their magnitude depends on the interpolaron distance
$R_{ab}$, a feature which is completely neglected in the AB
model. Moreover, it now appears clearly that the interacting
BMP pair in a paramagnetic DMS is composed of overlapping
objects, despite the simplifying assumption that $\Delta_{ab} = \Delta_{ba} \approx 0$.\]

2.4. Impurity-electron states

For the solution of the BMPM problem, $H_{\text{II}}$ must be completed
with the hydrogenic-like molecular part $H_{\text{II}}$, rewritten also
in the second quantization form. Such a representation $H_{\text{II}}$
is already known [25], so we can write it explicitly in the
second-quantized form:

\[
\begin{align*}
\hat{H}_{\text{II}} & \equiv \epsilon_n a_{n}^\dagger + \epsilon_p b_{p}^\dagger + t \sum_s \left( a_{sa}^\dagger a_{sb} + a_{sa}^\dagger a_{sb}^\dagger + a_{sb} a_{sa} + a_{sb}^\dagger a_{sa}^\dagger \right) + U_p n_{p1} n_{p2} - 2 f \hat{S}_1 \cdot \hat{S}_2 + (K + \frac{1}{2} J) n_{a} n_{b} \\
& \quad + J (a_{s1}^\dagger a_{s2} + a_{s2}^\dagger a_{s1} + \text{H.c.}) \quad + V \sum_s \left( n_{as} + n_{bs} \right) \\
& \quad + \frac{\Delta_{\text{ab}}}{\sqrt{2}} (a_{sa}^\dagger d_{s\uparrow} + a_{sb}^\dagger d_{s\downarrow}), \quad \text{(25)}
\end{align*}
\]

where, $\epsilon_n$ is the atomic level position, $t$ the hopping
between the impurity states and $U_p$ the interatomic Coulomb
(Hubbard) interaction. The remaining terms represent,
respectively, the ferromagnetic interatomic (Heisenberg)
exchange, the pair hopping, and the so-called correlated
hopping. In the singlet–triplet basis the matrix elements
$\langle m | \hat{H}_{\text{II}} | n \rangle = H_{\text{II}}$ has a relatively simple form [25], so we may
write the following Hamiltonian matrix:

\[
H_{\text{II}} = \left( \begin{array}{cccccc}
L & 0 & 0 & 0 & 0 & 0 \\
0 & L & 0 & 0 & 0 & 0 \\
0 & 0 & L & 0 & 0 & 0 \\
0 & 0 & 0 & L + 2 J & \Upsilon & 0 \\
0 & 0 & \Upsilon & L_{\text{d}} + 2 J & 0 & 0 \\
0 & 0 & 0 & 0 & L_{\text{d}} & 0
\end{array} \right), \quad \text{(26)}
\]

where:

\[
L = 2 \epsilon + K - J, \quad L_{\text{d}} = 2 \epsilon + U - J, \quad \text{and} \quad \Upsilon = 2 (t + V). \quad \text{(27)}
\]

For identical impurities, both atomic level positions $\epsilon_a$ and $\epsilon_b$
and the intraatomic Coulomb interactions $U_a$ and $U_b$ are equal
(i.e. $\epsilon_a = \epsilon_b = \epsilon$ and $U_a = U_b = U$). We note here, that $H_{\text{II}}$
is solved in the local coordinate system, with $z$ axis aligned with
the direction of the molecular bond, and then Hamiltonian $H_{\text{II}}$
is reoriented toward the global spin quantization axis. This
last step is quite trivial due to the spin rotational invariance
of $H_{\text{II}}$.\]

In section 3 we solve the redefined matrix form of
the total effective Hamiltonian $H = H_{\text{I}} + H_{\text{II}}$.\]

3. Solution of the BMP molecule Hamiltonian

Diagonalization of the total effective BMPM Hamiltonian
requires a solution of the eigenequation $HV = EV$, with $H$
given by:

\[
H = \begin{pmatrix}
L & Δ^+ \sin \theta e^{iϕ} & Δ^+ \sin \theta e^{-iϕ} & Δ^- & 0 & 0 \\
Δ^+ \sin \theta e^{-iϕ} L + Δ^- \cos \theta & 0 & 0 & 0 & 0 & 0 \\
Δ^+ \sin \theta e^{iϕ} & 0 & L - Δ^+ \cos \theta & 0 & 0 & 0 \\
Δ^- & 0 & 0 & L + 2j & γ & 0 \\
0 & 0 & 0 & γ & L_0 + 2j & 0 \\
0 & 0 & 0 & 0 & 0 & L_0 \\
\end{pmatrix}
\]  \hspace{1cm} (28)

Its form does not allow for an exact analytical diagonalization. Therefore, before solving it numerically, we discuss first some limiting situations in analytic terms.

3.1. Saturation limit for spins

We consider first the BMPM effective Hamiltonian for a pair of donors in the saturation limit, i.e. with \( M(r) = M_{\text{sat}} \). In this case, the solution is simplified greatly within our approach, because we can write:

\[
Δ^+ = \frac{α}{\hbar^2 μ_B} M_{\text{sat}} = 5/2αN \quad \text{and} \quad Δ^- = Δ_{\text{ab}} = 0,
\]

where \( N_5 \) is the number of Mn\(^{2+}\) ions within BMPM which contribute to magnetization. Subsequent diagonalization of \( H \) leads to the following exact solution for the eigenvalues \( E_i \), \( i = 1–6 \):

\[
\begin{pmatrix}
L \\
L + 5/2αN \\
L - 5/2αN \\
\frac{1}{2}[(L + L_0) + 4j + \sqrt{(L - L_0)^2 + 4γ^2}] \\
\frac{1}{2}[(L + L_0) + 4j - \sqrt{(L - L_0)^2 + 4γ^2}] \\
\end{pmatrix}
\]

The eigenvalues \( E_{4,5} \) contain an antiferromagnetic kinetic-exchange interaction [25, 26] in an explicit form, which competes with the direct Heisenberg ferromagnetic exchange. The nature of the ground state depends on the sign of the difference \( ΔE ≡ E_5 - E_3(Δ^+) \), which determines the dominant exchange interaction aligning individual polaron polarization clouds; for a positive value the BMPM ground state is ferromagnetic. Note that the states belonging to the polarization clouds; for a positive value the BMPM ground state. One sees also that if the two impurities are to close each other, then a nonmagnetic hydrogen-molecule spin-singlet ground state becomes the ground state.

In the s-type II–VI DMS, the magnetic susceptibility \( χ \) of localized magnetic spins at low temperatures takes an effective Curie–Weiss form \( χ = C/(T + T_0) \), with \( T_0 > 0 \) [13]. Therefore, the magnetization is certainly not saturated and the case considered here appears as describing a non-realistic situation. The situation changes for a p-type ferromagnetic III–V DMS in which magnetization saturates and the simplified model presented here provides a quantitative argument that for a certain range of interpolaron distances the interactions between BMPs can stabilize ferromagnetism. In figure 3, curve (b) was calculated for values of parameters corresponding to the p–d exchange case: \( N_0β = -1.2 \text{ eV} \), \( ε = 11 \), and \( m^*/m_0 = 0.5 \) (the values of the two last parameters may be attributed to GaAs, cf [32] and references therein), confirming our statement. Nevertheless, the nature of the hole wavefunction in zinc-blende semiconductors is more complicated than that used here for the s-type donor state. It should be stressed also that an additional reservation concerning the application of the BMPM model to III–V Mn-based DMS is based on the fact that now Mn atoms are not isovalent with the Ga\(^{3+}\) cations. Namely, each Mn\(^{2+}\) substituting for the Ga\(^{3+}\) cation introduces thus one hole. Therefore, our considerations may be applied only to a highly compensated III–V DMS. Nonetheless, the appropriateness of the last restriction has a solid empirical ground, as it has been proved that GaMnAs growth in the excess of As becomes highly compensated due to As antisite defects, each of which introduces two electrons, see e.g. [8]. In brief, the above considerations are only a crude representation of the acceptor BMPM case [27–31].

3.2. Collinear polarization clouds

Within our approach to the BMPM problem, we can write the resulting sixth-order eigenequation for matrix (28) in the following form:

\[
\Delta E \text{ dependence on interimpurity distance } R_{ab} \text{ for BMPM with an effective concentration } τ = 0.027 \text{ of magnetic ions. Lines } a \text{ and } b \text{ are calculated with material parameters corresponding to Cd}^{1–x}\text{Mn}_x\text{Se and Ga}^{1–x}\text{Mn}_x\text{As, respectively, within the s-state approximation for the impurity single-particle wavefunction.}
\]
and for \( \Delta = \Delta^- = \Delta^+ = 0 \), corresponding to the antiparallel orientation, and one with \( \Delta^+ = \Delta \) and \( \Delta^- = 0 \), for the parallel orientation of exchange fields. Calculations were carried out for four values of \( \Delta = 2k \overline{\Delta} \), where \( \overline{\Delta} \simeq 0.7 \) meV is the most probable value for the single BMP in Cd_{0.95}Mn_{0.05}Se. The parameter \( k \) emulates different materials (see main text).

This form does not allow for an analytical solution. The eigenvalue \( E_0 \) can be easily determined, but the presence of the term containing the vector product \((\Delta^+ \times \Delta^-)^2\) complicates calculations of the remaining five eigenvalues. However, at \( T = 0 \) K, when the system is in its ground state, one can expect that exchange fields are collinear, either parallel or antiparallel. In such situations, we may assume \( \Delta^+ \times \Delta^- = 0 \), which facilitates a factorization of the remaining part of equation. Practically, this leaves us with the solutions of a cubic and a quadratic equation. Formally, the cubic equation can be solved exactly, as in principle all roots of the third-order equation can be found analytically. However, the general solution has a complicated analytic form that precludes a further clear analysis in a clear form. Before deriving an approximate solution in this section, we plot in figure 4 the numerical results for the energy difference between the lowest energy eigenvalues for the collinear configurations of the exchange fields. Namely, for \( \Delta^- = \Delta \) and \( \Delta^+ = 0 \), corresponding to the antiparallel orientation, and for \( \Delta^- = \Delta \) and \( \Delta^+ = 0 \), for the parallel orientation of the exchange fields. Explicit calculations are carried out for three values of \( \Delta = 2k \overline{\Delta} \), where \( \overline{\Delta} \simeq 0.7 \) meV is the most probable value for the single BMP in Cd_{0.95}Mn_{0.05}Se, taking the values of the material parameters from [14] (and assuming that \( \overline{\Delta}(T = 0) \simeq \overline{\Delta}(T = 1.5\) K)) and \( k = 1, 4 \) and 9. As can be seen from figure 4, a weak ferromagnetic interaction is very small and can be identified for exchange fields \( \Delta \) of magnitude larger then 9 meV. Note that the exchange fields are fluctuating quantities, so this does not mean that the ground state of Cd_{0.95}Mn_{0.05}Se can be ferromagnetic.

Returning now to the approximate solutions of equation (31), we can overcome the above-mentioned difficulty. For this purpose we regard the expression in the internal \( \{ \cdot \} \) as a function of \( E \), say \( F(E) \), and write:

\[
F(E) = G(E) + A,
\]

where

\[
G(E) \equiv [(L + 2j - E)(L_6 + 2j - E) - \gamma^2 + (\Delta^-)^2)](E - L),
\]

and

\[
A \equiv (\Delta^-)^2(L_6 + 2j - L).
\]

Now, it is easy to observe that the quantity \( A \), which is independent of the energy \( E \), influences \( F(E) \) through an upward shift of \( G(E) \). Thus we need to analyze properly \( G(E) \) and take into account approximately the presence of \( A \). Namely, we expand \( F(E) \) in terms of a Taylor series for each eigenvalue separately around the zeros of \( G(E) \):

\[
F(E, \theta) = A + \frac{\partial G(E)}{\partial E} \bigg|_{E=E_{0i}} (E - E_{0i}) = A + \frac{1}{2} \frac{\partial^2 G(E)}{\partial E^2} \bigg|_{E=E_{0i}} (E - E_{0i})^2 + \cdots = 0.
\]

We solve those equations to the first order and obtain:

\[
E_i \approx E_{0i} - \frac{A}{\partial G(E,E_{0i})},
\]

for \( i = 1, 4 \) and 5. (36).

The six zero-order eigenvalues \( E_{0i} \) are:

\[
\begin{pmatrix}
L \\
L + \Delta^+ \\
L - \Delta^- \\
\frac{1}{2}[(L + L_6) + 4j + \sqrt{(L - L_6)^2 + 4(\gamma^2 + (\Delta^-)^2)}] \\
\frac{1}{2}[(L + L_6) + 4j - \sqrt{(L - L_6)^2 + 4(\gamma^2 + (\Delta^-)^2)}] \\
L_6
\end{pmatrix}.
\]

An explicit analysis shows that the nature of the ground state is determined again by the sign of \( \Delta E = E_5 - E_3 \), expressing the difference between antiparallel and parallel configurations of the exchange fields. Moreover, for \( \Delta^- = 0 \), the eigenvalues have the same functional form as in the exact solution, except that now \( \Delta^+ \) is not calculated for the saturated situation.

3.2.1. Particular case: asymptotic solution for \( R_{ab} \rightarrow \infty \).

In the limit \( R_{ab} \rightarrow \infty \), we can neglect the terms describing interactions between BMPs and the two ionic configurations. Naturally, in the resulting expression \( \Delta^+ \) and \( \Delta^- \) cannot be neglected. Next, we diagonalize \( H_{R_{ab} \rightarrow \infty} \), i.e. write:

\[
H_{R_{ab} \rightarrow \infty} \tilde{V} = E \tilde{V}.
\]

This equation leads to the related fourth-order equation of the form:

\[
0 = [(2E_{\infty} - E)^2 - (\Delta^+)^2][(2E_{\infty} - E)^2 - (\Delta^-)^2] - (\Delta^+)^2(\Delta^-)^2 \sin^2(\theta),
\]

for the parallel orientation of exchange fields. Calculations were carried out for four values of \( \Delta = 2k \overline{\Delta} \), where \( \overline{\Delta} \simeq 0.7 \) meV is the most probable value for the single BMP in Cd_{0.95}Mn_{0.05}Se. The parameter \( k \) emulates different materials (see main text).
where $E_\infty = e_a = e_b$. The corresponding four eigenvalues are:

\[
\begin{align*}
2E_\infty + \frac{1}{\sqrt{2}} & \left[ \sqrt{2(\Delta_a^2 + \Delta_b^2)} + 2\sqrt{\Delta_a^2 \Delta_b^2 - 4\Delta_a^2 \Delta_b^2 \cos^2(\theta)} \right], \\
2E_\infty - \frac{1}{\sqrt{2}} & \left[ \sqrt{2(\Delta_a^2 + \Delta_b^2)} - 2\sqrt{\Delta_a^2 \Delta_b^2 - 4\Delta_a^2 \Delta_b^2 \cos^2(\theta)} \right], \\
2E_\infty + \frac{1}{\sqrt{2}} & \left[ \sqrt{2(\Delta_a^2 + \Delta_b^2)} - 2\sqrt{\Delta_a^2 \Delta_b^2 - 4\Delta_a^2 \Delta_b^2 \cos^2(\theta)} \right], \\
2E_\infty - \frac{1}{\sqrt{2}} & \left[ \sqrt{2(\Delta_a^2 + \Delta_b^2)} + 2\sqrt{\Delta_a^2 \Delta_b^2 - 4\Delta_a^2 \Delta_b^2 \cos^2(\theta)} \right].
\end{align*}
\] (40)

Our asymptotic solutions still depend formally on the angle $\theta$ between $\Delta^+$ and $\Delta^-$, whereas the corresponding solutions for the two isolated BMPs have the form:

\[
\begin{align*}
2E_\infty - \frac{1}{2}(\Delta_a + \Delta_b), \\
2E_\infty - \frac{1}{2}(\Delta_a - \Delta_b), \\
2E_\infty + \frac{1}{2}(\Delta_a - \Delta_b), \\
2E_\infty + \frac{1}{2}(\Delta_a + \Delta_b).
\end{align*}
\] (41)

are clearly free of such angular dependence, which reflects the uncorrelated character of the spatial orientation of the polaron fields. In appendix B we prove the equivalence of our solutions, given by equation (40), with that for the two isolated polarons. Note that, for finite interpolaron distances, the limit $\Delta \to 0$ leads also to a correct solution for the hydrogenic-like molecular state of two impurity electrons with effective Bohr radius $a_B$.

4. Thermodynamics

4.1. The method

Thermodynamic fluctuations of the localized spins within the finite BMPM volume may strongly influence the behavior of the system. Having determined the eigenvalues, we can construct the free energy of the BMP pair. To determine these properties we extend the previous approach [14] devised for a single BMP. For the case of single BMP, the thermodynamics has been derived by including the contribution coming from localized magnetic moments starting from the Ginzburg–Landau Hamiltonian:

\[
H_3[M] = \int \left[ \frac{1}{2\kappa} \sum_{j=1}^{3} |\nabla M_j(r)|^2 + \frac{1}{2\chi} M^2(r) \right] d^3 r, \quad (42)
\]

where the two phenomenological parameters, $\kappa$ and $\chi^{-1}$, are the exchange stiffness constant and the inverse static susceptibility, respectively. For this case, the probability distribution of the exchange field has been found in the form:

\[
P(\Delta) \equiv \int D\mathbf{M}(r) P[\mathbf{M}(r)] \delta(\Delta - \Delta[\mathbf{M}(r)]), \quad (43)
\]

where the probability distribution of $\mathbf{M}(r)$ is defined as:

\[
P[\mathbf{M}(r)] = Ce^{-\beta H_3}e^{-\beta \Delta F}, \quad (44)
\]

and $C$ is a normalization constant and $\beta \equiv (k_B T)^{-1}$ with $k_B$ being the Boltzmann constant. The functional integration in equation (43) expresses a summation of the contributions coming from all space profiles $[\mathbf{M}(r)]$ of magnetization contributing to a given value of the exchange field $\Delta$. This necessitates the functional integration over all possible ‘paths’ of $[\mathbf{M}(r)]$, with the probability density $P[\mathbf{M}]$ [14].

Generalization of equations (42) and (43) to the $N$-polaron case leads to the following $N$-component Ginzburg–Landau Hamiltonian:

\[
H_3[M_1, \ldots, M_N] = \sum_{i=1}^{N} \int d^3 r_i \left[ \frac{1}{2\kappa} \sum_{j=1}^{3} |\nabla M_j(r_i)|^2 + \frac{1}{2\chi} |M_i(r_i)|^2 \right]. \quad (45)
\]

Next, we define the probability distribution of exchange fields $\Delta_i$, with $i = 1, \ldots, N$, in the following form:

\[
P(\Delta_1, \ldots, \Delta_N)
\]

\[
= C' \int \prod_{i}^{N} \mathcal{D} \mathbf{M}_i e^{-\beta H_3} \delta(\Delta_i - \Delta[M_i]) e^{-\beta \Delta F_T}, \quad (46)
\]

where $\Delta F_T$ is the electronic part of the total free energy of the $N$-polaron system, $C'$ is now the overall normalization factor, and functions $\Delta[M_i]$ are those presented in $\Delta F_T$. The transformation made in equation (46) defines our dynamic variables $\Delta_i$ and the functional integrations in this equation can be carried out for each $\Delta[M_i]$ with the methodology developed in [14].

4.2. Results

When coupling between BMPs is important, the two isolated BMPs problem transforms now to the problem of interacting polarons with three exchange fields: $\Delta^+$, $\Delta^-$, and $\Delta_{ab}$. The thermodynamics of this system can be derived from equation (46), which, after executing the functional integrations, provides the physical free energy:

\[
F = -k_B T \sum \int d^3 \Delta^+ d^3 \Delta^- d^3 \Delta_{ab} P(\Delta^+, \Delta^-, \Delta_{ab}), \quad (47)
\]

where the probability distribution of the exchange fields $\Delta^+, \Delta^-$ and $\Delta_{ab}$ is:

\[
P(\Delta^+, \Delta^-, \Delta_{ab}) = C'' \exp \left\{ -\frac{(\Delta^+)^2}{8\varepsilon_+ k_B T} \right\} \times \exp \left\{ -\frac{(\Delta^-)^2}{8\varepsilon_- k_B T} \right\} \exp \left\{ -\frac{(\Delta_{ab})^2}{8\varepsilon_{ab} k_B T} \right\} \times \sum_{i=1}^{6} \exp \left\{ -\frac{E_i}{k_B T} \right\}. \quad (48)
\]
and $E_i$ are the eigenvalues of equation (A.25), whereas the three new parameters, $\varepsilon_+\varepsilon_-\varepsilon$ and $f_{ab}$, are defined as follows:

$$
\varepsilon_{\pm}(R_{ab}) = \frac{1}{4(g\mu_B)^2} \int d^3r|w_a(r)|^2 \pm |w_b(r)|^2 \frac{d^3r}{2}, \tag{49}
$$

and

$$
\varepsilon_{ab}(R_{ab}) = \frac{1}{4(g\mu_B)^2} \int d^3r[w_a(r)w_b(r)] \frac{d^3r}{2}, \tag{50}
$$

where the equations in the respective last lines define $f_{\pm}(R_{ab})$ and $f_{ab}(R_{ab})$, respectively. Note that the material parameters dependent $\varepsilon_\pm\varepsilon_-$ and $\varepsilon_{ab}$ correspond to the single-polaron parameter $\varepsilon_p$ of DS [14]. Their $R_{ab}$ dependence is crucial, as it determine the fluctuations of either $\Delta^+, \Delta^-$ or $\Delta_{ab}$. In figure 5 we plot this dependence. One sees that, for all finite distances $f_+ \geq f_-$, i.e. the field responsible for the triplet configurations of the two impurity electrons is dominant. One can also observe that $f_{ab}$ is a small quantity for all distances, justifying our assumption that the presence of $\Delta_{ab}$ can be neglected at least for $R_{ab} > 3a_B$. Note also, that even though the parameter $\rho \to \infty$ in the limit $R_{ab} \to 0$, the corresponding functions $f_+(R_{ab})$ and $f_-(R_{ab})$ are then finite (see also below).

The dominating character of $\varepsilon_+$ may result in the ferromagnetic-ground-state appearance of the BMPM. However, to prove that explicitly we have to include also the effect of the thermodynamic fluctuations of exchange fields on the eigenvalues $[E_i]$. This can be done by determining the most probable values of the fields $\Delta^+$ and $\Delta^-$. For that purpose, we consider first the collinear solution at $\rightarrow 0$ K. In analogy to the single-polaron theory, we can now calculate the most probable values $\Delta^+$ and $\Delta^-$ from the conditions:

$$
[\partial P(\Delta^+, \Delta^-)/\partial \Delta^+]|_{\Delta^-} = [\partial P(\Delta^+, \Delta^-)/\partial \Delta^-]|_{\Delta^+} = 0, \tag{51}
$$

which lead to the following system of two coupled equations:

$$
\Delta^+ = \left(1 + B\Delta^- - 4\varepsilon_+\varepsilon_-ight) = 0, \tag{52}
$$

and

$$
\Delta^- = \left(1 + B^{-1}\Delta^- + 4\varepsilon_- \frac{d}{d\Delta^-}E_3(\Delta^-)\right) = 0, \tag{53}
$$

where $E_3$ stands for the lowest energy solution of the cubic equation $F(E) = 0$ and

$$
B = B(T) \equiv \exp\left(-\frac{E_3}{k_B T}\right) / \exp\left(-\frac{E_3}{k_B T}\right). \tag{54}
$$

In the limit $T = 0$, and depending on the value of $\Delta E = E_5 - E_3$, the $B$ may be equal to 0, 1 or $\infty$, leading to the following three pairs of solutions:

(i) $\Delta^- = -2\varepsilon_- \frac{d}{d\Delta^-}E_3(\Delta^-)$, and $\Delta^- = 2\varepsilon_+$, for $\Delta E = 0$, \tag{55}

(ii) $\Delta^- = 4\varepsilon_+$, and $\Delta^- = 0$, for $\Delta E > 0$, \tag{56}

and

(iii) $\Delta^- = 0$ and $\Delta^- = -4\varepsilon_- \frac{d}{d\Delta^-}E_3(\Delta^-)$, for $\Delta E < 0$. \tag{57}

Therefore, the driving interaction which at $\rightarrow 0$ aligns individual polaron polarization clouds can be defined in the following form:

$$
\Delta E \equiv E_5 \text{ (iii)} - E_3 \text{ (ii)} . \tag{58}
$$

The condition $\Delta E(R_{ab}) = 0$ defines the critical distance $R_c$; for $R_{ab} > R_c$ the ground state is ferromagnetic. For negative values this expression yields the ground state belonging to eigenvalue $E_3$, i.e. to a mixture of states belonging to $\Delta_{tot} = 0$ (singlets $[4]$, $[5]$ and triplet $[1]$) states of impurity electrons. Whereas, for $\Delta E(R_{ab}) > 0$ it leads to ferromagnetic, ($\Delta_{tot} = -1$) ground state with the corresponding eigenvalue $E_3$ (belonging to [3]).

We can now identify the material parameters responsible for the nature of the ground state. Namely, the condition $\Delta E > 0$, leading to appearance of ferromagnetic ground state, occurs if $\Delta^- = 4\varepsilon_+$ is sufficiently large. Therefore, the parameter $\chi$, which contains all the key material parameters that determine $R_c$, can be defined as:

$$
\chi = \frac{k}{\varepsilon_+\varepsilon_-} \frac{d^2 m_0}{4(g\mu_B)^2 m^*}, \tag{59}
$$

where $k$ is a numerical parameter allowing one to differentiate by material by defining $\chi(k = 1) = \xi_0$ as that corresponding to $Cd_{0.95}Mn_{0.05}Se$. We can now analyze the expression for $\Delta E$ numerically. The results for $\Delta E(R_{ab})$ are presented in figure 6, for the three values of $k = 1, 4$ and 9, allowing a direct comparison with the results of figure 4, the latter
containing the numerical results obtained for non-fluctuating $\Delta^+$ and $\Delta^-$. Note a huge enhancement in the strength of the ferromagnetic interaction due to the inclusion of the thermodynamic fluctuations of the exchange fields. The key factor causing such a spectacular enhancement is the difference in the most probable value of $\Delta^+$ (ii) and $\Delta^-$ (iii), governed by $\varepsilon_{a}(R_{ab})$ and $\varepsilon_{b}(R_{ab})$, respectively. From figure 6 one can see that the value $k = 9$ is now sufficient to induce a ferromagnetic ground state.

So far we have treated the magnetic susceptibility $\chi$ of localized magnetic ions as a material parameter neglecting its strong temperature dependence. In Cd$_{0.95}$Mn$_{0.05}$Se and at low temperature, $\chi$ takes the form of Curie–Weiss law, $\chi = C_M/(T + \Theta_0)$, with $\Theta_0 = 1.2$ K. Properties of the system at low temperatures may be deduced from the analysis of the averages of $\Delta^+$ and $\Delta^-$, as they reflect the properties of the most probable values $\Delta^+$ and $\Delta^-$ expressed via conditions (i)–(iii). In order to see that within our approach the ferromagnetic configuration of the impurity electron is possible in general (beyond the collinear approximation), we have shown in figure 7 the numerical averages of $\langle \Delta^+ \rangle$ and $\langle \Delta^- \rangle$ versus $T$ for exemplary value of the parameter $k = 25$. Such a value of $k$ for n-type DMS is naturally outside the accessible range of material parameter, but for the p-type DMS the p-d exchange is about 4–5 times stronger than that for the n-type, then such high value of $k$ is justified. For this value of $k$ one finds $\Delta E \approx 1.7$ meV ($\chi$ calculated at 2 K, with $\Theta_0 = 1.2$ K and for $R_{ab} = 3a_B$), which allows for a very rough upper estimate of the Curie temperature $T_C$, in the mean field approximation for a 3D cubic BMP lattice, and would be of the order of $T_C \sim (1/3)z\Delta E z_0(s + 1)/k_B \sim 60$ K, where $z = 6$ is the number of nearest neighbors.

For completeness, in figure 8 we plot the temperature dependence of the average values of $\langle \Delta^+ \rangle$ and $\langle \Delta^- \rangle$, taking the parameters for Cd$_{0.95}$Mn$_{0.05}$Se at $R_{ab} = 2$ and 4.25 $a_B$. As one can see, the numerical results confirm that for this material a nonmagnetic ($\delta_{00} = 0$) configuration of the impurity electrons is always stable.

5. Conclusions

In this work we have developed the theory of the bound-magnetic-polaron molecule (BMPM) consisting of two bound magnetic polarons (BMPs) which are arbitrarily separated, and the spin interactions are treated within a rotationally invariant approach. Their mutual interaction is accounted for accurately within the molecular impurity states. We have also succeeded in deriving the thermodynamics of these states and analyzed their properties at nonzero temperature. We found that the fluctuations can strongly stabilize the spin-triplet configuration, a precursor state of ferromagnetic ordering in a many-impurity system. The important role of accurately including the impurity-electron wavefunctions has been demonstrated. Furthermore, the microscopic interpretation of the WBD model [21, 22] Hamiltonian parameters has been provided. Also, the spin-triplet state of BMPM emulates the hydrogen-molecule-like parallel spin configuration.

Generally, our model of BMPM in DMS confirms the possibility of the appearance of a ferromagnetic ground state for certain DMS materials and for appropriate distances between polarons (appropriate concentration of impurities). One of the important and new results of our approach is
a direct incorporation of material parameters into the BMP molecule model. The numerical calculation presented here for the BMP molecule embodied in Cd_{0.95}Mn_{0.05}Se shows that, for all interpolaron distances, the polaronic molecule ground state spin configuration is \( \mathbf{s}^{\text{tot}} = 0 \) for the impurity electrons. Then, in this DMS a nonmagnetic ground state of BMP molecule is always stable. On the other hand, our model predicts a ferromagnetic (spin-triplet) ground state of the BMP molecule for material parameters corresponding to p-type DMS. The nature of the ground state depends on the magnitude of the parameter \( \alpha \), defined by equation (58), which is proportional to the magnetic susceptibility of the spin system of the DMS. Therefore, the RKKY interaction responsible for the low-temperature ferromagnetic ordering in p-type DMSs enhances the ferromagnetic ground state of BMPM when approaching the critical region from the high-temperature side. However, the results for the p-type DMS must be analyzed more carefully by taking into account the non-s-type character of the BMP carrier wavefunction, as well as non-Gaussian contributions to the free energy functional. Such a formulation of BMPM is possible as an extension of the present approach. We should be able to see progress along this line in the near future.

**Acknowledgments**

HB is grateful for the financial support in the frame of the project from the National Science Centre no. N N508 626840. JS thanks the Foundation for Polish Science (FNP) for financial support of the project TEAM, as well as the Ministry of Science and Higher Education, Grant No. N N202 128736.

**Appendix A. Matrix representation of \( H_1 \) within the first quantization formalism**

We assume that the exchange field acting on impurity electrons is relatively weak and that orbital moments can be disregarded. As a consequence, the two-impurity electron wavefunction may by written as a product of functions, which depend on the spatial and the spin variables separately, i.e., it is of the form:

\[
\Psi_c(\mathbf{S}_i); r_1, r_2, \sigma_1, \sigma_2 = \chi_S(\mathbf{S}_i) \chi_{\sigma_1 \sigma_2} \Psi_c(r_1, r_2). \tag{A.1}
\]

where \( \chi_S \) is the wave function of the spins, and \( \chi_{\sigma_1 \sigma_2} \) is the spin part of that for the carriers, whose spatial wavefunction is \( \Psi_c(r_1, r_2) \), where \( c \) runs over all pairs \((ab, aa, bb)\). We take the two-electron functions as composed of orthogonal molecular wavefunctions \( \psi_{ab}(r_1) \) introduced by equation (4). These orthogonal single-particle wavefunctions are used to construct the two-particle six-dimensional space. The space part is identical to that for the hydrogenic-like molecule. Therefore, we can write down directly the following six two-electron wavefunctions:

\[
\Psi_{1,4}\chi_{1,4} = \frac{1}{\sqrt{2}}[\psi_a(r_1)\psi_b(r_2) \mp \psi_a(r_2)\psi_b(r_1)] \\
\times \frac{1}{\sqrt{2}}[\chi_{1/2}(1)\chi_{-1/2}(2) \pm \chi_{-1/2}(1)\chi_{1/2}(2)], \tag{A.2}
\]

for the spin-triplet state with \( \mathbf{s}^{\text{tot}} = 0 \) and the singlet state, respectively,

\[
\Psi_{2,3}\chi_{2,3} = \frac{1}{\sqrt{2}}[\psi_a(r_1)\psi_b(r_2) - \psi_a(r_2)\psi_b(r_1)] \\
\times \{\chi_{\pm 1/2}(1)\chi_{\pm 1/2}(2)\}, \tag{A.3}
\]

for the remaining triplet states with \( \mathbf{s}^{\text{tot}} = \pm 1 \), and

\[
\Psi_{5,6}\chi_{5,6} = \frac{1}{\sqrt{2}}[\psi_a(r_1)\psi_b(r_2) \pm \psi_a(r_2)\psi_b(r_1)] \\
\times \frac{1}{\sqrt{2}}[\chi_{1/2}(1)\chi_{-1/2}(2) - \chi_{-1/2}(1)\chi_{1/2}(2)], \tag{A.4}
\]

for the remaining two ionic singlet states.

Next, we determine the matrix representation of \( H_1 \) by averaging over the spatial coordinates, but keeping the carrier spin still in the operator form:

\[
\langle \Psi_{n}\mid H_1 \mid \Psi_m \rangle = \frac{\alpha}{\mu B} \left[ \bar{\mathbf{s}}_1 \cdot \int \Psi_{n}^{\ast}(\mathbf{r}_1)\Psi_m d^3r_1 d^3r_2 \right. \\
\left. + \bar{\mathbf{s}}_2 \cdot \int \Psi_{n}^{\ast}(\mathbf{r}_2)\Psi_m d^3r_1 d^3r_2 \right]. \tag{A.5}
\]

Taking into account the symmetry properties of the spatial wavefunction with respect to the transposition of the carrier coordinates, the matrix elements of \( H_1 \) can be recast to the form:

\[
\langle \Psi_{n}\mid H \mid \Psi_m \rangle = \bar{\mathbf{s}}_1 \cdot \Delta_1[\mathbf{M}, \Psi_n, \Psi_m] + \bar{\mathbf{s}}_2 \cdot \Delta_2[\mathbf{M}, \Psi_n, \Psi_m] \\
= \bar{\mathbf{s}}_1 \cdot \text{sign}(\Psi_n)\text{sign}(\Psi_m)\bar{\mathbf{s}}_2 \cdot \Delta[\mathbf{M}, \Psi_n, \Psi_m], \tag{A.6}
\]

where the factor \( \text{sign}(\Psi_n)\text{sign}(\Psi_m) \) = \( \pm 1 \) expresses the parity of the corresponding wavefunction. The exchange fields, \( \Delta_l[\mathbf{M}, \Psi_n, \Psi_m] \), with \( l = 1, 2 \), are thus defined as:

\[
\Delta_l[\mathbf{M}, \Psi_n, \Psi_m] = \frac{\alpha}{\mu B} \int \Psi_{n}^{\ast}(\mathbf{r})\Psi_m d^3r_1 d^3r_2. \tag{A.7}
\]

Note that the fields \( \Delta_l[\mathbf{M}, \Psi_n, \Psi_m] \) are equal to \( \Delta_l[\mathbf{M}, \Psi_n, \Psi_m] \), except for the sign. An explicit calculation provides a remarkable reduction of the number of the exchange fields. Namely, only the following four appear in the final BMP pair Hamiltonian:

\[
\Delta^\pm[\mathbf{M}] = \frac{\alpha}{\mu B} \int d^3r [w_a(r)]^2 \pm [w_b(r)]^2 \mathbf{M}(r), \tag{A.8}
\]

\[
\Delta_{ab}[\mathbf{M}] = \frac{\alpha}{\mu B} \int d^3r w_a(r)^*w_b(r)\mathbf{M}(r), \tag{A.9}
\]

and

\[
\Delta_{ba}[\mathbf{M}] = \frac{\alpha}{\mu B} \int d^3r w_b(r)^*w_a(r)\mathbf{M}(r). \tag{A.9}
\]

The labeling of the exchange fields \( \Delta^+ \) and \( \Delta^- \) originates from the wavefunction parities (e.g. ‘+’ corresponds to the triplet–triplet matrix elements and ‘−’ to the singlet–triplet ones).
Here we outline the details of calculations of the matrix elements \( \langle \Phi_{\text{tot}}^{\xi} | H_{\text{BMP}} | \Phi_{\text{tot}}^{\xi} \rangle \) in the singlet–triplet basis. According to the definition given in the text: \( \Phi_{\text{tot}}^{\xi} = \Psi_{\text{t}} \chi_{\text{tot}}^{\xi} \) and, taking into account that Hamiltonian \( H_{\text{B}} \) does not contain explicitly the carrier spin variables, one can write:

\[
\langle \Phi_{\text{tot}}^{\xi} | H_{\text{BMP}} | \Phi_{\text{tot}}^{\xi} \rangle = (\chi_{\text{tot}}^{\xi} | H_{\text{BMP}} | \Psi_{\text{v}}) | \chi_{\text{tot}}^{\xi} \rangle = E_{\tau} \delta_{\tau} + (\chi_{\text{tot}}^{\xi} | (\mathbf{S}_{1} + \mathbf{S}_{2}) \cdot \Delta^{+} | \chi_{\text{tot}}^{\xi} \rangle,
\]

(A.10)

where \( \mathbf{S}_{1} = \mathbf{s}_{1} \otimes 1, \mathbf{S}_{2} = 1 \otimes \mathbf{s}_{2} \) are the carrier spin operators and the corresponding spinors \( \chi_{\text{tot}}^{\xi} \) are explicitly defined below:

\[
\begin{align*}
\chi_{+}^{0,0} & = \frac{1}{\sqrt{2}} (\chi_{1/2}(1) \otimes \chi_{-1/2}(2) - \chi_{-1/2}(1) \otimes \chi_{1/2}(2)), \\
\chi_{-}^{0,0} & = \frac{1}{\sqrt{2}} (\chi_{1/2}(1) \otimes \chi_{-1/2}(2) + \chi_{-1/2}(1) \otimes \chi_{1/2}(2)), \\
\chi_{1}^{1} & = \chi_{1/2}(1) \otimes \chi_{1/2}(2), \\
\chi_{-1}^{1} & = \chi_{-1/2}(1) \otimes \chi_{1/2}(2).
\end{align*}
\]

(A.11) - (A.13)

and

\[
\chi_{-1}^{1} = \chi_{-1/2}(1) \otimes \chi_{1/2}(2).
\]

(A.14)

To shorten the notation, the tensor product symbol is omitted below. We recall that we have fixed the direction of the spin quantization axis parallel to the local exchange field \( \Delta^{-} [\mathbf{M}] \).

Generally, one can assume that the selected coordinate system direction of \( \Delta^{-} [\mathbf{M}] \) is described by the angles \( \theta \) and \( \phi \). Therefore, evaluation of equation (A.10) with these exchange fields is simplest in the appropriately rotated spin basis, namely:

\[
\begin{align*}
\chi_{1/2} & = \chi_{\theta \phi} \begin{bmatrix} \cos \frac{\theta}{2} & \sin \frac{\theta}{2} \\ -\sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{bmatrix} \chi_{\xi \phi} = \chi_{\theta \phi} \chi_{\xi \phi} \sin \frac{\theta}{2} \cos \frac{\theta}{2}, \\
\chi_{-1/2} & = \chi_{\theta \phi} \begin{bmatrix} \cos \frac{\theta}{2} & \sin \frac{\theta}{2} \\ -\sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{bmatrix} \chi_{\xi \phi} = \chi_{\theta \phi} \chi_{\xi \phi} \cos \frac{\theta}{2} + \chi_{\theta \phi} \chi_{\xi \phi} \sin \frac{\theta}{2},
\end{align*}
\]

(A.15) - (A.16)

where the angles \( \theta \) and \( \phi \) describe the spin state with an arbitrary direction opposite to that defined by \( \theta \) and \( \phi \). From equation (A.10), the diagonal matrix elements can be written as follows:

\[
\langle \chi_{\text{tot}}^{\xi} | H_{\text{BMP}} | \chi_{\text{tot}}^{\xi} \rangle = E_{\tau} + (\chi_{\text{tot}}^{\xi} | (\mathbf{S}_{1} + \mathbf{S}_{2}) \cdot \Delta^{+} | \chi_{\text{tot}}^{\xi} \rangle.
\]

(A.17)

Note that \( \langle \chi_{\text{tot}}^{0,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{0,0} \rangle = E_{\tau} \), because \( \chi_{\text{tot}}^{0,0} \) transforms as a scalar under rotations, i.e.:

\[
\begin{align*}
\chi_{1/2}(1) \chi_{-1/2}(2) - \chi_{-1/2}(1) \chi_{1/2}(2) & = \chi_{\phi}(1) \chi_{\overline{\phi}}(2) - \chi_{\overline{\phi}}(1) \chi_{\phi}(2), \\
\chi_{-1/2}(1) \chi_{1/2}(2) + \chi_{1/2}(1) \chi_{-1/2}(2) & = \chi_{\phi}(1) \chi_{\overline{\phi}}(2) + \chi_{\overline{\phi}}(1) \chi_{\phi}(2).
\end{align*}
\]

(A.18)

Also, the matrix element \( \langle \chi_{\text{tot}}^{1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{-1,0} \rangle \) can be computed in the following manner:

\[
\begin{align*}
\langle \chi_{\text{tot}}^{1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{-1,0} \rangle & = \frac{1}{2} \sum_{i} \langle \chi_{1/2}(1) \chi_{-1/2}(2) + \chi_{-1/2}(1) \chi_{1/2}(2) | H_{\text{BMP}} | \chi_{1/2}(1) \chi_{-1/2}(2) + \chi_{-1/2}(1) \chi_{1/2}(2) \rangle \\
& = \frac{1}{2} \sum_{i} \sum_{\sigma,\sigma'} \langle \chi_{\sigma}(1) \chi_{\sigma'}(2) | (\mathbf{S}_{i} \cdot \Delta^{-} | \chi_{\sigma}(1) \chi_{\sigma'}(2) \rangle \\
& = \frac{1}{2} \sum_{i} \sum_{\sigma} \langle \chi_{\sigma}(i) | \mathbf{S}_{i} \cdot \Delta^{-} | \chi_{\sigma}(i) \rangle \Delta^{-}
\end{align*}
\]

(A.19)

and thus:

\[
\langle \chi_{\text{tot}}^{-1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{1,0} \rangle = \langle \chi_{\text{tot}}^{1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{-1,0} \rangle. \tag{A.20}
\]

(A.20)

The nonvanishing off-diagonal matrix elements containing \( \Delta \) are:

\[
\begin{align*}
\langle \chi_{\text{tot}}^{0,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{1,0} \rangle & = \Delta^{-} \langle \chi_{1/2}(1) \chi_{-1/2}(2) | (\mathbf{S}_{1} - \mathbf{S}_{2}) \chi_{1/2}(2) \rangle \chi_{\text{tot}}^{1,0} = \Delta^{-}, \tag{A.22}
\langle \chi_{\text{tot}}^{1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{1,1} \rangle & = \langle \chi_{\text{tot}}^{1,0} | (\mathbf{S}_{1} + \mathbf{S}_{2}) \cdot \Delta^{+} | \chi_{\text{tot}}^{1,1} \rangle \\
& = 2^{-1/2} \Delta^{+} \sin \theta \sin \phi, \tag{A.23}
\end{align*}
\]

(A.22) - (A.23)

and

\[
\langle \chi_{\text{tot}}^{1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{-1,1} \rangle = \langle \chi_{\text{tot}}^{1,0} | H_{\text{BMP}} | \chi_{\text{tot}}^{-1,1} \rangle^*. \tag{A.24}
\]

(A.24)

In a quite similar manner we computed the off-diagonal matrix elements containing \( \Delta_{ab} \).

Finally, the matrix representation of \( H_{1} \) in the six-dimensional basis of the two-electron wavefunctions is:

\[
\begin{pmatrix}
0 & \Delta_{+} \sin \theta \cos \phi & 0 & 0 & 0 & 0 \\
\Delta_{-} \sin \theta \cos \phi & 0 & 0 & 0 & 0 & 0 \\
0 & \Delta_{+} \sin \theta \sin \phi & 0 & 0 & 0 & 0 \\
\Delta_{-} \sin \theta \sin \phi & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

(A.25)
Appendix B. Equivalence of equation (35) with the solution for two isolated BMPs

In this appendix we demonstrate that our solution of the asymptotic case for $R_{ab} \to \infty$ is equivalent to that known for two isolated BMPs. Hence, we prove the equivalence between equations (40) and (41). To do this we need calculate first:

$$2 \Delta^+ \Delta^- \cos(\theta) = (\Delta^+ + \Delta^-)^2 - (\Delta^+)^2 - (\Delta^-)^2. \quad (B.1)$$

Taking now into account that:

$$(\Delta^+ + \Delta^-)^2 = 4 \Delta_n^2, \quad (B.2)$$

and

$$(\Delta^+)^2 + (\Delta^-)^2 = 2(\Delta_n^2 + \Delta_b^2), \quad (B.3)$$

one can find that:

$$\Delta^+ \Delta^- \cos(\theta) = \Delta_n^2 - \Delta_b^2. \quad (B.4)$$

Substituting equations (B.3)–(B.4) into equation (40) one obtains:

$$
\begin{align*}
&2E_\infty + \frac{1}{2}(\Delta_a + \Delta_b) \\
&2E_\infty - \frac{1}{2}(\Delta_a + \Delta_b) \\
&2E_\infty + \frac{1}{2}(\Delta_a - \Delta_b) \\
&2E_\infty - \frac{1}{2}(\Delta_a - \Delta_b)
\end{align*}
$$

(B.5)

These eigenvalues can be found also when taking an independent linear combination of the solutions for the two isolated polarons. This constitutes the equivalence between the two solutions.

References

[1] Jungwirth T, Sinowa J, Masek J, Kucera J and MacDonald A H 2006 Rev. Mod. Phys. 78 809
Dietl T et al 2000 Science 287 1019
[2] De Teresa J M et al 1997 Nature 386 256
[3] Coey J M, Venkatesan M and Fitzgerald C B 2005 Nature Mater. 4 173
[4] Kikoin K and Fleurov V 2006 Phys. Rev. B 74 174407
Kristajic P M, Peeters F M, Ivanov V A, Fleurov V and Kikoin K 2004 Phys. Rev. B 70 195215
[5] Jungwirth T et al 2007 Phys. Rev. B 76 125206
[6] Chapler B C et al 2011 Phys. Rev. B 84 081203(R)
[7] Bouzerar R, Bouzerar G and Ziman T 2006 Phys. Rev. B 73 024411
Bouzerar G, Ziman T and Kudrnovsky J 2005 Phys. Rev. B 72 125207
[8] Myers R C, Sheu B L, Jackson A W, Gossard A C, Schiffer P, Samarth N and Awschalom D D 2006 Phys. Rev. B 74 155203
[9] Govorov A O 2005 Phys. Rev. B 72 075358
Goverov A O 2005 Phys. Rev. B 72 075359
Zhang W, Dong T and Govorov A O 2007 Phys. Rev. B 76 075319
[10] Sellers I R et al 2010 Phys. Rev. B 82 195320
[11] Oszwaldowski R, żuć i and Petukhov A G 2011 Phys. Rev. Lett. 106 177201
[12] Lebedeva N, Holmberg H and Kuivalainen P 2008 Phys. Rev. B 77 245308
Lebedeva N, Varpula A, Novikov S and Kuivalainen P 2010 Phys. Rev. B 81 235307
Lebedeva N and Kuivalainen P 2009 Phys. Status Solidi b 246 1291
[13] Spalek J, Lewicki A, Tarnawski Z, Furdyna J K, Galazka R R and Oubusko Z 1986 Phys. Rev. B 33 3407
[14] Dietl T and Spalek J 1982 Phys. Rev. Lett. 48 355
Dietl T and Spalek J 1983 Phys. Rev. B 28 1548
[15] Ryabchenko S M and Semenov Y G 1983 Zh. Eksp. Teor. Fiz. 84 1419
Ryabchenko S M and Semenov Y G 1983 Sov. Phys.—JETP 57 825 (Engl. transl.)
Ryabchenko S M and Semenov Y G 1983 Sov. Phys.—JETP 57 825 (erratum)
[16] Heiman D, Wolff P A and Warnock J 1983 Phys. Rev. B 27 4848
Wolff P A and Warnock J 1984 J. Appl. Phys. 55 2300
[17] Golnik A, Ginter J and Gaj J 1983 J. Phys. C: Solid State Phys. 16 6073
[18] Nhung T H, Planel R, Benoît a la Guillaume C and Bhattacharjee A K 1985 Phys. Rev. B 31 2388
[19] Golnik A and Spalek J 1986 J. Magn. Magn. Mater. 54–57 1207
[20] Umehara M 2000 Phys. Rev. B 61 12209
[21] Wolff P A, Bhatt R N and Durst A C 1996 J. Appl. Phys. 79 5196
[22] Durst A C, Bhatt R N and Wolff P A 2002 Phys. Rev. B 65 235205
[23] Angelescu D E and Bhatt R N 2002 Phys. Rev. B 65 75211
[24] Bednarski H and Spalek J 2011 Acta Phys. Pol. A 120 967
[25] Spalek J, Podsialny J R, Wójcik M and Rycecz J 2000 Phys. Rev. B 61 15676
[26] Anderson P W 1959 Phys. Rev. B 115 2
Chao K A, Spalek J and Oleś A M 1977 J. Phys. C: Solid State Phys. 10 L271
[27] Berciu M and Bhatt R N 2003 Phys. Rev. Lett. 90 029702
Berciu M and Bhatt R N 2004 Phys. Rev. B 69 045202
[28] Kennett M P, Berciu M and Bhatt R N 2002 Phys. Rev. B 65 045207
Kennett M P, Berciu M and Bhatt R N 2002 Phys. Rev. B 65 115308
Berciu M and Bhatt R N 2002 Phys. Rev. B 66 085207
[29] Bney L and Gomez-Santos G 2003 Phys. Rev. B 68 115206
[30] Fiete G A, Zaran G, Damle K and Moca C P 2005 Phys. Rev. B 72 045212
Fiete G A, Zaran G and Damle K 2003 Phys. Rev. Lett. 91 097202
[31] Kaminski A and Das Sarma S 2002 Phys. Rev. Lett. 88 247202
Priour J and Das Sarma S 2006 Phys. Rev. Lett. 97 127201
[32] Yun P Y and Cardona M 1996 Fundamentals of Semiconductors (Berlin: Springer) p 70 and p 326