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Application of Pair Approximation to Bound-Magnetic-Polaron State in Diluted Magnetic Semiconductors

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We present a fully quantum-mechanical model of the bound magnetic polaron state in a diluted magnetic semiconductor based on the pair approximation to the exchange interaction among localized magnetic moments (spins). The model is applicable also to the limiting situation of the bound magnetic polaron magnetization due to the spins approaching saturation. On its basis, a simple extension of the Dietl–Spałek bound magnetic polaron theory is proposed and compared with the data for p-$Cd_{0.95}Mn_{0.05}Te$.

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1. Introduction

The bound magnetic polaron (BMP) is formed when an impurity charge carrier coupled to localized spins forms bound states in a diluted magnetic semiconductor (DMS). The importance of these nanoscale in size quantum objects have recently stimulated a growing interest in studies of ferromagnetic quantum dots [1–5], of ferromagnetic interaction between pair of coupled BMPs [6–8], as well as applied to novel materials such as ferromagnetic perovskites [9], diluted ferromagnetic oxides [10], and ferromagnetic DMSs [11, 12]. Here we develop a model of BMP in a DMS based on application of the pair approximation (PA) [13, 14]. Our work is motivated by the need to assess the appropriateness of a simple extension of the theory of BMP in DMS developed by Dietl and Spałek (DS) [15], which was developed taking into account the Gaussian thermodynamic fluctuations of the spins, to the limit of saturation of the magnetization due to those spins. Such situation takes place for example in p-type doped DMS or in the classic ferromagnetic semiconductor EuO:Eu. The extension comprises a formal replacement of the linear approximation to the system magnetization in DS by the magnetization itself.

The pair approximation accounts for the spin interactions in the Heisenberg form between the nearest neighbors (NN). In effect, the partition function of the system spins is factorized into the pair-contribution form. In practice, we consider Mn^{2+} based DMS with fcc lattice, with fraction x of all cationic sites occupied in a random fashion by the Mn^{2+} ions with spin $S = 5/2$. A single donor impurity is located in the center of the lattice and the range of interaction between the donor electron spin and magnetic ion spins is set to $5a_B$, where a_B is the effective Bohr radius.

For a given random distribution of Mn^{2+} ions the interacting pairs are determined numerically and accounted for within PA and by assuming additionally that the radial dependence of d–d exchange integral $J(R)$ can be parameterized by the power law in the following form $J(R) = J_0 R^{-6.8}$ [14]. Then, the partition function of BMP for this particular configuration of the spins is computed and the whole procedure repeated iteratively 10^3 times, starting with a new random configuration of Mn ions, in order to be able to perform the proper statistical averaging over the macroscopic spins configurations.

2. Modeling

2.1. Theoretical model — a reminder

We start by considering the impurity in DMS with a random distribution of localized magnetic moments $\boldsymbol{S}_i$ (spins) on the lattice sites labeled by index i. Apart from the effective-mass approximation, we assume also the spherical approximation to acceptor states which allow us to consider simultaneously the donor, as well as in the approximate manner, the acceptor type impurity [16, 17]. Thus we write the total wave function in the form [15]:

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$$\Psi(\{\boldsymbol{S}_i\};\boldsymbol{r},\sigma)=\chi\{\boldsymbol{S}_i\}\chi_\sigma u_0(r)\phi(r), \tag{1}$$

where $\chi\{\boldsymbol{S}_i\}$ is the wave function of spins, χ_σ is the spin part of that for the electron, $u_0(r)$ is the periodic part of the Bloch function (taken at Γ point) and $\phi(r)$ is the radial envelope function for the lowest $1s$ state of the impurity carrier

$$\phi(r)=\sqrt{\frac{1}{\pi a_{\mathrm{B}}^3}}\,\mathrm{e}^{-\frac{r}{a_{\mathrm{B}}}}, \tag{2}$$

with the effective Bohr radius a_{B}. Next, we write down the BMP Hamiltonian as composed of the three parts

$$H_{\mathrm{BMP}}=H_{\mathrm{sc}}+H_{s(p)-d}+H_{d-d}, \tag{3}$$

where H_{sc} contains contribution from impurity interactions (band states) within the host semiconductor, $H_{s(p)-d}$ — the $s(p)$–d exchange couplings of the donor electron to the localized spins and H_{d-d} is the exchange interaction among the spins. Explicitly, we take H_{sc} in usual form [18]:

$$H_{\mathrm{sc}}=-\frac{\hbar^2}{2m^*}\nabla^2-\frac{e^2}{\varepsilon}\frac{1}{r}, \tag{4}$$

where m^* is the effective mass, $\boldsymbol{r}$ is the impurity carrier position, e is the carrier charge, and ε is the static dielectric constant of the host semiconductor. $H_{s(p)-d}$ has the form of the spin exchange Hamiltonian

$$H_{s(p)d}=-\sum_i J_{\mathrm{c}}(\boldsymbol{r}-\boldsymbol{R}_i)\hat{\boldsymbol{S}}_i\cdot\hat{\boldsymbol{s}}-g^*\mu_{\mathrm{B}}\hat{\boldsymbol{s}}\cdot\boldsymbol{B}, \tag{5}$$

where J_{c} is the so-called Vonsovski–Zener exchange integral of the contact Fermi ($s(p)$–d) interaction between the localized spins $\{\hat{\boldsymbol{S}}_i\}$ and that of impurity carrier $\hat{\boldsymbol{s}}$, occupying the sites $\boldsymbol{R}_i$ and $\boldsymbol{r}$, respectively, g^* is the effective Landé factor, μ_{B} is the Bohr magneton and $\boldsymbol{B}$ is the external magnetic field. H_{d-d} contains the exchange interactions between the spins and their Zeeman energy

$$H_{d-d}=-\sum_{i,j}J_{ij}\hat{\boldsymbol{S}}_i\cdot\hat{\boldsymbol{S}}_j-g\mu_{\mathrm{B}}\sum_i\hat{\boldsymbol{S}}_i\cdot\boldsymbol{B}, \tag{6}$$

where J_{ij} is the corresponding exchange interaction integral and $g=2$ is the Landé factor for the spins. We assume also that the radial extension of d–d interaction $J_{ij}=J(R_{ij})$ can be parameterized by the power law in the following form: $J(R)=J_0R^{-6.8}$ [14]. This form has been advocated on the basis of the Bloembergen–Rowland mechanism. Parenthetically, this form can also parameterize in an approximated manner the distance dependence of the antiferromagnetic superexchange. The averaging of H_{BMP} with radial part of the carrier wave function, $u_0(r)\phi(r)$, influences both H_{sc} and $H_{s(p)-d}$, leads to the following expression for the effective Hamiltonian in the spin space:

$$\tilde{H}_{\mathrm{BMP}}\equiv H_{\mathrm{sc}}+H_{s(p)-d}=E_{\mathrm{D}}(a_{\mathrm{B}})-\gamma\sum_i|\phi(R_i)|^2\hat{\boldsymbol{S}}_i\cdot\hat{\boldsymbol{s}}-g^*\mu_{\mathrm{B}}\hat{\boldsymbol{s}}\cdot\boldsymbol{B}, \tag{7}$$

where

$$E_{\mathrm{D}}(a_{\mathrm{B}})=\frac{\hbar^2}{2m^*a_{\mathrm{B}}^2}-\frac{e^2}{\varepsilon a_{\mathrm{B}}}, \tag{8}$$

represents the donor mechanical energy, $\gamma=\langle u_0|J_{\mathrm{c}}(r)|u_0\rangle$ is the effective $s(p)$–d exchange constant (α for donors and β for acceptors), and $\hbar$ is the Planck constant divided by 2π. In this manner, we obtain finally, the BMP Hamiltonian in the spin-operator form, the starting point to any subsequent evaluation of BMP states and thermodynamics.

2.2. Pair approximation

Our principal aim is the application of the pair approximation to this Hamiltonian. The BMP possesses a natural spherical symmetry with the impurity at its center. It is valuable to exploit its property within PA. We consider the spins arrangement as divided into consecutive spin spheres. Thus, R_ν denotes distance to the ν-th coordination sphere with coordination number n_ν. Next, we introduce the spins pairing rule that the NN spins are paired starting from the center and preserve the radial symmetry. In effect, the partition function of the spins system is factorized to pair-contributions form. Additionally, we assume that the interaction between the impurity-carrier--spin and the localized magnetic moments can also be factorized to the form expressing interacting pairs of the spins. In effect, the obtained pair-approximated Hamiltonian H_ν of BMP for given pair can be written in the following form:

$$H_\nu=-\gamma\big(|\phi(R_\nu)|^2\hat{\boldsymbol{S}}_\nu+|\phi(R_{\mathrm{NN}_\nu})|^2\hat{\boldsymbol{S}}_{\mathrm{NN}_\nu}\big)\cdot\hat{\boldsymbol{s}}-2J(R_{\nu,\mathrm{NN}_\nu})\hat{\boldsymbol{S}}_\nu\cdot\hat{\boldsymbol{S}}_{\mathrm{NN}_\nu}-g\mu_{\mathrm{B}}(\hat{\boldsymbol{S}}_\nu+\hat{\boldsymbol{S}}_{\mathrm{NN}_\nu})\cdot\boldsymbol{B}, \tag{9}$$

where NN_ν denotes the nearest magnetic neighbor of ν, $R_{\mathrm{NN}\nu}$ is the distance from the center to NN_ν and R_{ν,NN_ν} is the distance between the nearest neighbors (ν and NN_ν). Finally, the BMP free energy can now be written in the standard form

$$F=E_{\mathrm{D}}-k_{\mathrm{B}}T\sum_{\{\nu\}}\ln\mathrm{Tr}\exp(-\beta H_\nu), \tag{10}$$

where the sum is running over all pairs of the spins.

To simplify computations we limit ourselves to the case with $\boldsymbol{B}=\boldsymbol{0}$ and derive analytical solution of the related eigenvalue problem. For this purpose, we chose for the $\{\nu,\mathrm{NN}_\nu\}$ pair the spin-state-space representation parameterized by the following quantum numbers: $s,(S_\nu S_{\mathrm{NN}\nu})S_{\nu,\mathrm{NN}\nu},J,M$ where $\hat{\boldsymbol{S}}_{\nu,\mathrm{NN}_\nu}=\hat{\boldsymbol{S}}_\nu+\hat{\boldsymbol{S}}_{\mathrm{NN}_\nu}$, $\hat{\boldsymbol{J}}=\hat{\boldsymbol{s}}+\hat{\boldsymbol{S}}_\nu+\hat{\boldsymbol{S}}_{\mathrm{NN}_\nu}$, and $M=-J\ldots J$. This assumption allows us to write the matrix representation of H_ν in the following diagonal form:

$$\begin{aligned}\langle s,S_{\nu,\mathrm{NN}\nu};JM|H_\nu|s,S_{\nu,\mathrm{NN}\nu};JM\rangle&=\Big\{-\gamma\frac{|\phi_\nu|^2+|\phi_{\mathrm{NN}_\nu}|^2}{4}\Big[J(J+1)\\&-S_{\nu,\mathrm{NN}\nu}(S_{\nu,\mathrm{NN}\nu}+1)-s(s+1)\Big]\\&-J_{\nu,\mathrm{NN}\nu}[S_{\nu,\mathrm{NN}\nu}(S_{\nu,\mathrm{NN}\nu}+1)-2S(S+1)]\Big\}\\&\times|s,S_{\nu,\mathrm{NN}\nu};JM\rangle.\end{aligned} \tag{11}$$

Such representation allows for a direct numerical evaluation, as discussed next.

3. Numerical results and comparison with experiment

Next, we apply the developed approach to assess the appropriateness of the simple extension of the BMP theory developed by Dietl and Spałek (DS) [15] to the situation with saturation of the magnetization due to the spins. For this purpose, we compare the expectation value of H_{sd} calculated in three different ways. Namely, we use the approach developed here, obtained according to the DS theory, and finally determined by replacement of the linear approximation to the system magnetization in the DS solution by the magnetization itself. Additionally, we compare numerical results with experimental data of Ref. [19] for acceptor type BMP in $Cd_{0.95}Mn_{0.05}Te$. Our and DS approaches are derived explicitly within the spherical approximation. Nonetheless, we apply them also to the acceptor-type BMP. Generally, this is not quite correct but we would like to test if the detailed treatment of spin fluctuations suffices to describe the system, independently of the detailed shape of the impurity-carrier wave function.

The expectation value of $\langle H_{s(p)-d}\rangle$ within DS can be calculated analytically, it has the following simple form:

$$\langle H_{s(p)-d}\rangle = 4s^2\varepsilon_{\rm p}\frac{4s^2\varepsilon_{\rm p}+3k_{\rm B}T}{4s^2\varepsilon_{\rm p}+k_{\rm B}T}, \tag{12}$$

with the following replacement:

$$\varepsilon_{\rm p} \equiv \left(\frac{\gamma}{g\mu_{\rm B}}\right)^2 \frac{\chi}{32\pi a_{\rm B}^3} \Rightarrow \frac{\gamma}{g\mu_{\rm B}}\langle M\rangle, \tag{13}$$

where $\varepsilon_{\rm p}$ is the principal parameter obtained within DS theory, χ is the magnetic susceptibility and $\langle M\rangle$ is the mean spin magnetization [15]. Let us note that within the replacement in Eq. (13) we extend formally DS approach to the situation when magnetization due to the spins can approach the saturated state. In order to calculate $\langle H_{s(p)-d}\rangle$, we need to fix values of the Hamiltonian parameters. For the BMP in p-$Cd_{0.95}Mn_{0.05}Te$, we have: $s = 3/2$ and $a_{\rm B} = 10$ Å according to Ref. [19], $x = 0.05$, $n = 6.8$, $J_0 = -7$ K [14], for the acceptor impurity.

All the calculated curves shown in Fig. 1 are adjusted to the experimental data with one parameter only. Namely, the exchange integral γ is treated as the adjustable parameter within our approach. The curves within DS approach are obtained with χ and M determined within PA (more precisely, with an extended version of PA which accounts also for the triple-spin configurations; see e.g. Refs. [14] and [20]) and are adjusted by the following parameterizations: $\varepsilon_{\rm p} = c\chi = cB_{\rm mf}^{-1}M(B_{\rm mf})$, with c and $B_{\rm mf}$ being the adjustable parameters. Those results for $\langle H_{s(p)-d}\rangle$ are also shown in Fig. 1. As one can see, the calculated curves provide a satisfactory representation of the experimental data.

The determined value of the mean molecular field $B_{\rm mf} = 4.5$ T shows why a linear approximation to the magnetization (at $B = 0$) fails in this particular case. The value of γ, determined within our model, corresponds to taking $N_0\beta = 0.75$ eV. This value obtained

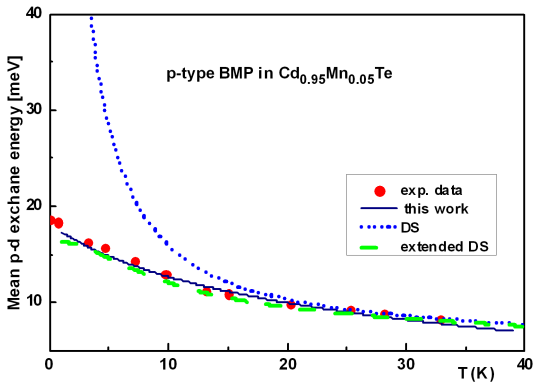

Fig. 1. Comparison with experimental data of Ref. [19] the temperature dependence of the effective p–d exchange energy calculated using the original DS approach [15] (dotted line), PA method of the present work (solid line), as well as including a simple extension of DS theory (dashed line).

for a bound-hole-impurity state compares well with the free-hole value 0.88 eV (cf. Refs. [19] and [21]). Finally, the determined value of $\varepsilon_{\rm p} = 13.76$ meV (at $T = 1$ K) is significantly larger than the corresponding value for donor case. Let us note however, that due to differences in the values of $N_0\alpha$ and $N_0\beta$, the enhancement factor of ≈ 16 appears. The additional enhancement is provided by a smaller value of the impurity-orbit diameter (also a factor of 4).

4. Conclusions

In this work we have developed a model of BMP in DMS based on the pair approximation. The results illustrated in Fig. 1 demonstrate the appropriateness of a simple extension of the BMP approach of Dietl and Spałek (DS) [15] to the situations when magnetization due to the spins can approach the saturated state. The value of the molecular field $B_{\rm mf}$ acting on the polaron depends on magnetic–ion concentration and on material parameters which can be estimated within our present approach. The detailed shape (apart from its size) of the impurity wave function seems not to be crucial in this case. This approach can also be applied to BMP located inside a quantum dot in a straightforward manner.

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