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Electron Correlations and Spatial Configuration of the Bipolaron

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Bipolaron energy is calculated for various distances between the centers of polarization wells of two polarons with accounting the electron correlations. A singlet bipolaron is stable at a rather high energy of ion binding $\eta \le \eta_m \approx 0.143$, $\eta = \varepsilon_\infty/\varepsilon_0$ (ε_∞ and ε_0 are the high-frequency and static dielectric constants, respectively). The unique energy minimum corresponds to a one-center bipolaron (analog of a helium atom). The bipolaron binding energy constitutes up to 25.8 % of a double polaron energy at $\eta \to 0$. A triplet bipolaron (analog of ortho-helium) is energetically disadvantageous. The one-center configuration of a triplet bipolaron corresponds to a maximum on the distance dependence of the total energy $J_{\mathrm{Bp}}(R)$. The exchange interaction between polarons has antiferromagnetic character. A prediction is made about a possibility of Wigner crystallization of a polaron gas, which occurs with antiferromagnetic ordering in the polaron system.

1. Introduction

The bipolaron problem has revived again due to the discovery of high-temperature superconductivity (HTSC). Unfortunate numerical errors made in earlier papers devoted to bipolarons (see, for example, review [1]) were corrected in [2]. The energy minimum obtained in [2] for a one-center bipolaron (or Pekar bipolaron [3]) was much lower than a corresponding minimum originally found in [4] for a two-center bipolaron. In terms of a spatial configuration, a one-center is analogous to a helium atom, while a two-center bipolaron is an analog of a hydrogen molecule. Subsequently the results of [2] were repeatedly reproduced for both Slater and Gaussian functions [1, 5–7].

The deciding factor in choosing a bipolaron probe wave function (WF) in variational calculations was its nonmultiplicative form. For a two-center bipolaron the probe WF was chosen [4] as a symmetrized product of one-electron WFs. For a one-center bipolaron correlation effects were taken into account by introducing a term depending on the difference in the electron coordinates [2, 5, 6] (in what follows this particular type of correlation effects will be referred to as electron correlations).

However, until the present time no calculations have been performed to take into account both effects at the same time. Therefore the question, which of the two bipolaron configurations is energetically more advantageous, still remains open. Thus, the works [8, 9] continue to explore a two-center bipolaron (despite the fact that the authors of [2] obtained a deeper minimum for a one-center bipolaron than in [8, 9]). A quantitative estimate of the contribution of electron correlations into the energy of two-electron systems of the type of large-radius paired centers in polar crystals (F_2-center

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or “a hydrogen molecule placed in a phonon field”) is yet to be obtained, too. To solve the problems of this sort one should consider both the permutation symmetry of the WF and the electron correlation at the same time.

The energy of a one-center bipolaron can be calculated with the use of wave functions of Pekar type [3]

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = N(1 + \gamma r_{12})\,(1 + \alpha r_1)\,(1 + \alpha r_2)\exp\left(-\alpha(r_1 + r_2)\right), \tag{1}$$

where N is a normalization multiplier, $\mathbf{r}_1, \mathbf{r}_2$ are the coordinates of the first and the second electrons, respectively, $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$ is the distance between the first and the second electron, $r_1 = |\mathbf{r}_1|$, $r_2 = |\mathbf{r}_2|$, γ and α are variational parameters.

The bipolaron energy is calculated analytically and the relevant functional is subsequently varied over the parameter γ (α is uniquely determined by γ). The deciding factor in the formation of a bound two-electron state is the availability of a correlation term proportional to the difference in the electron coordinates γr_{12}.

Calculations of a two-center bipolaron are concerned with the consideration of the permutation symmetry of the electron system. The wave function of the two-electron system is chosen in the symmetrized form

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = \Phi(\mathbf{r}_1, \mathbf{r}_2) + \Phi(\mathbf{r}_2, \mathbf{r}_1). \tag{2}$$

In early papers devoted to the calculation of bipolaron states, $\Phi(\mathbf{r}_1, \mathbf{r}_2)$ was chosen as a product of hydrogen-like WFs centered at various points [4]. In the recent paper [8], which deals with the calculation of a two-center bipolaron, $\Phi(\mathbf{r}_1, \mathbf{r}_2)$ is chosen as a product of Pekar polaron functions $(1 + \alpha r_{a1})\exp(-\alpha r_{a1})\,(1 + \alpha r_{b2})\exp(-\alpha r_{b2})$ (where use is made of the notation traditional for the two-center coordinate system: $\mathbf{r}_{a1}(\mathbf{r}_{a2})$ is the radius-vector of the first (second) electron reckoned from the center a, $\mathbf{r}_{b1}(\mathbf{r}_{b2})$ is the same for the center b). Both in [4] and in [8] the energy minima were much higher than a corresponding minimum obtained in [2] with the use of the wave function (1). The essence of variational calculations suggests, that preference should be given to the WF, which gives a deeper energy minimum. However final conclusions, which of two bipolarons is more energetically advantageous can be made only in the case, when the electron correlations in the wave function (2) are taken into account. This means the choice

$$\Phi(\mathbf{r}_1, \mathbf{r}_2) = (1 + \alpha r_{a1})\,(1 + \alpha r_{b2})\,(1 + \gamma r_{12})\exp\left(-\alpha(r_{a1} + r_{b2})\right). \tag{3}$$

However, though the wave function (3) has rather a simple form, one runs into severe obstacles when trying to find analytically the two-center integrals. Therefore the problem of a two-center bipolaron cannot be solved in the way as it was done for a one-center bipolaron. But if the WF is chosen in the form of a combination of Gaussian orbitals, analytical calculations become straight forward and the problem is reduced to a variation of a multiparametric functional.

Recently the interest in the polaron and bipolaron subject field has surfaced again due to an expansion of this field of investigations to anisotropic crystals, low dimensional structures and systems with quantum wells [7, 10–13]. The problem of the choice of a bipolaron spatial configuration and consideration of electron correlations is actual for such systems, too.

The aim of this work was to calculate the bipolaron energy depending on the distance between the centers of polarization wells taking into account both electron correlations and the permutation symmetry of the electron system.

2. Main Formulae and Relations

Let us choose the Hamiltonian of a system consisting of two electrons and a phonon field in the form

$$H = \hbar\omega \sum_{\mathbf{k}} a^{+}_{\mathbf{k}} a_{\mathbf{k}} + \sum_{\mathbf{k}} V_{\mathbf{k}} (a_{\mathbf{k}} - a^{+}_{-\mathbf{k}}) \left(\exp\,(i\mathbf{k}\mathbf{r}_1) + \exp\,(i\mathbf{k}\mathbf{r}_2)\right) - \frac{\hbar^2}{2m^*}\,\Delta_1 - \frac{\hbar^2}{2m^*}\,\Delta_2 + \frac{e^2}{\varepsilon_\infty |\mathbf{r}_1 - \mathbf{r}_2|}\,, \tag{4}$$

where $V_{\mathbf{k}} = -i\,\dfrac{e}{k}\sqrt{\dfrac{2\pi\hbar\omega}{V\tilde{\varepsilon}}}$, $\dfrac{1}{\tilde{\varepsilon}} = \dfrac{1}{\varepsilon_\infty} - \dfrac{1}{\varepsilon_0}$, V is the crystal volume, ω is the frequency of optical phonons, $\mathbf{k}$ is the wave vector of the phonons, $a^{+}_{\mathbf{k}}$ and $a_{\mathbf{k}}$ are the operators of creation and annihilation of phonons with the wave vector $\mathbf{k}$, $\mathbf{r}_1$, and $\mathbf{r}_2$ are electron coordinates.

The first term in (4) corresponds to the Hamiltonian of the field of optical phonons, the second one represents the Hamiltonian of the electron–phonon interaction in a two-electron system written in the Fröhlich form, the third and the fourth terms stand for the kinetic energy of electrons, the last term describes the Coulomb repulsion of electrons.

Canonical transformation of the Hamiltonian (4) $\exp\,(S_{\mathrm{a}})\,H\exp\,(-S_{\mathrm{a}})$ with $S_{\mathrm{a}} = \sum_{\mathbf{k}} C_{\mathbf{k}} (a^{+}_{\mathbf{k}} - a_{\mathbf{k}})$ and variation over parameters $C_{\mathbf{k}}$ (shift transformation) yields the following expression for the functional of the bipolaron ground: state

$$\bar{H} = \bar{V}_{\mathrm{ee}} + \bar{T} + \sum_{\mathbf{k}} \frac{U^2_{\mathbf{k}}}{-\hbar\omega}\,, \tag{5}$$

$$\bar{V}_{\mathrm{ee}} = \left\langle \Psi(\mathbf{r}_1, \mathbf{r}_2) \right| \frac{e^2}{\varepsilon_\infty r_{12}} \left| \Psi(\mathbf{r}_1, \mathbf{r}_2) \right\rangle, \tag{6}$$

$$\bar{T} = -\frac{\hbar^2}{2m^*} \left\langle \Psi(\mathbf{r}_1, \mathbf{r}_2) \right|\, \Delta_1 + \Delta_2 \left| \Psi(\mathbf{r}_1, \mathbf{r}_2) \right\rangle, \tag{7}$$

$$U_{\mathbf{k}} = V_{\mathbf{k}} \left\langle \Psi(\mathbf{r}_1, \mathbf{r}_2) \right| \exp\,(i\mathbf{k}\mathbf{r}_1) + \exp\,(i\mathbf{k}\mathbf{r}_2) \left| \Psi(\mathbf{r}_1, \mathbf{r}_2) \right\rangle. \tag{8}$$

In what follows we will use effective atomic units, i.e. the unit of energy will be $e^4 m^*/\hbar^2 \varepsilon^2_\infty$ and the unit of length will be effective Bohr's radius $a^*_0 = \hbar^2 \varepsilon_\infty / m^* e^2$.

Summing the functional (5) over the wave vector and integrating it we express it in the form traditional for bipolaron problems

$$J_{\mathrm{Bp}} = \bar{T} + \bar{V}_{\mathrm{ee}} + \bar{V}_{\mathrm{ef}}\,, \tag{9}$$

$$\bar{V}_{\mathrm{ef}} = -\frac{2e^2}{\tilde{\varepsilon}} \int \frac{|\Psi(\mathbf{r}_1, \mathbf{r}_2)|^2\, |\Psi(\mathbf{r}_3, \mathbf{r}_4)|^2}{r_{13}}\, \mathrm{d}\tau_{12}\, \mathrm{d}\tau_{34}\,. \tag{10}$$

Note, that in this formulation of the problem we do not use the concept of adiabatic approximation anywhere. We perform variational calculations of the initial Hamiltonian (4), which after canonical transformation changes to functional (5). A transformation of this sort leads to the results of the strong-coupling limit and gives the first term in the expansion of the polaron (bipolaron) energy in terms of $1/\alpha$ (where α is the Fröhlich binding constant, $\alpha \gg 1$) proportional to α^2,

$$\alpha = \frac{e^2}{\hbar \tilde{\varepsilon}} \sqrt{\frac{m^*}{2\hbar\omega}} \,. \tag{11}$$

The probe WF will be chosen as a linear combination of Gaussian orbitals

$$\Psi_S(\mathbf{r}_1, \mathbf{r}_2) = \Phi(\mathbf{r}_1, \mathbf{r}_2) + (-1)^S \, \Phi(\mathbf{r}_2, \mathbf{r}_1) \,, \tag{12}$$

where $S = 0$ for the singlet state (symmetric with respect to the operation of permutation of electron coordinates) and $S = 1$ for the triplet (antisymmetric) bipolaron state.

In turn

$$\Phi(\mathbf{r}_1, \mathbf{r}_2) = \sum_{i=1}^{n} C_i \exp\left(-a_{1i} r_{\mathrm{a}1}^2 - 2a_{2i}(\mathbf{r}_1 \cdot \mathbf{r}_2) - a_{3i} r_{\mathrm{b}2}^2\right), \tag{13}$$

where $\mathbf{r}_1 (\mathbf{r}_2)$ is the radius-vector of the first (second) electron reckoned from the coordinate origin placed halfway between the points a and b. The axis $0z$ is directed from the center a to the center b. The distance between the points a and b is equal to R. The quantities C_i, a_{1i}, a_{2i}, a_{3i} are variational parameters. The electron correlations are taking into account by the term $2a_{2i}(\mathbf{r}_1 \cdot \mathbf{r}_2)$ in (13), $(\mathbf{r}_1 \cdot \mathbf{r}_2)$ is the scalar product of vectors $\mathbf{r}_1$ and $\mathbf{r}_2$.

The polaron WF is chosen in the form

$$\Psi_{\mathrm{p}}(\mathbf{r}) = \sum_{i=1}^{n} c_i \exp\left(-\alpha_i r^2\right), \tag{14}$$

where c_i, α_i are variational parameters.

In going to a one-center bipolaron at $a_{1i} = a_{3i}$ the WF (13) is equivalent (with an accuracy of the notation of variational parameters) to the WF used in [5] for a one-center bipolaron and in the limit case of the absence of anisotropy it coincides with the wave function suggested in [6] for the calculation of the energy of a one-center bipolaron in anisotropic crystals.

3. Calculation Results

3.1 Singlet bipolaron

Quantitative analysis of correlation effects can conveniently be carried out with the use of the wave function (13) with five exponents.

Figure 1 shows the dependencies of the energy of the ground (singlet) state on the distance between the centers of polarization wells of two polarons at $\eta = 0$ (curve 2). It is seen, that as the distance between the polarons increases, the functional of the bipolaron system breaks down into functionals corresponding to the two polarons, the bipolaron energy tends to doubled polaron energy calculated in this approximation (-0.1085128), and the importance of electron correlations decreases. Figure 1 also displays the dependencies of corresponding energies for the system without regard for electron correlations at $\eta = 0$ (curve 1). In going to the multiplicative form at $R = 0$ (curve 1 is calculated for $a_{1i} = a_{3i}$, $a_{2i} = 0$ and corresponds to the symmetrized product of the polaron WF), the bipolaron functional breaks down into a doubled polaron functional, and the unique minimum at $R \neq 0$ corresponds to a two-center bipolaron. Tak-

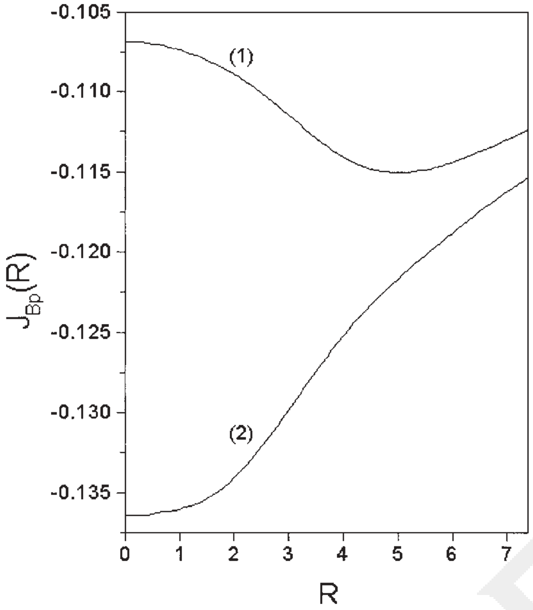

Fig. 1. Distance dependence of the bipolaron energy: curve 1 – without regard for the correlation ($a_{1i} = a_{3i}$, $a_{2i} = 0$, $n = 5$), curve 2 – the most general function (variational parameters a_{1i}, a_{3i}, a_{2i}, $n = 5$)

ing into account electron correlations result in a unique minimum at $R = 0$, which corresponds to a one-center bipolaron (curve 2 calculated for $a_{1i} \neq a_{3i}$, $a_{2i} \neq 0$), so the two-center bipolaron obtained without regard for electron correlations is associated with a less appropriate choice of the WF.

As the number of exponents in expression (13) increases, the qualitative picture shown in Fig. 1 does not change. The unique energy minimum corresponds to the one-center configuration as before.

Figure 2 shows the dependencies of various contributions into the bipolaron energy (kinetic energy $T(R)$, energy of electron–electron interaction $V_q(R)$, phonon contibution into the total energy $V_f(R)$) for five terms of the wave function (13).

So, a two-center bipolaron is energetically disadvantageous in an isotropic crystal, and the energy minimum arising at $R > 0$ [4, 8, 9] is associated with an inappropriate choice of the WF without electron correlations.

In many papers relative values of the bipolaron binding energy are given (in the units of polaron energy

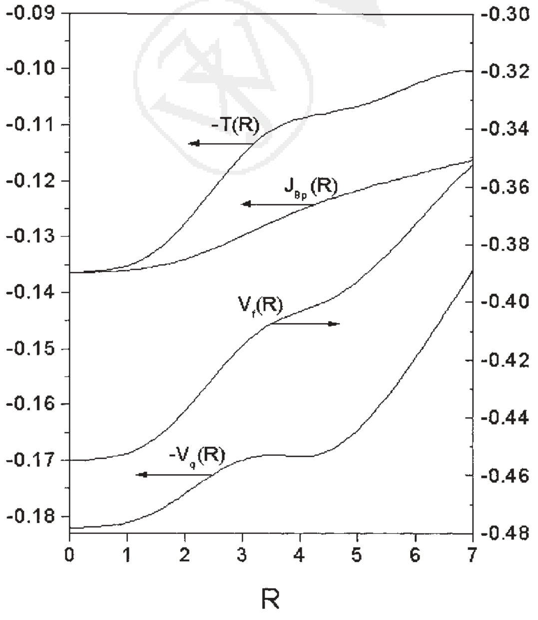

Fig. 2. Dependence of various contributions into the bipolaron energy on the distance between the centers of polarizations wells. $T(R)$ is the bipolaron kinetic energy, $V_q(R)$ is the energy of interelectron interaction, $V_f(R)$ is the phonon contribution into the bipolaron energy ($T(R) > 0$, $V_q(R) > 0$, $V_f(R) < 0$). $J_{Bp}(R)$ is the total bipolaron energy. The calculations are performed for variational parameters a_{1i}, a_{3i}, a_{2i}, $n = 5$

calculated in the same approximation) which leads to a misunderstanding in comparing the values of energy minima obtained by different authors. To remedy this we give absolute values of the energy found with the use of various WFs.

The Pekar function (1) leads at $\eta = 0$ to the ground state energy $J_{\rm Bp} = -0.1346292$. The polaron binding energy is given by

$$\Delta E = J_{\rm Bp} - 2J_{\rm p}\,. \tag{15}$$

Or, in relative units $\Delta E/2J_{\rm p} \approx 0.241$, where $J_{\rm p} = -0.0542564$ is the exact value of the polaron energy obtained numerically in the limit of strong binding in [14]. This value of $J_{\rm p}$ with all the significant digits is found with the use of a polaron WF in the form (14) for $n = 5$. The region of existence of the bipolaron for a Pekar WF is determined by $\eta \le \eta_{\rm m} = 0.132$.

In [2] absolute values of the ground state energy are not given, and slightly overestimated results for the binding energy and the region of existence of the bipolaron ($\Delta E/2J_{\rm p} \approx 0.25$, $\eta_{\rm m} \approx 0.14$) were associated with the fact that the bipolaron energy was calculated with respect to a slightly overestimated value of the polaron energy obtained for the WF chosen in the form $(1 + \alpha r)\exp(-\alpha r)$.

The best value of the energy of the bipolaron ground state obtained by us at $\eta = 0$ ($n = 11$) was

$$J_{\rm Bp} = -0.136512\,, \tag{16}$$

or in dimensionless units $\Delta E/2J_{\rm p} \approx 0.258$, $\eta_{\rm m} \approx 0.143$.

In the strong-coupling regime the present approach provides a larger binding energy and a broader region of the bipolaron stability than those obtained in [2, 4, 5, 8, 15].

Figure 3 shows the dependencies $\Delta E(R)$ (singlet state) for different η.

The results obtained will be generalized to crystals with intermediate electron–phonon coupling in a special paper. Notice only, that the conclusion about the energetic disadvantage of a two-center bipolaron remains unchanged [16]. A generalization of the results obtained by the strong coupling method to the range of intermediate values of the electron–phonon interaction parameter α with the use of the formula

$$J_i = 2J_{\rm Bp}\alpha^2\hbar\omega\,, \tag{17}$$

(where $J_{\rm Bp}$ corresponds to the bipolaron energy in the strong coupling limit), naturally gives underestimated values for the bipolaron energy and the critical parameter $\eta_{\rm m}$ as com-

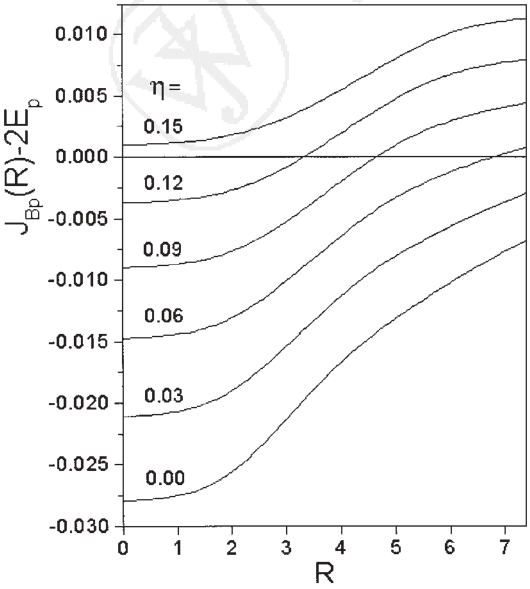

Fig. 3. The bipolaron binding energy as a function of distance between the centers of polarization wells for various parameters η

pared to the results obtained by the all-coupling methods [15, 17]. Thus, the lowest values of the bipolaron energy for $\alpha < 9$ are obtained in [17], where at $\alpha = 7$, J_{Bp} is calculated to be $-16.28\hbar\omega$, while formula (17) yields $-13.38\hbar\omega$.

Notice that the variation parameter **a**, which is treated in [17] as the mean distance at which electrons fluctuate, is analogous to the variation parameter R used in this paper. Our results correlate well with the fact, that the only energy minimum of a bipolaron corresponds to $\mathbf{a} = 0$ [17].

In the range $9 < \alpha < 25$, the lowest value of the bipolaron functional corresponds to the results of [15] and for $25 < \alpha$ the upper bound of the bipolaron energy is determined by the formula (17).

3.2 Exchange interaction between polarons

We have calculated the energy of a triplet term of bipolaron (one-center configuration of triplet bipolaron is analog of ortho-helium). As the distance between the centers of polarization wells increases, the energy of the triplet term of a bipolaron (WF (12) at $S = 1$) monotonically decreases in complete analogy with the $^3\Sigma_{\mathrm{u}}$-term of a hydrogen molecule. At $R = 0$ the distance dependence of the energy exhibits rather a sharp maximum ($J_{\mathrm{or}} = -0.076072$ at $\eta = 0$), which suggests instability of a triplet 2^3S state which can be arise, for example, under nonequilibrium conditions during an exchange scattering at bipolarons of zone electrons. In this case bipolarons break down into isolated polarons.

Note also that the use of the WFs (12), (13) for the calculation of the energy of pair- and ortho-helium has enabled us to get full agreement with experimental results, which suggests a considerable flexibility of the WFs used.

Noteworthy is the occurrence of antiferromagnetic (AF) exchange interaction between polarons. At rather large distances the WF of a polaron can be presented as a symmetrized (singlet state) or antisymmetrized (triplet state) product of polaron WFs. Then the interaction energy of two polarons has the form (with an accuracy of the terms quadratic in overlapping integral K)

$$E_{\mathrm{int}} = E_1 - J_{\mathrm{ex}} S_1 S_2 \,, \tag{18}$$

where S_1 and S_2 are spins of the first and the second electron,

$$E_1 = \frac{1}{\varepsilon_0} \int \frac{a(1)^2 b(2)^2}{r_{12}} \, \mathrm{d}\tau_{12} \,, \tag{19}$$

$$J_{\mathrm{ex}} = \frac{1}{\varepsilon_\infty} K_1 - \frac{4}{\tilde{\varepsilon}} K_2 K \,, \tag{20}$$

$$K_1 = \int \frac{a(1)\, b(1)\, a(2)\, b(2)}{r_{12}} \, \mathrm{d}\tau_{12} \,, \qquad K_2 = \int \frac{a(1)\, b(1)\, b(2)^2}{r_{12}} \, \mathrm{d}\tau_{12} \,,$$

$$K = \int a(1)\, b(1) \, \mathrm{d}\tau_1 \,,$$

where we use the notation traditional for two-center coordinate systems: $a(1)$, $b(1)$ are the WFs of a polaron centered at points a and b respectively ($a(1) \equiv \Psi_{\mathrm{p}}(\mathbf{r}_{\mathrm{a}1})$, $b(1) \equiv \Psi_{\mathrm{p}}(\mathbf{r}_{\mathrm{b}1})$).

The first term in (20) corresponds to ferromagnetic Coulomb exchange, the second one describes antiferromagnetic (AF) interaction between polarons caused by phonon contributions.

Thus, at large distances (if $\varepsilon_0 \gg \varepsilon_\infty$, $J_{\text{ex}} \approx -3K^2/\varepsilon_\infty R$) polarons push off and the spin-depending part of the interaction (total exchange) has AF character. Therefore Wigner crystallization accompanied by AF ordering can occur in a polaron system.

It also follows that for a bipolaron state to be formed the potential barrier should be overcome.

AF interaction between polarons can be responsible for the decrease in the paramagnetic component in the magnetic susceptibility of a polaron gas as the polaron concentration increases even though bound bipolaron states are not formed.

Instability of triplet bipolaron with respect to its breaking down into individual polarons obtained by us in the framework of Fröhlich Hamiltonian describing the interaction of electrons with optical phonons does not exclude a possibility for the formation of triplet states in a two-electron system caused by the interaction of electrons with elementary excitations of different nature, for example, with spin waves. The formation of self-localized electron states in antiferromagnetics with low Neel temperature ("spin polaron" in an AF crystal) was considered in [18, 19]. The author of [20] proposed, that high-temperature superconductivity is associated just with spin polarons, which form a bound state by full analogy with a bipolaron.

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Editors notes

- The English had to be polished somewhat, please check.
- Is pair helium correct or should it be instead para helium (page 8, last line).
- Are there some further information for Ref. [3]. Is it a Patent?