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The regular perturbation theory on the stability of the strong-coupling bipolaron

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Abstract. The regular perturbation method for the strong-coupling bipolaron is developed systematically. The ground-state energy of a bipolaron is calculated by the variational method as a function of the Fröhlich coupling constants α and η , the ratio of the optic to the static dielectric constant. It is found that the bipolaron is stable in a broad region of η . The estimated upper bound of η turns out to be 0.165.

1. Introduction

The idea of the bipolaron appears to have been mooted in 1951 by Pekar [1] who at that time conjectured that the formation of the bipolaron was energetically unfavourable. This conclusion has since been proved wrong by several workers [2–12]. In 1961, Vinetskii and Gitterman [2] reconsidered the problem and were the first to claim that the formation of the bipolaron was possible only in a suitable lattice environment. Since then the study of the bipolaron has continued to remain the focus of attention from both academic interest and technological importance viewpoints. Anderson [13] first advocated the bipolaron concept in ‘negative- U centres’ to study the electronic properties of amorphous semiconductors. Lakkis *et al* [14] proposed an intersite bipolaron to explain some properties of Ti_4O_7 . Recently it has attracted much attention because some workers [15–18] have conceived the idea that the bipolaron may be a possible candidate for explaining high- T_c superconductivity. However, all these propositions primarily demand the feasibility of the bipolaronic complex.

In this paper we shall consider the question of the first-principles existence of the bipolaron in a material medium from an *ab-initio* calculation. In such a study, one needs in the simplified case a Hamiltonian of two electrons, in the low-lying conduction band of a polar crystal, interacting with the lattice modes. The only such available Hamiltonian appears to have been obtained from the usual polaron Hamiltonian by introducing a second electron and its interaction with its environment. Regarding the question of the stability of the Fröhlich bipolaron a general consensus, however, seems to be lacking; the majority of workers have maintained that a Fröhlich bipolaron can indeed exist in polar crystals, provided that certain conditions are satisfied by the material parameters. In the present paper we have studied the formation and stability of the Fröhlich bipolaron on the basis of this standard Hamiltonian in the strong-coupling limit using the well known variational technique in combination with the Rayleigh–Schrödinger perturbation theory (RSPT). We first make a canonical transformation through the well known Lee–Low–Pines (LLP) [19] technique [20] in order to reduce it to a suitable form for use later in this paper. The reduced Hamiltonian is then split into an unperturbed Hamiltonian H_0 having a definable

basis and the remainder H_{int} as a perturbation. In the following we carry out the Rayleigh–Schrödinger perturbation procedure up to the desired order and then apply the variational principle to obtain the upper bound to the ground-state energy of the Fröhlich bipolaron. It will be seen that the energy calculated in this way to the first order in H_{int} is essentially the Landau–Pekar (LP) variational result [7, 8]. However, the LP approximation does not take into account the remainder term which is the difference between the actual Hamiltonian and the effective Hamiltonian corresponding to the Pekar *ansatz*. We have taken care of this remainder term through the RSPT in the present approach to the bipolaron problem. The remainder term which is neglected in the LP approximation in the language of our paper can be treated as a small parameter governing the perturbation expansion. It is seen that in the second-order RSPT all the negative second-order corrections, however small they may be, added to the first-order result produce a better stability criterion than do the LP variational result and others [2–12]. Corrections have been limited to the second order in H_{int} for the purpose of the present paper.

2. Formulation

The standard Hamiltonian for two electrons interacting with the optical modes in a polar lattice is given by (in Fröhlich units)

$$\tilde{H} = \sum_{i=1}^2 p_i^2 + \sum_{\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} + \frac{\mu}{r_{12}} + \sum_{i=1}^2 \sum_{\mathbf{q}} (\xi_{\mathbf{q}r_i} b_{\mathbf{q}} + \text{HC}) \quad (1)$$

where

$$\xi_{\mathbf{q}r_i} = -i \left(\frac{4\pi\alpha}{V} \right)^{1/2} \frac{1}{q} \exp(i\mathbf{q} \cdot \mathbf{r}_i) \quad (1a)$$

$$\mu = 2\alpha/(1 - \eta) \quad \eta = \varepsilon_{\alpha}/\varepsilon. \quad (1b)$$

ε_{α} and ε are the high frequency and the static dielectric constants, respectively, V is the volume and α is the usual Fröhlich coupling constant. The Hamiltonian given by equation (1) can be written as

$$\tilde{H} = h_1 + h_2 + h_{12} + h_{12}^{(0)} \quad (2)$$

where

$$h_1 = \sum_{i=1}^2 p_i^2 \quad (2a)$$

$$h_2 = \sum_{\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} \quad (2b)$$

$$h_{12} = \sum_{i=1}^2 \sum_{\mathbf{q}} (\xi_{\mathbf{q}r_i} b_{\mathbf{q}} + \text{HC}) \quad (2c)$$

$$h_{12}^{(0)} = \mu/r_{12}. \quad (2d)$$

The Hamiltonian \tilde{H} is now reduced by a standard canonical transformation [20] for use later in the paper, which leads to

$$\tilde{H} \rightarrow H = \exp(-S)\tilde{H} \exp S \tag{3}$$

where

$$S = \sum_q [(\xi_{q_1} + \xi_{q_2})b_q + \text{HC}] \tag{3a}$$

and

$$\xi_{q_i} = \langle P | \xi_{q r_i} | P \rangle \quad i = 1, 2. \tag{3b}$$

The averaging function is $|P\rangle$ to be defined presently. The transformed Hamiltonian H is now given by

$$H = h_1^{(1)} + h_2 + h_{12}^{(1)} + h_{12}^{(0)} \tag{4}$$

where

$$h_1^{(1)} = - \sum_{i=1}^2 \nabla_{r_i}^2 + \sum_{i=1}^2 \sum_{j=1}^2 \sum_q \xi_{q_i} \xi_{q_j}^* - \sum_{i=1}^2 \sum_{j=1}^2 \sum_q (\xi_{q r_i} \xi_{q_j}^* + \text{HC}) \tag{4a}$$

$$h_{12}^{(1)} = \sum_{i=1}^2 \sum_q [(\xi_{q r_i} - \xi_{q_i})b_q + \text{HC}]. \tag{4b}$$

It turns out that the transformed Hamiltonian retains the structure of the original Hamiltonian ($\tilde{H} = h_1 + h_2 + h_{12} + h_{12}^{(0)}$) changing, however, the individual components. Thus h_1 becomes $h_1^{(1)}$ which acquires an extra potential term entirely due to the coupling of the two systems and which is furthermore explicitly attractive. This must then be at the cost of the interaction h_{12} which in the new form $h_{12}^{(1)}$ may be seen to be less effective than h_{12} . As a result we obtain an effective Hamiltonian $H_{\text{eff}} = h_1^{(1)} + h_2 + h_{12}^{(0)}$ which already contains much of the phonon-induced interaction and $h_{12}^{(1)}$ is the exact remainder term. This method [20] may be iterated to any desired order. Finally there still remains a residual interaction term which can now be treated by a suitable conventional method.

Our approach is essentially directed at repartitioning of the exact Hamiltonian H into an unperturbed Hamiltonian H_0 and a perturbation H_{int} such that H_0 admits a basis. This is arrived at in such a way that in the lowest order one obtains the LP variational result [7] and then the next higher-order contribution leads to a better stability criterion.

For a single polaron the LP approximation in its simplest form implies replacing the induced potential by an effective potential (which corresponds to the variational wavefunction) in the electronic equation. Such an effective wavefunction [21] method is known to have provided very good results for the strong-coupling polaron. Following this we repartition the Hamiltonian (4) as follows:

$$H = H_0 + H_{\text{int}} \tag{5}$$

where

$$H_0 = - \sum_{i=1}^2 \nabla_{r_i}^2 + \sum_q b_q^\dagger b_q + V_{\text{eff}}(r_1, r_2) \tag{5a}$$

and

$$H_{\text{int}} = H^{(1)} + H^{(2)} + H^{(3)} \quad (5b)$$

$$H^{(1)} = - \sum_{i=1}^2 \sum_{j=1}^2 \sum_q (\xi_{qr} \xi_{qj}^* + \text{HC}) - V_{\text{eff}}(\mathbf{r}_1, \mathbf{r}_2) + \frac{\mu}{r_{12}} \quad (5c)$$

$$H^{(2)} = \sum_{i=1}^2 \sum_{j=1}^2 \sum_q \xi_{qi} \xi_{qj}^* \quad (5d)$$

$$H^{(3)} = \sum_{i=1}^2 \sum_q [(\xi_{qr_i} - \xi_{q_i}) b_q + \text{HC}]. \quad (5e)$$

The motivation behind the partition of the Hamiltonian and the inclusion of an effective potential, as described above, is to find an appropriate basis for the perturbative calculation. With a particular choice of the effective potential the basis of H_0 then becomes

$$\psi_{kv}(\mathbf{r}_1, \mathbf{r}_2, \dots, v_q, \dots) = \varphi_k(\mathbf{r}_1, \mathbf{r}_2) \chi(\dots v_q \dots) \quad (6)$$

where $\varphi_k(\mathbf{r}_1, \mathbf{r}_2)$ is the k th eigenstate of $-\nabla_{\mathbf{r}_1}^2 - \nabla_{\mathbf{r}_2}^2 + V_{\text{eff}}(\mathbf{r}_1, \mathbf{r}_2)$ and $\chi(\dots v_q \dots)$ is the general eigenstate of $\sum_q b_q^\dagger b_q$. The ground state of H_0 is given by

$$|g, 0\rangle = \varphi_g(\mathbf{r}_1, \mathbf{r}_2) \chi(0) \quad b_q \chi(0) = 0. \quad (7)$$

We now choose the averaging function in equation (3b) to be $\varphi_g(\mathbf{r}_1, \mathbf{r}_2)$. Then a systematic RSPT in different order gives the following:

$$\begin{aligned} \text{zero order:} \quad E^{(0)} &= \langle g, 0 | H_0 | g, 0 \rangle \\ &= \langle g | p_1^2 + p_2^2 + V_{\text{eff}}(\mathbf{r}_1, \mathbf{r}_2) | g \rangle \end{aligned} \quad (8)$$

$$\begin{aligned} \text{first order:} \quad E^{(1)} &= \langle g, 0 | H_{\text{int}} | g, 0 \rangle \\ &= \langle g | H^{(1)} | g \rangle + H^{(2)} \end{aligned} \quad (9)$$

$$\begin{aligned} \text{second order:} \quad E^{(2)} &= \sum_k' \frac{|\langle g, 0 | H_{\text{int}} | k, v \rangle|^2}{E_{g,0} - E_{k,v}} \\ &= \sum_k' \frac{|\langle g, 0 | H^{(1)} + H^{(2)} + H^{(3)} | k, v \rangle|^2}{E_{g,0} - E_{k,v}} \\ &= \sum_k' \frac{|\langle g | H^{(1)} | k \rangle|^2}{E_{g,0} - E_{k,0}} + \sum_k' \frac{|\langle g, 0 | H^{(3)} | k, 1 \rangle|^2}{E_{g,0} - E_{k,1}} \end{aligned} \quad (10)$$

where the prime in the summation indicates that $|k\rangle \neq |g\rangle$ and $H^{(2)}$ does not enter into the final expression of $E^{(2)}$ because of the orthogonality of the ground-state and excited-state wavefunctions.

3. Results and discussion

For an explicit calculation of the perturbative result we choose the effective potential

$$V_{\text{eff}}(\mathbf{r}_1, \mathbf{r}_2) = \lambda^4 r_1^2 + \lambda^4 r_2^2 + 2\beta^4 \mathbf{r}_1 \cdot \mathbf{r}_2 \tag{11}$$

such that it accounts for the correlated motion the two electrons in the bipolaronic complex. Moreover this choice of the effective potential does not destroy the generality of the system in any way because this term is only added and subtracted from the actual Hamiltonian in order to repartition this Hamiltonian for a convenient perturbative calculation.

Now the perturbative calculation is easily accomplished by introducing the new variables

$$\zeta = (\mathbf{r} + \mathbf{r}_2)/\sqrt{2} \quad \gamma = (\mathbf{r}_1 - \mathbf{r}_2)/\sqrt{2}. \tag{12}$$

In the new representation

$$V_{\text{eff}}(\mathbf{r}_1, \mathbf{r}_2) = \lambda_1^4 \zeta^2 + \lambda_2^4 \gamma^2 \tag{13}$$

where

$$\lambda_1^4 = \lambda^4 + \beta^4 \quad \text{and} \quad \lambda_2^4 = \lambda^4 - \beta^4; \tag{14}$$

$$H_0 = (p_\zeta^2 + \lambda_1^4 \zeta^2) + (p_\gamma^2 + \lambda_2^4 \gamma^2) + \sum_q b_q^\dagger b_q. \tag{15}$$

The basis of H_0 is then given by

$$\varphi_{ij}(\zeta, \eta) = \phi_i(\zeta)\phi_j(\gamma) \tag{16}$$

where $\phi_i(\zeta)$ is the excited state of $p_\zeta^2 + \lambda_1^4 \zeta^2$ and $\phi_j(\gamma)$ is the j th excited state of $p_\gamma^2 + \lambda_2^4 \gamma^2$.

With the corresponding basis the perturbation result up to first order gives

$$\begin{aligned} E = E^{(0)} + E^{(1)} &= \frac{3(\lambda_1^2 + \lambda_2^2)}{2} + \mu\lambda_2 \left(\frac{2}{\pi}\right)^{1/2} - \lambda_1\lambda_2 \frac{4\sqrt{2}\alpha(2/\pi)^{1/2}}{(\lambda_1^2 + \lambda_2^2)^{1/2}} \\ &= \frac{3\lambda_1^2(1+t^2)}{2} + \mu t\lambda_1 \left(\frac{2}{\pi}\right)^{1/2} - 4\sqrt{2}\alpha\lambda_1 t \frac{(2\pi)^{1/2}}{(1+t^2)^{1/2}} \end{aligned} \tag{17}$$

where

$$t = \lambda_2/\lambda_1.$$

The variational principle demands that

$$\partial E/\partial\lambda_1 = 0 \quad \partial E/\partial t = 0 \tag{18}$$

$$\Rightarrow \lambda_1 = 2\alpha \frac{(2/\pi)^{1/2} t}{3(1+t^2)} \left(\frac{2\sqrt{2}}{(1+t^2)^{1/2}} - \frac{1}{1-\eta} \right) \tag{19}$$

and

$$\frac{1-t^2}{(1+t^2)^{1/2}} = \frac{1}{2\sqrt{2}(1-\eta)}. \tag{20}$$

Solving (19) and (20) for λ_1 and t we obtain the energy of the bipolaron as a function of α^2 and η . The result obtained here by canonical transformation followed by first-order RSPT is identical with what one would obtain by LP variational treatment. Now we need to examine the stability criterion for the formation of the bipolaron which is

$$E - 2E_P < 0$$

where $E_P (= -\alpha^2/3\pi)$ is the energy of a single polaron calculated in the same procedure. The above-mentioned condition thus leads to the desired criterion, namely that, for $\eta < \eta_c (= 0.08)$, the bipolaron is stable which compares well with the other values of η_c reported in the literature [6–8]. However, for $\beta = 0$ we obtain [9]

$$\lambda_1 = \lambda_2 = \frac{\alpha}{3} \left(\frac{2}{\pi} \right)^{1/2} \left(2 - \frac{1}{1-\eta} \right) \tag{21}$$

and

$$E - 2E_P = \frac{2\alpha^2}{3\pi} \left(\frac{1}{1-\eta} - 1 \right) \tag{22}$$

where $\eta = \varepsilon_\alpha/\varepsilon$ is always less than unity. It therefore follows that the energy of the bipolaron (for the $\beta = 0$ case) to the first order always lies above the energy of the two independent polarons. This is the Pekar [1] case and the bipolaron formation is not favoured to this order of approximation. This negative result of Pekar occurs because no correlation is taken into account between the electrons in the bipolaronic wavefunction. Thus we are inclined to include that the effect through a systematic perturbation theory with an appropriate choice of the effective potential which takes care of the appropriate interaction between the electrons in the bipolaronic complex and we see that our choice of the effective potential ($v_{\text{eff}}(r_1, r_2) = \lambda^4 r_1^2 + \lambda^4 r_2^2 + 2\beta r_1 \cdot r_2$) represents the formation of a bipolaron in the first order whereas in the Pekar case $v_{\text{eff}}(r_1, r_2) = \lambda^4 r_1^2 + \lambda^4 r_2^2$, which does not take into account any interaction between the electrons in the first order of the approximation and no formation of a bipolaron occurs in the Pekar approach. The crux of this contention is that the LP approximation in the language of the present paper, which includes only first-order effects, may not be regarded as the final criterion of stability or instability. Higher-order corrections may enhance the stability criterion which we shall presently see.

The second-order correction to the first-order result (equation (17)) is given by equation (10). Evaluating all the matrix elements we finally obtain

$$E^{(2)} = E_1^{(2)} + E_2^{(2)} \tag{23}$$

where

$$\begin{aligned} E_1^{(2)} = & -\frac{4\alpha^2}{\pi^2} \sum_N \sum_{N'} [1 - (-1)^{N+N'+1}] \frac{(\Gamma(N + N' + 1)/2)^2}{(N + N' + 1)N!N!} \frac{t^{2(N+1)}}{(N + N't^2)(1 + t^2)^{N+N'+1}} 16 \\ & - \frac{3(\lambda_1^2 + \lambda_2^2)}{8} - \frac{\mu^2}{4\pi^2} \sum_N [1 - (-1)^{N+1}] \frac{2^{N+1}(\Gamma(N + 1)/2)^2}{NN!} \\ & + \frac{16\mu\alpha}{\sqrt{2}\pi^2} \sum_N [1 - (-1)^{N+1}] \frac{2^{N+1}(\Gamma(N + 1)/2)^2}{NN!(1 + t^2)^{(N+1)/2}} + \frac{4\alpha\lambda t}{2\sqrt{\pi}(1 + t^2)^{3/2}} \\ & - \frac{(2/\pi)^{1/2}\mu\lambda_1 t}{4} \end{aligned} \tag{24}$$

and

$$E_2^{(2)} = -\frac{2\alpha}{\pi\lambda_1} \sum_N \sum_{N'} \frac{t^{2N+1} \Gamma(N + N' + 1/2)}{2^{(N+N'-1/2)} N! N'! (N + N' t^2) (1 + t^2)^{N+N'+1/2}} \times \left[1 + \sum_{P=12} (-1)^P \left(\frac{1}{N + N' t^2} \right)^P \frac{1}{2\lambda_1^{2P}} \right]. \tag{25}$$

Equation (23) together with (17) will give the bipolaron energy in the second order of approximation. Appropriate variations have been made in the strong-coupling limit in order to obtain the ground-state energy of the bipolaron in the second-order RSPT which may be found in the following form:

$$E_{BP} = E^{(0)} + E^{(1)} + E_1^{(2)} + C + O(1/\alpha^2) \tag{26}$$

where

$$C = -\frac{2\alpha}{\pi\lambda_1} \sum_N \sum_{N'} \frac{t^{2N+1} \Gamma(N + N' + 1/2)}{2^{N+N'-1/2} N! N'! (N + N' t^2) (1 + t^2)^{N+N'+1/2}}. \tag{27}$$

In the limit of the localized state solution, λ_1 and t in the second order of approximation are also given by equations (19) and (20). The contributions to the bipolaron energy from successive orders of approximation are shown in table 1. It is found that the second-order contribution, however small it may be, enhances the stability of the bipolaronic complex. We have calculated the binding energy of the bipolaron given by $B_{BP} = 2\bar{E}_P - E_{BP}$ where \bar{E}_P is the energy of a single polaron calculated in the framework of the same method in order to obtain the stability criterion for the formation of the bipolaron. In this order of approximation, \bar{E}_P turns out to be given by

$$\bar{E}_P = -0.1077\alpha^2 + 3 \ln 2. \tag{28}$$

Table 1. Successive contributions to the energy of the bipolaron obtained in the successive orders of approximation in our perturbative approach based on the RSPT and variational method: $E^{(0)}$, energy of the bipolaron in the zeroth-order perturbative calculation; $E^{(1)}$, first-order perturbative correction to the bipolaron energy; $E^{(2)}$, second-order perturbative correction to the bipolaron energy; E_{BP} , energy of the bipolaron in the second-order perturbative calculation.

η	$E^{(0)}$	$E^{(1)}$	$E^{(2)}$	E_{BP}
0.00	$0.2435\alpha^2$	$-0.4871\alpha^2$	$-0.0291\alpha^2$	$-0.2727\alpha^2$
0.05	$0.2238\alpha^2$	$-0.4476\alpha^2$	$-0.0322\alpha^2$	$-0.2560\alpha^2$
0.10	$0.2031\alpha^2$	$-0.4062\alpha^2$	$-0.0358\alpha^2$	$-0.2389\alpha^2$
0.15	$0.1815\alpha^2$	$-0.3631\alpha^2$	$-0.0411\alpha^2$	$-0.2227\alpha^2$

The stability criterion has been obtained by demanding that B_{BP} is positive and we find a value of $\eta = \eta_c = 0.165$, below which this desired stability condition is satisfied. For the $\beta = 0$ case this result becomes worse. η_c becomes of the order of 0.10. However, this result is quite impressive in the sense that we can overcome Pekar's [1] negative argument in this approach. Using the same Pekar wavefunction we can go beyond Pekar's result through a systematic perturbation procedure.

It is now worth noting that the required constraint on η is very liberal compared with the other results reported in the literature. Suprun and Moizhes [3] using Pekar-type products of one electron wavefunctions with built-in correlation factors obtained $\eta_c = 0.14$, which was much later obtained by Adamowskii [5] in the strong-coupling limit. Sil and co-workers [8], by introducing simple correlation factors, have recently obtained an n_c value of 0.132. The path integral calculation of Hiramoto and Toyozu [4] yielded an n_c value of 0.079, also obtained otherwise by Bassani *et al* [7]. Devreese and co-workers [6] in a recent work obtain an upper bound of $\eta = 0.131$ for a large value of α . Recently we [12] have obtained $\eta_c = 0.14$ in a variational calculation using the concept of a two-centre bipolaron [2]. The common feature of all these works is the estimate of η_c below which the bipolaron is stable. It is seen that the region of the bipolaron stability obtained by the present method is much broader than those of others [2–12]. Our approach is close to that of Pekar with additional interaction extracted from the remainder term. Correlation need not be introduced artificially in this approach, as it is built into the formalism. As for the energy of the bipolaron, although it has been shown that the strongly coupled bipolaron energy is proportional to α^2 , the complete numerical value has not been reported [5–9] so far. In table 2 we compare the binding energy of the bipolaron with the other result [7] available in the literature.

Table 2. Comparison of the binding energy η in the present calculation with the binding energy (BE) from Bassani *et al* [7]: BE1, binding energy of the bipolaron in the first-order perturbative calculation; BE2, binding energy of the bipolaron in the second-order perturbative calculation.

η	BE [7]	BE1	BE2
0.000	$0.0314\alpha^2$	$0.0314\alpha^2$	$0.0572\alpha^2$
0.035	$0.0177\alpha^2$	$0.0177\alpha^2$	$0.0456\alpha^2$
0.070	$0.0035\alpha^2$	$0.0035\alpha^2$	$0.0338\alpha^2$

In conclusion the present paper supplies theoretical support in favour of the formation of the optical bipolaron in a material medium. We have shown in this paper that, through the inclusion of interaction beyond the LP approximation, a condition for the optical bipolaron exists. Higher approximations are made through a regular perturbation procedure. First-order correction essentially gives the stability criterion which is otherwise obtained by the LP variational method. Second-order correction considerably improves that result. Higher-order corrections are only likely to strengthen these conclusions.

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