

## SOLUTION TO LEE-LOW-PINES EQUATIONS IN THE BIPOLARON THEORY

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We have found numerical solutions to Lee-Low-Pines bipolaron equations. According to these solutions the bipolaron energy exceeds the energy of two uncoupled polarons over the whole range of parameter variation. We suggest also the existence of metastable bipolaron states.

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### 1. Introduction

We witness an increased interest in the bipolaron problem, particularly in the context of high-temperature superconductivity [1-2]. Complete solution to this problem is still to be found. Exact solutions are presently found only in the strong coupling limit [3]. Thus, in the whole range of variation of electron-phonon coupling constant  $\alpha$  except for as  $\alpha \rightarrow \infty$ , only variational estimates of the bipolaron energy are available. Sometimes the variational approach

leads to an effective one-particle Schrödinger equation whose solutions may be of interest in the bipolaron theory. In this paper we study numerical solutions to this equation, found by using the Lee-Low-Pines (LLP) variational method.

### 2. LLP equations in the bipolaron theory

The Pekar-Fröhlich Hamiltonian for two electrons interacting with the phonon field is written as

$$H = -\frac{\hbar^2}{2m}\Delta_{r_1} - \frac{\hbar^2}{2m}\Delta_{r_2} + \sum_k \hbar\omega_k b_k^\dagger + b_k + U_c(|r_1 - r_2|) + \sum_k [b_k C_k (\exp(ikr_1) + \exp(ikr_2)) + \text{h.c.}], \quad (1)$$

where  $r_1$  and  $r_2$  are the electron coordinates,  $\omega_k$  are the phonon frequencies, and  $C_k$  are some constants.

Following the LLP method, Hamiltonian (1) can be canonically transformed [4, 5],

$$U_1 = \exp\left[i \frac{r_1 + r_2}{2\hbar} \sum_k \hbar k b_k^\dagger b_k\right], \quad (2)$$

$$\tilde{H} = U_1 H U_1^{-1}. \quad (3)$$

As a result  $\tilde{H}$  is expressed as

$$\begin{aligned} \tilde{H} = & -\frac{\hbar^2}{2m}\Delta_{r_1} - \frac{\hbar^2}{2m}\Delta_{r_2} + U_c(|r_1 - r_2|) + \sum_k b_k^\dagger b_k \left[ \hbar\omega_k + i \frac{\hbar^2 k}{2m} (\nabla_{r_1} + \nabla_{r_2}) \right] \\ & + \frac{1}{4m} \left( \sum_k \hbar k b_k^\dagger b_k \right)^2 + \sum_k \left[ b_k C_k \left( \exp\left[-\frac{ik(r_1 - r_2)}{2}\right] + \exp\left[\frac{ik(r_1 - r_2)}{2}\right] \right) + \text{h.c.} \right]. \end{aligned} \quad (4)$$

Applying to (4) a canonical transformation of the form

$$U_2 = \exp \left[ \sum_k (f_k^* b_k - b_k f_k) \right], \quad (5)$$

$$\tilde{H} = U_2 \tilde{H} U_2^{-1}, \quad (6)$$

where  $f_k$  are the LLP coefficients

$$f_k = -\frac{2C_k^* \rho_k^*}{\hbar\omega_k + \hbar^2 k^2/4m}, \quad (7)$$

$$\rho_k = \frac{1}{2} \int dV_1 dV_2 \left\{ \exp \left[ -\frac{ik(r_1 - r_2)}{2} \right] + \exp \left[ \frac{ik(r_1 - r_2)}{2} \right] \right\} |\Phi(r_1, r_2)|^2$$

we write for the wave function  $\Phi(r_1, r_2)$  the Schrödinger equation

$$\left[ -\frac{\hbar^2}{2m} \Delta_{r_1} - \frac{\hbar^2}{2m} \Delta_{r_2} + U(r_1, r_2) \right] \Phi(r_1, r_2) = W \Phi(r_1, r_2), \quad (8)$$

with the effective potential

$$U(r_1, r_2) = U_c(|r_1 - r_2|) - 2 \sum_k \frac{|C_k|^2}{\hbar\omega_k + \hbar^2 k^2/4m} \left\{ \rho_k^* \left( \exp \left[ -\frac{ik(r_1 - r_2)}{2} \right] + \exp \left[ \frac{ik(r_1 - r_2)}{2} \right] \right) + \text{h.c.} \right\} \quad (9)$$

Let us pass in (8) and (9) to coordinates of the center of mass and expressing the wave function  $\Phi$  as

$$\Phi(r_1, r_2) = \frac{1}{\sqrt{V}} \Psi(r_1 - r_2), \quad (10)$$

in the case of ionic crystals

$$C_k = ek^{-1} \sqrt{\frac{2\pi\omega_k}{\epsilon V}}, \quad \omega_k = \omega_0, \\ U_c(|r_1 - r_2|) = \frac{e^2}{\epsilon_\infty |r_1 - r_2|}. \quad (11)$$

Then Eq. (8) changes to

$$\left[ -\frac{\hbar^2}{m} \Delta_r + U(r) \right] \Psi(r) = W \Psi(r), \quad (12)$$

$$U(r) = -\frac{8e^2}{\epsilon} \int dV' \frac{|\Psi(r')|^2}{|r - r'|} \\ \times \left[ 1 - \exp \left( -\frac{\lambda|r - r'|}{2} \right) \right] + \frac{e^2}{\epsilon_\infty r}, \quad (13)$$

$$\lambda = \sqrt{\frac{4m\omega_0}{\hbar}}. \quad (14)$$

Thus, the LLP method leads to an effective one-particle Schrödinger equation, numerical solutions to which are to be studied below.

### 3. Solutions to the LLP bipolaron equation

Schrödinger equations (12) and (13) can be presented as a set of differential equations

$$\frac{\hbar^2}{\mu} \Delta \Psi - \frac{e^2 \Psi}{\epsilon_\infty r} + e(\Pi_1 - \Pi_2) \Psi - |W| \Psi = 0, \\ \Delta \Pi_1 + \frac{32\pi e}{\epsilon} \Psi^2 = 0, \\ \Delta \Pi_2 - \frac{\lambda^2}{4} \Pi_2 + \frac{32\pi e}{\epsilon} \Psi^2 = 0. \quad (15)$$

Written in dimensionless variables

$$\Psi(r) = \frac{|W|}{e\hbar} \sqrt{\frac{m\epsilon}{32\pi}} \frac{\xi(x)}{x}, \quad \Pi_1(r) = \frac{|W|}{e} \frac{\eta_1(x)}{x}, \\ \Pi_2(r) = \frac{|W|}{e} \frac{\eta_2(x)}{x}, \quad r = \frac{\hbar}{\sqrt{m|W|}} x, \quad (16)$$

Table 1.

$\alpha$	$T$	$N$	$Q$	$R$	$\alpha$	$T$	$N$	$Q$	$R$
2.0	1.0973	7.43234	2.2075	29.22416	0.37	1.0485	8.40033	2.1893	37.07256
1.9	1.0969	7.43800	2.2074	29.26748	0.36	1.0469	8.44036	2.1887	37.41627
1.8	1.0965	7.44459	2.2072	29.31794	0.35	1.0453	8.48273	2.1880	37.78171
1.7	1.0960	7.45232	2.2071	29.37722	0.34	1.0435	8.52762	2.1873	38.17081
1.6	1.0954	7.46147	2.2069	29.44746	0.33	1.0417	8.57524	2.1865	38.58571
1.5	1.0948	7.47240	2.2066	29.53151	0.32	1.0398	8.62583	2.1857	39.02882
1.4	1.0940	7.48561	2.2064	29.63319	0.31	1.0378	8.67965	2.1849	39.50286
1.3	1.0930	7.50177	2.2060	29.75773	0.3	1.0358	8.73697	2.1840	40.01088
1.2	1.0918	7.52179	2.2056	29.91241	0.29	1.0336	8.79813	2.1831	40.55635
1.1	1.0903	7.54698	2.2051	30.10762	0.28	1.0314	8.86349	2.1821	41.14323
1.0	1.0884	7.57926	2.2044	30.35855	0.27	1.0291	8.93346	2.1811	37.77605
0.9	1.0859	7.62146	2.2035	30.68812	0.26	1.0267	9.00850	2.1800	42.46000
0.8	1.0828	7.67797	2.2024	31.13208	0.25	1.0241	9.08915	2.1789	43.20109
0.7	1.0785	7.75584	2.2009	31.74870	0.24	1.0215	9.17601	2.1776	44.00630
0.6	1.0727	7.86689	2.1987	32.63795	0.23	1.0187	9.26980	2.1764	44.88379
0.59	1.0720	7.88051	2.1985	32.74784	0.22	1.0158	9.37131	2.1750	45.84314
0.58	1.0713	7.89469	2.1982	32.86240	0.21	1.0128	9.48150	2.1736	46.89574
0.57	1.0706	7.90946	2.1979	32.98190	0.2	1.0096	9.60148	2.1720	48.05516
0.56	1.0698	7.92484	2.1977	33.10662	0.19	1.0063	9.73256	2.1704	49.33776
0.55	1.0690	7.94088	2.1974	33.23688	0.18	1.0028	9.87631	2.1686	50.76342
0.54	1.0682	7.95760	2.1970	33.37299	0.17	0.99923	10.0346	2.1668	52.35657
0.53	1.0674	7.97506	2.1967	33.51531	0.16	0.99542	10.2098	2.1647	54.14760
0.52	1.0665	7.99328	2.1964	33.66422	0.15	0.99143	10.4046	2.1626	56.17474
0.51	1.0656	8.01232	2.1960	33.82013	0.14	0.98724	10.6227	2.1603	58.48686
0.5	1.0646	8.03222	2.1957	33.98349	0.13	0.98285	10.8684	2.1578	61.14748
0.49	1.0636	8.05304	2.1953	34.15477	0.12	0.97824	11.1475	2.1550	64.24069
0.48	1.0626	8.07484	2.1949	34.33450	0.11	0.97339	11.4676	2.1521	67.88041
0.47	1.0616	8.09766	2.1945	34.52323	0.1	0.96829	11.8387	2.1488	72.22491
0.46	1.0605	8.12159	2.1941	34.72158	0.09	0.96293	12.2748	2.1453	77.50103
0.45	1.0593	8.14669	2.1936	34.93021	0.08	0.95727	12.7957	2.1413	84.04624
0.44	1.0582	8.17303	2.1932	35.14985	0.07	0.95130	13.4312	2.1369	92.38610
0.43	1.0569	8.20070	2.1927	35.38129	0.06	0.94498	14.2273	2.1320	103.3881
0.42	1.0557	8.22979	2.1922	35.62539	0.05	0.93830	15.2613	2.1263	118.5978
0.41	1.0544	8.26040	2.1917	35.88310	0.04	0.93123	16.6747	2.1197	141.0695
0.4	1.0530	8.29264	2.1911	36.15547	0.03	0.92377	18.7635	2.1119	177.8368
0.39	1.0516	8.32662	2.1906	36.44363	0.02	0.91606	22.2953	2.1020	249.6668
0.38	1.0501	8.36247	2.1900	36.74887	0.01	0.90903	30.3263	2.0881	458.2225

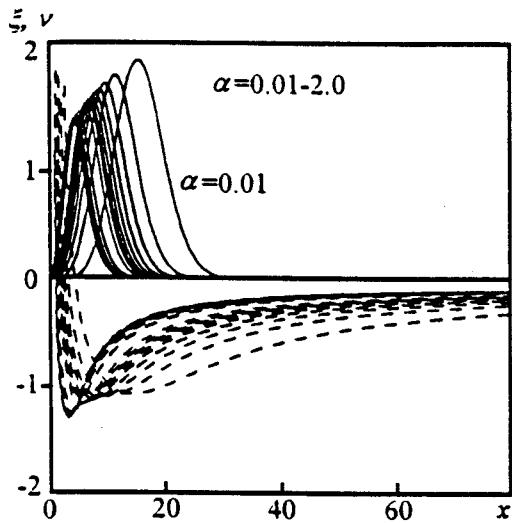


Figure 1. Solutions of (17) for various parameters  $\alpha$  and potential energy of the system: solid lines correspond to  $\xi$ , dashed lines correspond  $\nu$ ;  $\nu = N/4x - [\eta_1(x) - \eta_2(x)]x^{-1}$ .

equation (15) in the spherically-symmetric case takes on the form

$$\begin{aligned} \xi''(x) - \xi(x) - \frac{N}{4x}\xi(x) + \frac{1}{x}(\eta_1(x) - \eta_2(x))\xi(x) &= 0, \\ \eta_1''(x) + \frac{\xi^2(x)}{x} &= 0, \\ \eta_2''(x) - \alpha^2 \frac{N^2}{4} \eta_2(x) + \frac{\xi^2(x)}{x} &= 0. \end{aligned} \quad (17)$$

This set is subject to the boundary conditions

$$\xi(0) = \xi(\infty) = \eta_1(0) = \eta_1'(\infty) = \eta_2(0) = \eta_2(\infty) = 0. \quad (18)$$

Using the notation

$$N = \frac{\Gamma}{2} \frac{\tilde{\epsilon}}{\epsilon_\infty}, \quad \alpha = \frac{\lambda}{64} \frac{\hbar^2 \epsilon_\infty}{e^2 m}, \quad \Gamma = \int_0^\infty \xi^2 dx, \quad (19)$$

energy  $W$ , total energy  $F$ , and bipolaron radius  $\langle r \rangle$  are written as

$$|W| = 16 \frac{e^4 m}{\hbar^2 \epsilon_\infty^2} \frac{1}{N^2}, \quad (20)$$

$$F = \frac{|W|}{4N} \frac{\tilde{\epsilon}}{\epsilon_\infty} \left\{ T - 2N \frac{\epsilon_\infty}{\tilde{\epsilon}} + \frac{1}{4} N Q \right\}, \quad (21)$$

$$\langle r \rangle = \int r \Psi^2 dV = \frac{\hbar^2 \tilde{\epsilon}}{2em} R. \quad (22)$$

Figure 1 shows solutions to system (17) for various parameters  $\alpha$ . Table 1 lists quantities  $N$ ,  $Q$ , and  $R$ , where

$$T = \int_0^\infty \xi'^2 dx, \quad Q = \int_0^\infty \frac{\xi^2}{x} dx, \quad R = \int_0^\infty \xi^2 dx. \quad (23)$$

To solve set (17) we used schemes and software developed in [6-8], which yield reasonable accuracy of results. Using relations (19)-(22) and Table 1 one can determine the bipolaron energy and radius over a wide range of parameter  $\alpha$  variation.

#### 4. Possible application of the results

Data on the parameter  $\alpha$  listed in Table 1 enables one to calculate bipolaron states (BS) for a number of ionic crystals. From the calculations follows that bipolaron energy (21) always exceeds  $-2\alpha\hbar\omega_0$ , which is the upper limit for energy of two uncoupled polarons. Thus, the LLP equations do not yield a stable BS. Note, that these equations have solutions for all  $\alpha > 0$ , including those leading to positive total polaron energy. In this case localized solutions exist as the electron energy is negative. Therefore, LLP equations point to the possible existence of unstable BSs. These states may be of physical interest if they are separated from the delocalized polaron states by a potential barrier.

It should be emphasized that our results are based only on the solution of approximate LLP equations and can be modified when using more exact approximations.

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