© 2009

TWO-SITE MODEL FOR A SMALL POLARON: MASS RENORMALIZATION AND OPTICAL CONDUCTIVITY

B. Yavidov[*]

Institute of Nuclear Physics, Academy of Sciences of Uzbekistan
100214, Tashkent, Uzbekistan

The renormalization of the mass of an electron interacting with many ions of a lattice via the long-range (Fröhlich) electron–phonon interaction and optical absorption of electrons are studied at zero temperature. Ions are assumed to be isotropic three-dimensional oscillators. The optical conductivity and the renormalized mass of small adiabatic Fröhlich polarons are calculated and compared with those of small adiabatic Holstein polarons.

1. INTRODUCTION

Polarons have been extensively studied since a seminal paper of Landau [1]. They are divided into small and large polarons in accordance with the size of their wave function. In the first case, a carrier is coupled to intramolecular vibrations and self-trapped on a single site. Its size is the same as the size of the phonon cloud, both are about the lattice constant (so-called small Holstein polaron, SHP). In the case of large polarons, the size of the polaron is also the same as the size of the phonon cloud, but the polaron extends over the distance of many lattice constants.

Polarons with a very different internal structure were introduced in [2, 3]. They were called small Fröhlich polarons (SFP). An SFP size is about the lattice constant, but its phonon cloud spreads over the whole crystal. Within the model in [3], the renormalized mass appears to be much smaller compared with that in the canonical Holstein model [4]. Recently [5], this model was extended to the adiabatic limit and it was found that the SFP mass is renormalized much less than the mass of the SHP in this limit. An electron interacting with vibrations of a chain of ions polarized perpendicular to the chain was considered in [5]. The model was introduced in order to mimic high-T_c cuprates, where in-plane CuO_2 carriers are strongly coupled to the c-axis polarized vibrations of apical oxygen ions [6]. However, apical ions vibrate in all directions. To de-
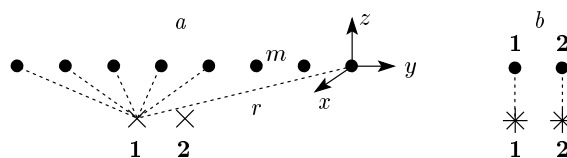
Fig. 1. Electron hops between sites **1** and **2**. In our model, i.e., in the Fröhlich or extended Holstein model (a), the electron at site **1** interacts with $3D$ vibrations of ions (circles in the upper row) in the Holstein model (b), it interacts only with $3D$ vibrations of a single ($m = 1$) ion

scribe a more realistic case, we consider an electron hopping between two sites and interacting with three-dimensional ($3D$) vibrations of ions of the chain, as shown in Fig. 1a. In addition, we calculate the optical conductivity of the system to show that the long-range electron–phonon interaction qualitatively changes the polaron hopping and the optical conductivity compared with these in the Holstein model, Fig. 1b.

2. MASS RENORMALIZATION

We first derive an analytic expression for the renormalized hopping integral of an SFP in the nonadiabatic limit and in the adiabatic limit in order to elucidate the effect of ion's longitudinal vibrations in the renormalized hopping integral. The Hamiltonian of the model

is [2–5]

$$H = H_e + H_{ph} + H_{e-ph},\qquad(1)$$

where ($\hbar = 1$)

$$H_{ph} = \sum_m \left(-\frac{1}{2M}\frac{\partial^2}{\partial \mathbf{u}_m^2} + \frac{M\omega^2 \mathbf{u}_m^2}{2} \right)\qquad(2)$$

is the Hamiltonian of vibrating ions,

$$H_{e-ph} = \sum_{\mathbf{i=1,2}} \sum_m c_{\mathbf{i}}^+ c_{\mathbf{i}} \mathbf{f}_m(\mathbf{i}) \cdot \mathbf{u}_m\qquad(3)$$

describes the interaction between the electron and ions, and

$$H_e = -t(c_{\mathbf{1}}^+ c_{\mathbf{2}} + \text{H.c.})$$

is the electron hopping energy. Here, t is the bare hopping integral between the neighbour sites, $c_{\mathbf{i}}^+$ ($c_{\mathbf{i}}$) is the creation (destruction) operator of an electron on site $\mathbf{i}$, M is the mass of a vibrating ion, ω is its frequency, $\mathbf{u}_m$ is the displacement of an ion on site m, $\mathbf{f}_m(\mathbf{i})$ is the force between the electron on site $\mathbf{i}$ and the mth ion, $\mathbf{f}_m(\mathbf{i}) = \text{const} \cdot r^{-3}\mathbf{e}_{\mathbf{i},m}$, r is the distance between sites $\mathbf{i}$ and m, and $\mathbf{e}_{\mathbf{i},m}$ is the unit vector directed from site $\mathbf{i}$ to site m. Similarly to Refs. [3, 5], the renormalized hopping integral in the nonadiabatic case is

$$\tilde{t} = t\exp(-g^2),\qquad(4)$$

where

$$g^2 = \frac{1}{2M\omega^3}\left[\mathbf{f}_m^2(\mathbf{1}) - \mathbf{f}_m(\mathbf{1}) \cdot \mathbf{f}_m(\mathbf{2}) \right].\qquad(5)$$

We can rewrite renormalized hopping integral as

$$\tilde{t} = t\exp(-\gamma E_p/\omega),\qquad(6)$$

where

$$E_p = \frac{1}{2M\omega^2}\sum_m \mathbf{f}_m^2(\mathbf{1})\qquad(7)$$

is the polaronic shift and

$$\gamma = 1 - \frac{\displaystyle\sum_m \mathbf{f}_m(\mathbf{1}) \cdot \mathbf{f}_m(\mathbf{2})}{\displaystyle\sum_m \mathbf{f}_m^2(\mathbf{1})}.\qquad(8)$$

The value of the factor γ depends on the geometry of the lattice. In our case, the distance between ions along the chain and between sites **1** and **2** is $a = 1$, and the distance from the chain and ion **1** (or **2**) is $b = 1$. In the nonadiabatic limit, a summation over the whole lattice can be performed in (8) with the result $\gamma_{3D} = 0.727797$; in the one-dimensional ($1D$) case, the value $\gamma_{1D} = 0.28678$ has been obtained [7]. We see that
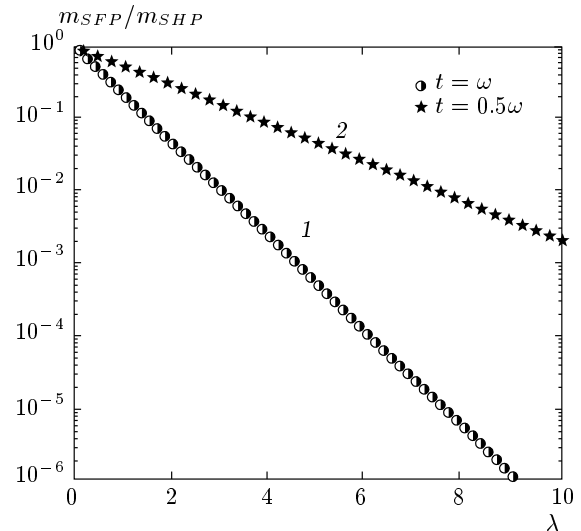

Fig. 2. The ratio of masses of Fröhlich and Holstein small polarons as a function of the electron–phonon coupling constant λ at different values of ω/t in the nonadiabatic regime: 1 — $\omega/t = 1$; 2 — $\omega/t = 2$

an increase in the factor γ in the exponent of (6) increases the mass of an SFP with $3D$ vibrations of ions compared to the mass of an SFP with ions vibrations polarized perpendicular to the chain. Nevertheless, the mass still turns out to be small compared to the mass of a nonadiabatic SHP, because $\gamma = 1$ for the SHP (Fig. 2).

The ratio of masses of the nonadiabatic SFP with $3D$ vibrations of ions to the nonadiabatic SFP with vibrations polarized perpendicular to the chain for the same polaronic shift is given by

$$\frac{m_{3D}}{m_{1D}} = \exp\left(\frac{(\gamma_{3D} - \gamma_{1D})E_p}{\omega} \right).$$

For $t = \omega$ and $\lambda = 2.6$, we obtain $m_{3D}/m_{1D} = 10$.

In the opposite adiabatic regime, we use the Born–Oppenheimer approximation representing the wave function as the wave function $\chi(\mathbf{u}_m)$ describing the "vibrating" ions times the electron wave function $(\psi(\mathbf{u}_m)\,\varphi(\mathbf{u}_m))^T$ with "frozen" ion displacements (T denotes the transposed matrix and $\psi(\mathbf{u}_m)$ and $\varphi(\mathbf{u}_m)$ stand for the respective electron wave functions at site **1** and site **2**). Terms with the first and second derivatives of the "electron" functions $\psi(\mathbf{u}_m)$ and $\varphi(\mathbf{u}_m)$ are small compared with the corresponding terms with derivatives of $\chi(\mathbf{u}_m)$. The wave function of the "frozen" state satisfies the equations

$$\left[E(\mathbf{u}_m) - \sum_m \mathbf{f}_m(\mathbf{1}) \cdot \mathbf{u}_m \right] \psi(\mathbf{u}_m) - t\varphi(\mathbf{u}_m) = 0,$$

$$-t\psi(\mathbf{u}_m) + \left[E(\mathbf{u}_m) - \sum_m \mathbf{f}_m(\mathbf{2}) \cdot \mathbf{u}_m \right] \varphi(\mathbf{u}_m) = 0. \quad (9)$$

The lowest energy is

$$E(\mathbf{u}_m) = \frac{1}{2} \sum_m \mathbf{f}_m^+ \cdot \mathbf{u}_m -$$

$$- \left[\frac{1}{4} \left(\sum_m \mathbf{f}_m^- \cdot \mathbf{u}_m \right)^2 + t^2 \right], \quad (10)$$

where $\mathbf{f}_m^+ = \mathbf{f}_m(\mathbf{1}) + \mathbf{f}_m(\mathbf{2})$ and $\mathbf{f}_m^- = \mathbf{f}_m(\mathbf{1}) - \mathbf{f}_m(\mathbf{2})$. The lowest enrgy plays the role of potential energy in the equation for $\chi(\mathbf{u}_m)$,

$$[E - H_{ph} - E(\mathbf{u}_m)] \chi(\mathbf{u}_m) = 0. \quad (11)$$

Because of the infinite number of variables, Eq. (11) cannot be reduced to a double-well potential problem [4]. Here, we restrict ourselves to the simple case of three ions in the upper chain (ions $m = -1, 0, +1$). Even this simplified five-site model is qualitatively different from the canonical two-site Holstein model (Fig. 1b) and maintains the features of the long-range Fröhlich interaction. By generalizing the transformation formulas in Ref. [5],

$$f_{+1\alpha}^+ u_{-1\alpha} + f_{-1\alpha}^- u_{+1\alpha} = q_\alpha X_\alpha,$$

$$f_{-1\alpha}^- u_{-1\alpha} + (f_{+1\alpha}^+ - f_{-1\alpha}^-) u_{0\alpha} - f_{+1\alpha}^+ u_{+1\alpha} = q_\alpha Y_\alpha$$

and

$$f_{-1\alpha}^- (f_{+1\alpha}^+ - f_{-1\alpha}^-) u_{-1\alpha} - (f_{-1\alpha}^{-2} + f_{+1\alpha}^{+2}) u_{0\alpha} -$$
$$- f_{+1\alpha}^+ (f_{+1\alpha}^+ - f_{-1\alpha}^-) u_{+1\alpha} = (f_{-1\alpha}^{-2} + f_{+1\alpha}^{+2}) Z_\alpha,$$

where $q_\alpha = \sqrt{2(f_{-1\alpha}^{-2} - f_{-1\alpha}^- f_{+1\alpha}^+ + f_{+1\alpha}^{+2})}$, $\alpha = x, y, z$, and introducing a new variable $\xi = Y_x + Y_y$, we can integrate out eight of the nine vibration modes and reduce the problem to the well-known double-well potential problem [4]

$$\left(E - 4\omega + \frac{5}{8} E_p + \frac{\partial^2}{2\mu \, \partial \xi^2} - U(\xi) \right) \chi(\xi) = 0. \quad (12)$$

Here,

$$U(\xi) = \frac{\mu \omega^2 \xi^2}{2} - \sqrt{\frac{3}{4} \mu \omega^2 E_p \xi^2 + t^2} \quad (13)$$

is the familiar double-well potential and $\mu = M/2$. Standard procedure yields for energy splitting $\Delta E = \Delta \exp(-g_F^2)$, where

$$\Delta = \frac{\tilde{\omega}}{\pi} \sqrt{\frac{3E_p}{4\omega}} k^{3/2} \left[1 - \left(\frac{3E_p}{4\omega} k^{3/2} \right)^{-1} \right],$$
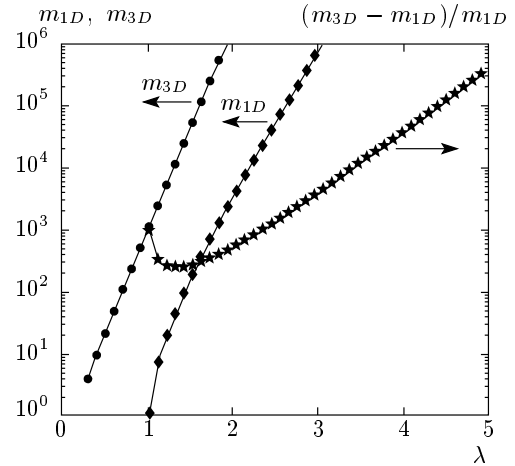

Fig. 3. Mass of the small Fröhlich polaron in units of the band electron mass ($m = 1/2ta^2$) with polarized (m_{1D}) and vector (m_{3D}) ion vibrations, and their relative change as functions of λ in the adiabatic regime, $t/\omega = 5$

$$g_F^2 = \frac{3E_p}{4\omega} k^{3/2} \sqrt{1 - \left(\frac{3E_p}{4\omega} k^{3/2} \right)^{-1}}.$$

Here, $\tilde{\omega} = \omega \sqrt{k}$ is the renormalized phonon frequency, $k = 1 - 1/36\lambda^2$, and $\lambda = E_p/2t$.

The mass of the adiabatic SFP with polarized (perpendicular to the chain) and $3D$ vibrations of ions is plotted in Fig. 3. The relative change in the adiabatic SFP mass, $(m_{3D} - m_{1D})/m_{1D}$, is also plotted in Fig. 3. The magnitude of the mass change of the SFP in passing from polarized phonons to isotropic phonons is even higher in the adiabatic limit. For $\lambda = 1.5$ and $t = 5\omega$, we have $m_{3D}/m_{1D} \approx 377$. It follows that the longitudinal (parallel to the chain) component of ion vibrations increases the SFP mass compared with the SHP mass, as expected [7, 8]. Nevertheless, the net contribution of all vibrations provides much a lighter adiabatic SFP than the adiabatic SHP even with the $3D$ vibrations of ions (Fig. 4).

3. OPTICAL CONDUCTIVITY

Optical conductivity of both small [9–13] and large [14–20] polarons have been studied extensively. In our case of the adiabatic small polaron, the optical absorption is a nearly adiabatic process, and hence we can apply the familiar Franck–Condon principle. Here, we adopt a general formula for the optical conductivity of small polarons, which at zero temperature ($T = 0$) is written as [21]
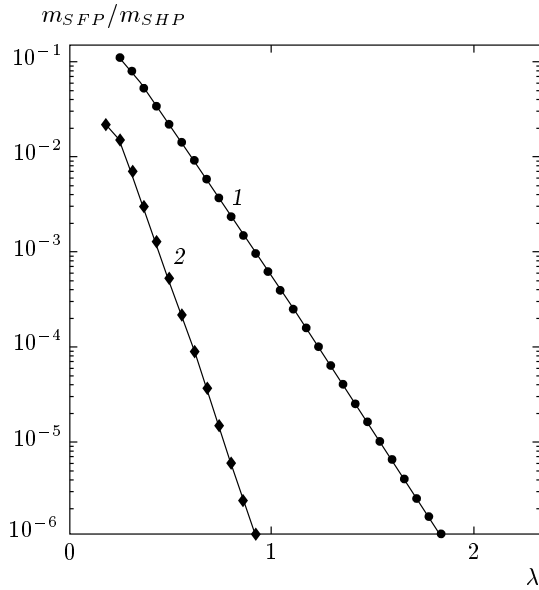
Fig. 4. The ratio of the SFP mass to the SHP mass as a function of λ in the adiabatic regime: $1 - t/\omega = 5$; $2 - t/\omega = 10$

$$\sigma(\nu) = \frac{\sigma_0 \tilde{t}^2}{\hbar\nu\sqrt{2E_a\hbar\omega_{ph}}} \times$$
$$\times \exp\left[-\frac{(\nu-4E_a)^2}{\left(2\sqrt{2E_a\hbar\omega_{ph}}\right)^2}\right], \quad (14)$$

where σ_0 is a constant, ω_{ph} is the phonon frequency, ν is the photon frequency, and E_a is an activation energy for the hopping process. The main difference between polarons with the Holstein and the Fröhlich interactions is that in the former case, the electron deforms only the site where it sits, while in the latter case, it also deforms many neighboring sites. This difference can be seen in diagonal transitions of a polaron from site to site, which ensures a lighter polaron in the Fröhlich model, and in the optical absorption spectra. Due to the photon absorption, the SHP hops to an undeformed site, and $E_a = E_p/2$. But the SFP hops to a deformed neighbouring site, and hence $E_a = \gamma E_p/2$.

As a result, the optical conductivities of the SHP and SFP are very different, as is shown in Fig. 5. In our model, the optical conductivity of the SFP has a more asymmetric Gaussian shape. It is also different from the one in Refs. [14–20], where large Fröhlich polarons were studied using the effective mass approximation, with detailed crystal structure being irrelevant. The optical conductivity of large polarons has an asymmetric shape with a threshold at the optical phonon frequency ω_{ph}. This shape also depends on the
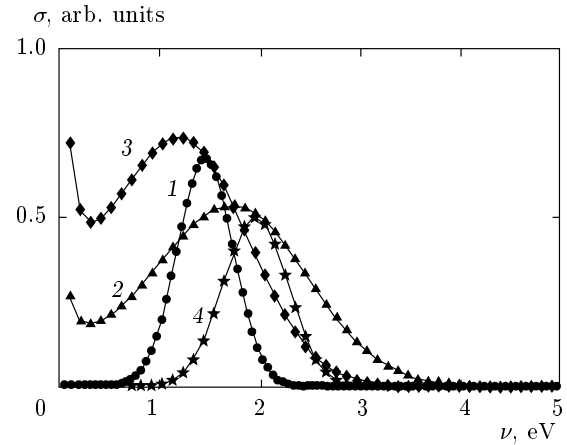

Fig. 5. Optical conductivity of the SFP (curves 1 and 2) and SHP curves (3 and 4) as functions of ν at $E_p = 1$ eV for $\omega_{ph} = 0.1$ eV (curves 1 and 4) and $\omega_{ph} = 0.5$ eV (curves 2 and 3)

many-body (polaron–polaron) interactions [19] and on the magnitude of the polaron coupling constant [15, 20]

$$\alpha_p = \frac{e^2}{2\hbar\omega_{ph}\tilde{\varepsilon}}\sqrt{\frac{2m^*\omega_{ph}}{\hbar}},$$

where m^* is the effective mass of an electron, $\tilde{\varepsilon}^{-1} = \varepsilon_\infty^{-1} - \varepsilon_0^{-1}$, and ε_∞ and ε_0 are the high-frequency and static dielectric constants of the lattice. It was established that optical conductivity spectra of large Fröhlich polarons exhibit relaxed state peaks at moderate and large values of α_p [15].

However, recent quantum Monte Carlo simulations showed that in the strong-coupling regime, peaks due to relaxed excited states are "washed out" by large broadening of these states [20], and optical conductivity spectra of large polarons obtained in [15] are restricted to the region $0 < \alpha_p < 6$. In our discrete model, the optical conductivity is different, its gross features being more reminiscent of the canonical shape of the large-polaron optical conductivity [14, 15].

4. CONCLUSION

We have solved an extended Holstein model with a long-range Fröhlich interaction generalized for the $3D$ vibrations. The small adiabatic Fröhlich polaron is found to be many orders of magnitude lighter than the small Holstein polaron in both the nonadiabatic (see Fig. 2) and adiabatic (see Fig. 4) regimes even with isotropic vector vibrations of ions. The component of ion vibrations parallel to the chain gives rise to a

larger increase in the SFP mass, in agreement with Refs. [7, 8]. But the common effect of all vibrations provides a much smaller renormalization of the SFP mass compared with that of the SHP mass. Optical conductivity of small-size Fröhlich adiabatic polarons has been analyzed and compared with the Holstein model.

REFERENCES

1. L. D. Landau, Phys. Z. Sowjetunion **3**, 664 (1933).

2. A. S. Alexandrov, Phys. Rev. B **53**, 2863 (1996).

3. A. S. Alexandrov and P. E. Kornilovich, Phys. Rev. Lett. **82**, 807 (1999).

4. T. Holstein, Ann. Phys. **8**, 325, 343 (1959).

5. A. S. Alexandrov and B. Ya. Yavidov, Phys. Rev. B **69**, 073101 (2004).

6. T. Timusk, C. C. Homes, and W. Reichardt, in *Anharmonic Properties of High-T_c Cuprates*, ed. by D. Mihailovic et al., World Sci., Singapore (1995), p. 171.

7. S. A. Trugman, J. Bonča, and Li-Chung Ku, Int. J. Mod. Phys. B **15**, 2707 (2001).

8. P. E. Kornilovitch, Phys. Rev. B **59**, 13531 (1999).

9. D. M. Eagles, Phys. Rev. **130**, 1381 (1963); **181**, 1278 (1969); **186**, 456 (1969).

10. H. G. Reik, Sol. St. Comm. **1**, 67 (1963).

11. M. I. Klinger, Phys. Lett. **7**, 102 (1963).

12. H. Böttger and V. V. Bryksin, *Hopping Conduction in Solids*, Academie-Verlag, Berlin (1985).

13. A. S. Alexandrov, V. K. Kabanov, and D. K. Ray, Physica C **224**, 247 (1994).

14. E. Kartheuser, R. Evrard, and J. T. Devreese, Phys. Rev. Lett. **22**, 94 (1969).

15. J. Devreese, J. De Sitter, and M. Goovaerts, Phys. Rev. B **5**, 2367 (1972).

16. J. T. Devreese, in *Polarons in Ionic Crystals and Polar Semiconductors*, North-Holland, Amsterdam (1972).

17. F. M. Peeters and J. T. Devreese, Phys. Rev. B **28**, 6051 (1983).

18. J. T. Devreese, in *Encyclopedia of Applied Physics*, Vol. 14, ed. by G. L. Trigg, VCH Publishers, New York (1996), p. 383.

19. J. Tempere and J. T. Devreese, Phys. Rev. B **64**, 104504 (2001).

20. A. S. Mishchenko, N. Nagaosa, N. V. Prokof'ev, A. Sakamoto, and B. V. Svistunov, Phys. Rev. Lett. **91**, 236401 (2003).

21. G. D. Mahan, *Many-Particle Physics*, Kluwer Acad./Plenum Publ. (2000).