Long-range electron-phonon interactions lead to superlight small bipolarons

J P Hague¹, P E Kornilovitch², J H Samson¹ and A S Alexandrov¹

¹ Department of Physics, Loughborough University, Loughborough, LE11 3TU, UK
² Hewlett-Packard Company, 1000 NE Circle Blvd, Corvallis, Oregon 97330, USA

E-mail: J.P.Hague@lboro.ac.uk

Abstract. A finite-range Fröhlich electron-phonon interaction (EPI) with c-axis polarized optical phonons has been identified in cuprate superconductors by photoemission spectroscopy, in agreement with an earlier proposal by Alexandrov and Kornilovitch [1]. In this article, we discuss the consequences of long-range interactions on phonon-mediated local pairing. First, we examine the effects of modifying interaction range and lattice geometries with regard to analytical strong-coupling/non-adiabatic results for ladder systems. To test the applicability of the analytic results to experimentally achievable couplings and phonon frequencies, we apply a continuous time quantum Monte-Carlo algorithm (CTQMC) to the computation of the effective mass and pairing radius of lattice bipolarons. We demonstrate that bipolarons can be simultaneously small and light due to a novel crab-like motion. Such light, small bipolarons are a necessary precursor to high-temperature Bose-Einstein condensation in solids.

1. Model and motivation

There are a considerable variety of effects due to electron-phonon interactions in solids. The balance between Coulomb repulsion and phonon-mediated attraction is often very subtle, leading to quite different materials such as superconductors, metals and Peierls insulators. Under certain conditions, polarons (which are composite particles consisting of an electron with a phonon cloud) may form, acting like heavy electrons. The balance can be tipped with a small increase in attraction, where the same mechanisms can lead to pairs or intersite bipolarons [1]. Determining the existence and properties of bipolarons is an important question with significant consequences for the theory of superconductivity.

Here, we study the Coulomb-Fröhlich model for electron-phonon interactions on chains [1],

\[
H = -t \sum_{\langle nn' \rangle} c_{n'\sigma}^\dagger c_{n\sigma} + \sum_{nn'\sigma} V(n, n') c_{n\sigma}^\dagger c_{n'\sigma}^\dagger c_{n'\bar{\sigma}} + \sum_m \frac{P_m^2}{2M} + \sum_m \frac{\xi_m^2 M \omega^2}{2} - \sum_{nm\sigma} f_m(n) c_{n\sigma}^\dagger c_{n\sigma} \xi_m.
\]

Sites are numbered by the indices \( n \) or \( m \) for electrons and ions respectively. Operators \( c \) annihilate electrons. The phonon subsystem is a set of independent oscillators with frequency \( \omega \) and mass \( M \). Each vibrating ion can displace by \( \xi_m \), and \( P_m \) is the ion momentum operator.

In the model in figure 1, electrons are mobile along the chain, and hence in-chain Coulomb
with the convention $A_i^\dagger = c_{2x_i-r_i}^\dagger c_{1x_i}^\dagger$, $C_i^\dagger = c_{2x_i+r_i}^\dagger c_{1x_i}^\dagger$, and $B_i^\dagger = c_{2x_i}^\dagger c_{1x_i}^\dagger$, i.e. the index $i$ for the pair is defined on leg 1 of the ladder. States A and C are higher in energy by $V$, since effective attractive interaction is smaller. The Hamiltonian is a specially projected tight binding one, where only hops that result in one of the three states are allowed,

$$\begin{align*}
\hat{H} A_i^\dagger &= -t \left( B_i^\dagger + B_{i-1}^\dagger \right) + VA_i^\dagger \\
\hat{H} B_i^\dagger &= -t \left( A_i^\dagger + C_i^\dagger + C_{i-1}^\dagger + A_{i+1}^\dagger \right) \\
\hat{H} C_i^\dagger &= -t \left( B_i^\dagger + B_{i+1}^\dagger \right) + VC_i^\dagger 
\end{align*}$$

2. Strong coupling antiadiabatic limit: Some simple results for the staggered and rectangular ladders

2.1. Rectangular ladder

For the system in the top panel of figure 1, one may write a wavefunction as a linear combination of the 3 states A, B and C:

$$|\psi\rangle = \frac{1}{N} \sum_i e^{i k \cdot r} \left( a A_i^\dagger + b B_i^\dagger + c C_i^\dagger \right) |0\rangle$$

with the convention $A_i^\dagger = c_{2x_i-r_i}^\dagger c_{1x_i}^\dagger$, $C_i^\dagger = c_{2x_i+r_i}^\dagger c_{1x_i}^\dagger$, and $B_i^\dagger = c_{2x_i}^\dagger c_{1x_i}^\dagger$, i.e. the index $i$ for the pair is defined on leg 1 of the ladder. States A and C are higher in energy by $V$, since effective attractive interaction is smaller. The Hamiltonian is a specially projected tight binding one, where only hops that result in one of the three states are allowed,

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\hat{H} C_i^\dagger &= -t \left( B_i^\dagger + B_{i+1}^\dagger \right) + VC_i^\dagger 
\end{align*}$$

The dimensionless interaction parameter $\lambda = E_p/W = \sum_m f_m^2(0)/2M\omega^2 z t$ where $W$ is the magnitude of the energy of the tight-binding electron. In combination with $\lambda$, the functions $\Phi_0[r_i, r_j] = \sum_m f_m[r_i]f_m[r_j]$ give a universal definition of coupling in models with long-range interaction. In the antiadiabatic limit, $\sigma_{ii'} \to t_{ii'}$, where the renormalised hopping includes the exponential term. Thus, long range interaction is expected to lead to light polarons, which has been demonstrated using CTQMC [2, 3, 4, 5]. $g_{ij} = f_{r_i}(r_j)\sqrt{n/2M\omega^3}$. 

Figure 1. Low energy subspace of the bipolaron problem when the phonon frequency is the highest energy scale. Top panel shows the rectangular ladder. State B is the lowest energy state, followed by A and C. The higher energy states have been projected out (reasonable when coupling is large). The lower panel shows the staggered ladder. States A and B are degenerate, and may be reached via a single hop. No hopping is allowed between legs.
Figure 2. The superlight small bipolaron on rectangular and staggered ladders. The inverse mass and bipolaron radius have been computed for \( \omega = 1 \) and \( \omega = 4 \) and a range of \( \lambda \). There are orders of magnitude difference between inverse masses on rectangular and staggered ladders over significant regions of the strong coupling parameter space (see especially \( \omega = 1 \) results, top). For the very long range Fröhlich interaction used here, the radius is similar on both types of lattice. This washing out effect has been discussed previously with regard to polarons [5].

Substituting into the Schrödinger equation, reindexing the sum on \( i \), and projecting onto states \( \langle A \rangle, \langle B \rangle \) and \( \langle C \rangle \) in turn, it is a simple matter to determine the secular equations:

\[
(E + V)a/\tilde{t} = -b \left( 1 + e^{-i k \tau} \right), \quad (E + V)c/\tilde{t} = -b \left( 1 + e^{i k \tau} \right)
\]

\[
Eb/\tilde{t} = -a \left( 1 + e^{i k \tau} \right) - c \left( 1 + e^{-i k \tau} \right)
\]

which have the solution:

\[
E = V, \left( -V \pm \sqrt{V^2 + 32 \tilde{t}^2 \cos^2(k \tau/2)} \right) / 2
\]

the inverse mass, \( 1/m^{**} = -\partial^2 E/\partial k^2 \big|_{k=0} \) and the inverse polaron mass, \( 1/m^* = 2\tilde{t}\tau^2/\hbar^2 \)

\[
m^{**} = m^* \sqrt{\frac{V^2 m^* 2 \tau^4}{\hbar^4} + 8}
\]

For large \( V/\tilde{t} \), bipolaron mass increases as polaron mass squared: \( m^{**} = [m^*]^2 V \tau^2/\hbar^2 \).

2.2. Staggered ladder
States on the staggered ladder are degenerate, and the wavefunction is just a linear combination of 2 states, A and B. Operation of the hopping term on these states is,

\[
\hat{H} A_i^\dagger = -t \left( B_i^\dagger + B_{i-1}^\dagger \right), \quad \hat{H} B_i^\dagger = -t \left( A_i^\dagger + A_{i+1}^\dagger \right)
\]
leading to secular equations,

\[ \frac{E_a}{\tilde{t}} = -b \left( 1 + e^{i k \cdot \tau} \right), \quad \frac{E_b}{\tilde{t}} = -a \left( 1 + e^{-i k \cdot \tau} \right) \]

with the solution,

\[ E = \pm 2t \cos \left( \frac{k \tau}{2} \right) \quad (6) \]

\[ m^{**} = 4m^* \quad (7) \]

Therefore, one expects bipolarons on staggered ladders, and more generally on systems made of triangular plaquettes to be significantly lighter than those on rectangular systems where dimer bonds must be broken for bipolarons to move [6].

3. Superlight small bipolarons revealed by QMC and future outlook

We compute QMC results for bipolarons moving on staggered and rectangular ladders with periodic boundary conditions for a range of \( \lambda \) with \( \omega/t = 1 \) and \( \omega/t = 4 \). If one is to obtain a superconducting state via the Bose-Einstein condensation of bipolarons, there are two pre-conditions. First, the pair must be light (since mass is inversely proportional to the transition temperature of the BEC). Second, the pairing radius must be small, since this helps keep bipolarons non-overlapping with increasing density (BEC temperature grows with the density). We demonstrate the differences between the inverse masses and radii of the bipolaron in figure 2. There is more than an order of magnitude difference between the masses of bipolarons on staggered and rectangular ladders over significant regions of the strong coupling parameter space. In fact, the bipolaron mass on the staggered ladder has of the same order as the polaron mass over a wide range of \( \lambda \) and \( \omega \) values [7].

We have demonstrated that one of the precursors for a BEC of pairs above the mK range may be met on the staggered ladder arrangement, but we also require small pairs, with non-overlapping wavefunctions. Also shown in figure 2 is the variation of bipolaron size with \( \lambda \). Not only is the bipolaron on the staggered ladder extremely light when compared to the bipolaron on the rectangular ladder, it also has a small radius for wide regions of the parameter space, making such bipolarons an excellent prospect for Bose-Einstein condensation. Here, the very long range interaction leads to similar radii on staggered and rectangular lattices [5]. For shorter range interactions, the bipolaron on the staggered ladder tends to be slightly smaller (see reference [8] for results from interaction ranges on the order of a lattice spacing). These properties extend into 3D systems, and very high transition temperatures may be estimated from them [7].

The final condition for a BEC is the absence of clustering between the pairs. This is likely to occur in a narrow region of the parameter space where the pair binding energy and kinetic energy are of similar size. We are currently extending out algorithm to deal with large numbers of particles to determine the conditions under which bipolaronic superconductors can exist.

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