How can bipolarons manage to stay slim?

1. **Semi-classical theory of bipolaronic superconductivity in a bond-modulated electron-phonon model**
   Authors: Kyung-Su Kim, Zhaoyu Han, John Sous
   Phys. Rev. B 109, L220502 (2024)

2. **Bipolaronic superconductivity out of a Coulomb gas**
   Authors: J. Sous, C. Zhang, M. Berciu, D. R. Reichman, B. V. Svistunov, N. V. Prokof'ev, A. J. Millis
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3. **Bipolaronic high-temperature superconductivity**
   Authors: C. Zhang, J. Sous, D. R. Reichman, M. Berciu, A. J. Millis, N. V. Prokof'ev, B. V. Svistunov
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Recommended with a Commentary by Massimo Capone, Scuola Internazionale Superiore di Studi Avanzati (SISSA) and CNR-IOM, Via Bonomea 265, I-34136, Trieste, Italy

The concept of polaron is almost as old as solid state physics. It has been indeed proposed in 1933 by L. Landau[1], while the name polaron was introduced in 1946 by S. Pekar[2], who developed the compelling image of an electron digging its own potential well. The tool to dig the well is, at least for conventional polarons, the coupling with the lattice vibrations. The idea is simply that an electron which is strongly coupled with the phonons can not move without carrying a phonon cloud. This leads, intuitively, and in several simple models, to an exponentially large effective mass which is often seen as the signature of polaron "self trapping".

Despite its long history, the study of polarons is very timely, as their fingerprints appear in a variety of systems, including strongly correlated materials, where electron-electron interactions dominate and the simultaneous relevance of electron-phonon (e-ph) coupling seems to contradict the common wisdom that these interactions are mutually exclusive. The investigation of polarons is nowadays among the most active fields also in the context of electronic-structure calculations[3], and in cold-atom systems, where polarons can be realized via impurity atoms in a quantum fluid[4].

Last, but not least, polarons can give rise to superconductivity. It is not hard to see that polarons tend to bind in pairs. The mechanism, in a nutshell, is that two electrons

with opposite spin can share the same lattice deformation. In this way, both gain energy via electron-phonon coupling, while the energetic cost of lattice distortion is the same as for a single electrons. Those tightly bound pairs of polarons are called bipolarons, and they can in turn condense, leading to a superconducting state. Such a bipolaronic superconducting state can be seen as the strong-coupling counterpart of the well-known BCS state. Increasing the e-ph coupling the characteristic length of the pairs is reduced and the bipolarons are more and more localized.

A natural question is whether such a bipolaronic superconductor can reach a large critical temperature exploiting the large coupling strength. The general answer is negative, and it follows precisely from the large effective mass. For example in three dimensions the BEC temperature is

$$T_c = \frac{1}{2\pi\zeta(3/2)^{2/3}} \frac{\hbar^2}{m*} n^{2/3} \tag{1}$$

showing that, in the BEC regime, a large effective mass almost inevitably leads to a small T_c (similar results are obtained in two-dimensions using the Berezinskii–Kosterlitz–Thouless expression).

The reduction of T_c in strong coupling leads to a dome-shaped behavior as a function of the coupling and sets an upper bound for phonon-mediated superconductivity*. This scenario is indeed realized in microscopic calculations for the Holstein model, where the local occupation of lattice electrons couples with local distortions, and it essentially the same physics of the attractive Hubbard model.

The papers that I am discussing propose and demonstrate a way out from this destiny. In a nutshell, they provide evidence that, for a model in which the phonons do not couple with the charge, but on "bond" variables, one can reach a bipolaronic regime, with short bipolaronic radius, without paying the price of a large effective mass, or in other words one can realize mobile bipolarons.

This perspective is extremely exciting, as they suggest that the ceiling for phonon-driven superconductivity can be higher than expected. Incidentally, a non-local e-ph coupling is also naturally less affected by a Hubbard U or other local interactions like the Hund's coupling. This implies that the superconductivity of mobile bipolarons can survive to strong correlations or even benefit from them, analogously to what happens in multiorbital models where the coupling involves internal degrees of freedom rather than the total charge.

The proposal of light bipolaron has been put forward in series of recent papers[5–7] addressing two different models with e-ph coupling of the SSH (or Peierls) kind, where the electron hopping is coupled to phonon degrees of freedom, respectively the difference between displacements[5] and a bond phonon[6, 7]. In both cases the key point is that the phonons are not coupled with the electronic charge density, as it happens in the Holstein model. The evidence for the mobile bipolarons has been so far mainly numerical, using variational exact diagonalization and powerful diagrammatic Quantum Monte Carlo methods. In the most

*We do not address here the dependence on the phonon frequency, which appears as a prefactor in the BCS expression for superconductivity, and it is the origin of hogh-temperature superconductivity in high-pressure hydrogen-based compounds

recent of works featured here[8] the authors provide a very welcome physical picture of the reason why this kind of e-ph coupling does not lead to very large effective masses in the case of the bond-SSH model

$$H = -\sum_{\langle i,j\rangle\sigma}(t+\alpha X_{\langle ij\rangle})(c^{\dagger}_{i\sigma}c_{j\sigma}+h.c.)+\sum_{\langle i,j\rangle}(\frac{kX^2_{\langle ij\rangle}}{2}+\frac{P^2_{\langle ij\rangle}}{2M}) \tag{2}$$

In a nutshell, in the adiabatic limit, where the phonons become classical variables, one can define an effective potential which depends on the various phononic displacements associated to each bond $V_{eff}(X_{\langle ij\rangle})$. In the atomic limit of zero hopping, one finds that, in two dimensions, the configurations where $X_{\langle ij\rangle}$ is non-zero on the four bonds connected to a single site form a degenerate energetic minimum. When the hopping is switched on, the configuration shown in Fig. 1 prevails. Increasing the hopping the bipolaronic configurations become more extended in space, even if the growth of the radius of these configurations is quite slow. The existence of this manifold of degenerate configurations is the key to allow for

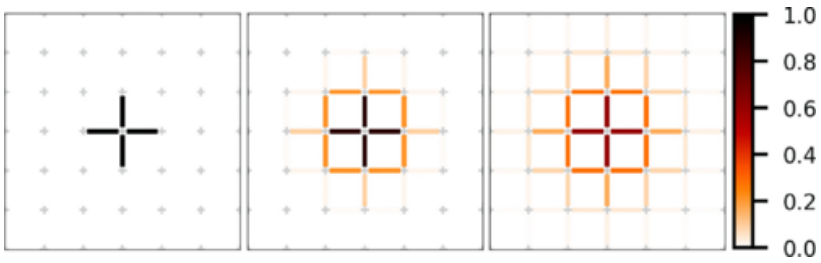

Figure 1: Evolution of the distortion pattern associated with the bipolaron as the ratio between hopping and electron-phonon coupling is increased (Taken from the featured reference 1., where the details of parameters can be found)

a mobile bipolaron. As opposed to the Holstein model, where moving a bipolaron requires to destroy and re-create the phonon cloud, here the bipolaron can slide (using the authors' language) almost freely from one site to another without adjusting the lattice configuration within the degenerate manifold (which means, without energy costs). A pictorial view of the evolution is shown in Fig. 2 which describes the imaginary-time evolution of the process in which the bipolaron moves from one site to another.

In previous work, using accurate estimates of the polaronic radius and effective mass from diagrammatic Quantum Monte Carlo, the authors have been able to estimate the critical temperature in two dimensions including also a screened Coulomb repulsion of the Hubbard form. The results confirms that the mobile bipolaron can lead to higher critical temperature with respect to the standard expectations for electron-phonon theories. In Fig. 3 the authors display the estimated critical temperature for a BKT transition where the pairs are light bipolarons. We see that the result exceeds substantially the Migdal-Eliashberg estimates and they reach a fraction of the phonon frequency not far from 0.2. These results are the measure of the enhancement of critical temperature brought by the mobility of the bipolarons.

The curves as a function of U display that the interplay with the Coulomb repulsion is non-trivial and it follows again a dome-shaped curve. This means that, at least in a

Figure 2: Imaginary-time evolution of the distortion pattern as the bipolaron moves from one site to another (Taken from the featured reference 1., where the details of parameters can be found)

wide window of interactions, the Hubbard repulsion enhances the critical temperature. As a matter of fact even for $U/t \simeq 20$ the critical temperature exceeds that for $U = 0$, even if the effect is maximal for an intermediate U of the order of the bandwidth 8t. These results are consistent with the idea that a phonon mechanism involving the electronic hopping rather than the charge can coexist with the Coulomb interaction[9]. This scenario has been addressed very accurately in the case of alkali-metal doped fullerides[10, 11], where a correlation-driven boost of superconductivity has been explicitly shown within DMFT, and it has been conjectured for SSH-like models, where more indirect signatures have been reported[12].

Also the dependence on the "adiabatic ratio" Ω/t is intriguing and non-trivial. This parameter is indeed often overlooked as in conventional materials it is typically very small because of the large mass difference between ions and electrons which reflects in their characteristic energy scales, but advances in the engineering of quantum materials can lead to design materials with properties that raise Ω/t as, for instance, flat bands.

One of the main messages of the works I discussed is that several conventional ideas about the e-ph interactions are much more specific than believed, ranging from the exponentially large effective mass of the bipolarons to the value of the maximum value of T_c one can achieve.

These results have been obtained in the model of Eq. (2). A fundamental question is whether the model and the mobile bipolaron physics can be relevant for materials of interest. A possible realization of a two-dimensional bond electron-phonon coupling comes indeed from layered materials in which the bonding orbitals of in-plane atoms are coupled with the perpendicular displacement of out-of-plane atoms. This is the case of iron-based superconductors[6], where the position of the pnictogen atoms plays a crucial role[13] and the role of electron-phonon coupling has been revived by observations on monolayer FeSe[14]. While the current evidence does not point towards a purely phononic mechanism for superconductivity in these materials, a potential role of mobile bipolarons should not be overlooked.

Moreover, the advances of the ab-initio methods, can lead us to reliable estimates of the electron-phonon coupling and of the nature of the relevant phonons and their coupling (even for strongly correlated materials), providing us with candidate materials where light bipolarons can be observed.

Last, but not least, for electron-phonon couplings involving the hopping, we expect that electron-electron correlations can coexist with the phonon-driven pairing. This means that conventional (phonon-driven) and unconventional (correlation-driven) superconductivity are

not two words apart and the two mechanisms can have a rich interplay including scenarios in which the they cooperate to increase the critical temperature. Obviously this depends on the specific properties of the materials, in particular on the nature of the relevant phonon modes.

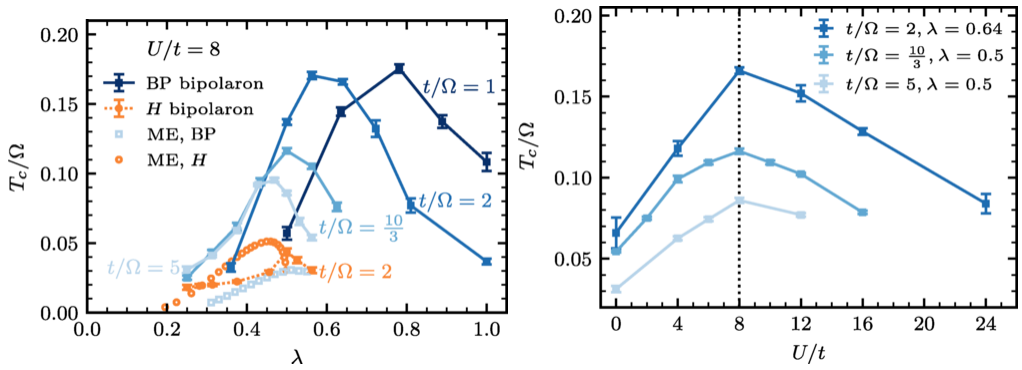

Figure 3: left: Superconducting critical temperature T_c in units of the phonon frequency Ω as a function of the electron-phonon coupling for fixed U and different values of the inverse adiabatic ratio t/Ω. The curves have a maximum that exceeds Migdal-Eliashberg predictions and the typical ceiling for electron-phonon superconductors. Right: critical temperature as a function of U, showing that the Hubbard repulsion can enhance superconductivity and the optimal interaction is of the order of the bandwidth. Taken from the featured reference 3.

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