Two-dimensional superconductivity induced by electronic excitations

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As a nonphonon mechanism of superconductivity, Little has proposed models in which the pairing force is induced by electronic polarizability of molecules embedded in the conductor. Hirsch and Scalapino have studied a model of this type in one dimension and have found that superconductivity is dominated by charge-density wave. Here, their calculations are extended to two dimensions. Numerical data from quantum Monte Carlo calculations suggest that the charge-density-wave formation is limited to a region close to half filling. Outside of that superconductivity develops unhindered. The theoretical results might be relevant to high-temperature superconductors.

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Soon after Bardeen, Cooper, and Schrieffer[1] proposed their theory of phonon-based superconductivity, the search has been underway for an alternate mechanism for superconductivity. In 1964, Little proposed a specific model of electronic (excitonic) origin.[2] He considered a quasi-one-dimensional conductor with densely packed polarizable molecules surrounding the conducting spine as a most likely candidate of excitonic superconductors.

Little's idea is very appealing and has a very broad impact on the experimental search for high-temperature organic superconductors in the following decades. In contrast, rigorous theoretical work on the Little model has been scarce. An exception is a comprehensive work by Hirsch and Scalapino.[3](HS) Analytical as well as quantum Monte Carlo methods were employed by HS to study the competition between various instabilities in the one-dimensional Little model. The unfortunate conclusion is that superconductivity is suppressed by the formation of charge-density wave (CDW). This result is actually not surprising because in one-dimension, CDW can exist for any band filling. Moreover, with the discrete symmetry of its order parameter, CDW is less susceptible to thermal fluctuations and retardation effect than superconductivity, which has a continuous symmetry.

From this viewpoint, two dimensions would provide a new distinct possibility for superconductivity to thrive. The reason is that in two dimensions, CDW has a characteristic periodicity that can exist only near half filling. The case is particularly clear in the Holstein model[4] in which the CDW dominates near half filling, but superconductivity emerges elsewhere in the carrier-concentration range. Following HS, we thus study the following Hamiltonian in two dimensions:

$$
\begin{aligned}
H = & -t\sum_{\langle ij\rangle\sigma}\left[c_{i\sigma}^{\dagger}c_{j\sigma}+\text{H.c.}\right]-t_d\sum_i\left[d_{i1}^{\dagger}d_{i2}+\text{H.c.}\right] \\
& +\sum_{i\sigma}\left[Un_{i\sigma}d_{i1}^{\dagger}d_{i1}-\mu n_{i\sigma}\right] \\
& +\sum_i\left[V_1 d_{i1}^{\dagger}d_{i1}+V_2 d_{i2}^{\dagger}d_{i2}-\mu_d\left(d_{i1}^{\dagger}d_{i1}+d_{i2}^{\dagger}d_{i2}\right)\right],
\end{aligned}
\tag{1}
$$

where $n_{i\sigma}=c_{i\sigma}^{\dagger}c_{i\sigma}$ is a density operator of the conduction electrons. The polarizable side group next to site i is modeled by two atomic orbitals with corresponding creation operators $d_{i1}^{\dagger}$, $d_{i2}^{\dagger}$ and potential energies V_1, V_2, respectively. t_d is the overlap integral between those two orbitals. A possible arrangement of the side group consists of the two atomic orbitals lined up in a direction perpendicular to the conduction plane with orbital 1 being closer to the conduction plane than orbital 2. Along with that we are going to assume that the potential V_1 is lower than V_2 and that the conduction electrons couple to the side groups through a repulsive density-density interaction. The chemical potentials are introduced to control the density of the conduction electrons and to maintain the condition $d_{i1}^{\dagger}d_{i1}+d_{i2}^{\dagger}d_{i2}=1$.

A simple picture of the above Hamiltonian is that the conduction electrons excite (polarize) a side group by displacing the d-electron density away from orbital 1, or equivalently by promoting the d electron from the bonding orbital to the antibonding orbital. Obviously, there exist other ways to implement the coupling between conduction electrons and polarizable molecules. We adopt the model Hamiltonian (1) to facilitate comparison with HS's work in one dimension.

The local virtual excitation can mediate an effective interaction between the conduction electrons. A central question is whether this interaction favors CDW or superconductivty. To answer this question, we carry out quantum Monte Carlo calculations with the Blankenbecler-Scalapino-Sugar (BSS) method suitably extended to handle numerical stability at low temperatures.[5]

A straightforward application of the BSS auxiliary field Monte Carlo methodology leads to large error bars in the CDW correlation function at low temperatures. We find that the large fluctuations are caused by a small percentage (typically less than a few percent) of samples (auxiliary field configurations). The rest of the auxiliary field configurations follow a very smooth probability distribution in the magnitude of the CDW correlation. An extrapolation of this smooth distribution would predict a probability much less than 1% for the occurrence of the aforementioned large fluctuations. To speed up the calculations and to reduce the error bars, in averaging over the auxiliary field configurations we simply

skip those small percentage of samples. Although the fluctuation in superconducting correlation is not as bad as CDW correlation, the Monte Carlo samples are simultaneosly regularized with respect to the superconducting correlation. The regularization of the auxiliary field is not absolutely necessary, but the unregularized sampling takes longer computer time to converge with the result not significantly different from that obtained by the regularized sampling.[6] In other words, regularization provides a convenient yet good approximation.

To present these results, we focus on the two equal-time correlation functions, the *s*-wave pair-field correlation function

$$P_s = \langle \Delta \Delta^\dagger \rangle, \tag{2}$$

with

$$\Delta^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{l}} c^\dagger_{\mathbf{l}\uparrow} c^\dagger_{\mathbf{l}\downarrow} \tag{3}$$

and the charge-density structure factor

$$C(\mathbf{q}) = \langle \rho_{\mathbf{q}} \rho^\dagger_{\mathbf{q}} \rangle \tag{4}$$

with

$$\rho^\dagger_{\mathbf{q}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{l}} e^{i\mathbf{q}\cdot\mathbf{l}} (n_{\mathbf{l}\uparrow} + n_{\mathbf{l}\downarrow}). \tag{5}$$

In two dimensions, the only relevant wave vector of the CDW correlation function is the (π,π) component, $C(\pi,\pi)$. Based on what is learnt from the Hubbard model, the long-range part of the pair-field correlation is a better indicator[7,8] than the fully intergrated one. Therefore, we exclude near neighbors in the double summation (over $\mathbf{l}$ and $\mathbf{l}'$) in Eq. (2). For the 8×8 lattice results to be described, neighbors ($\mathbf{l}$ and $\mathbf{l}'$) with distance less than three lattice spacings are excluded. Other than the regularization problem, the sign problem is also present in the Monte Carlo calculations at low temperatures. In the lowest temperature T we have reached $\beta = 1/T = 14$, the average sign is about 0.04 in the worst case.

Our first set of calculations is done with parameters chosen to produce CDW, at least that is what happens in the one-dimensional case, i.e., $U=2.83$, $t=1$, $t_d=0.5$, $V_1=-U$, and $V_2=0$. The variation of $C(\pi,\pi)$ with the electron site density per spin n (the band filling) is depicted in Fig. 1 for inverse temperature $\beta=12$. Figure 2 exhibits the corresponding variation of the integrated long-range part of the pair-field correlation function P_3. Monte Carlo samples with the magnitude of $C(\pi,\pi)$ larger than 70 or with the magnitude of P_3 larger than 1.5 are discarded. The width of the probability distribution in $C(\pi,\pi)$ is about 10, which is small compared with the cutoff of $|C(\pi,\pi)|$ at 70. Similarly, the width of the distribution in P_3 (of the order of 0.1) is small compared to the cutoff of $|P_3|$. Varying the cutoffs changes the size of the error bars without affecting the essential features.

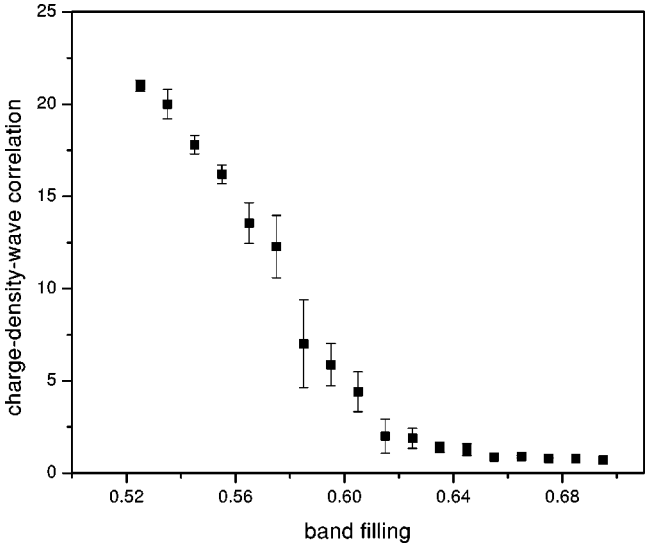

FIG. 1. Charge-density-wave correlation $C(\pi,\pi)$ as a function of band filling for the first set of parameters.

From Figs. 1 and 2, it is clear that $C(\pi,\pi)$ peaks at half filling ($n=0.5$) with a peak value consistent with long-range CDW order and it decreases almost linearly away from half filling. Beyond $n=0.62$, only residual short-range CDW exists. There seems to be a metal-insulator transition near $n=0.62$. In contrast, the pairing-correlation function is small at half filling and rises to a maximum near the region where the long-range CDW disappears. From there on it decays slowly with increasing band filling. Compared to the pairing-correlation function of the noninteracting electron gas (the bottom curve), there is a considerable enhancement in the two-dimensional (2D) exciton model.

The results depicted in Figs. 1 and 2 are strikingly similar to those obtained by Noack, Scalapino, and Scalettar[9] and

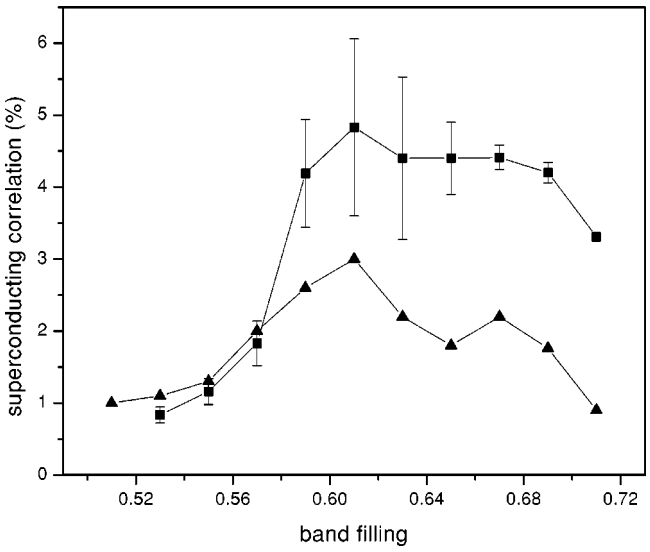

FIG. 2. Long-range superconducting correlation P_3 (in units of 0.01) as a function of band filling for the first set of parameters. The upper curve is for the exciton model, the lower curve is for noninteracting electron gas on the same 8×8 lattice at the same temperature.

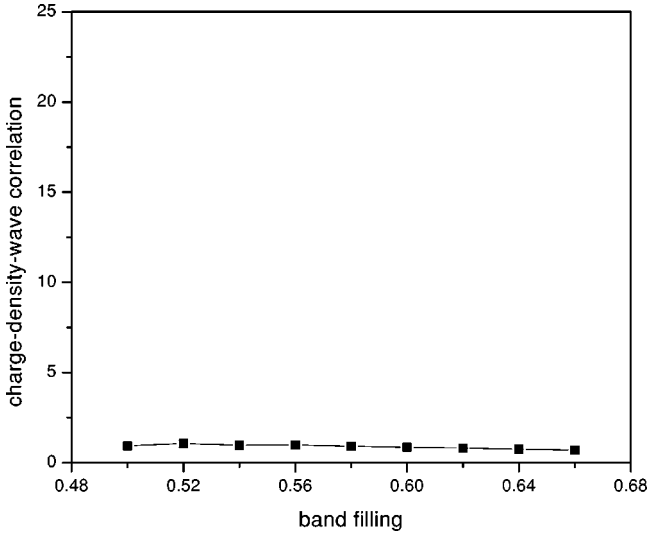

FIG. 3. Charge-density-wave correlation $C(\pi,\pi)$ as a function of band filling for the second set of parameters.

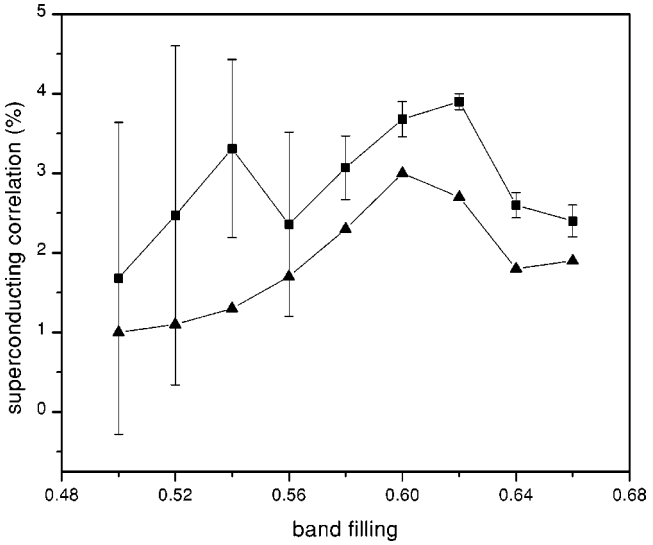

FIG. 4. Long-range superconducting correlation P_3 (in units of 0.01) as a function of band filling for the second set of parameters. The upper curve is for the exciton model, the lower curve is for noninteracting electron gas on the same 8×8 lattice at the same temperature.

others[10] for the 2D Holstein model. Numerical data on the Holstein model indicate[11–14] that outside of the CDW region, the pair-field susceptibility is reasonably well described by the Eliashberg theory, implying the existence of long-range superconductivity. We invoke this analogy to argue that long-range superconductivity exists in the 2D exciton model outside of the CDW region.

As is well known, the massless or high-frequency limit of the Holstein model is the attractive Hubbard model, in which the CDW formation is limited to exactly half filling.[15,16] To simulate this limiting case, we carry out another set of calculations for the following parameters $U=2$, $t=1$, $t_d=2$, $V_1=-U/2$, and $V_2=0$. The inverse temperature is $\beta=14$. The exciton energy given by $\sqrt{4t_d^2+V_1^2}$ is larger than that for the previous parameter set, rendering the effective interaction mediated by an exciton more likely to be attractive. Because of the sign problem, we choose a smaller U than the one-dimensional counterpart considered by HS. Results paralleling Figs. 1 and 2 are contained in Figs. 3 and 4. It is seen from Fig. 3 that CDW remains short ranged even at half filling, at the same time the enhancement in superconducting correlation seems to extend to half filling. These resemble the results of the attractive Hubbard model. A closer comparison requires further study. In this connection, we note that the addition of a next-nearest-neighbor hopping could also significantly suppress CDW and enhance superconductivity near half filling.[14,17]

Based on the results obtained so far, I think that the two-dimensional Little model indeed supports superconductivity. As such it would be one of the very few nontrivial microscopic models of nonphonon superconductivity. To be applicable to realistic materials, certain electron-electron repulsive interactions as well as electron-phonon interaction probably need to be considered.

Instead of the on-site *s*-wave pairing induced by an on-site exciton, one can imagine extended pairing induced by excitons situated between two lattice sites. This could lead to an attractive nearest-neigbor interaction which has been used to model *d*-wave superconductivity[18] as occurred in the cuprates. The on-site *s*-wave pairing would not be favorable because of large repulsion caused by the small copper and oxygen orbitals, as opposed to the large C_{60} orbitals.

It is not clear what part of the cuprates provides the excitons. There has been an interesting discussion[19] about the role of apical oxygen[20] in this context. In general, the layers sandwiched between the CuO_2 planes could be a source of excitons so long as they are polarizable.

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