\( \kappa - (BEDT - TTF)_2X \) organic crystals: superconducting versus antiferromagnetic instabilities in an anisotropic triangular lattice Hubbard model

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A Hubbard model at half-filling on an anisotropic triangular lattice has been proposed as the minimal model to describe conducting layers of \( \kappa - (BEDT - TTF)_2X \) organic materials. The model interpolates between the square lattice and decoupled chains. The \( \kappa - (BEDT - TTF)_2X \) materials present many similarities with cuprates, such as the presence of unconventional metallic properties and the close proximity of superconducting and antiferromagnetic phases. As in the cuprates, spin fluctuations are expected to play a crucial role in the onset of superconductivity. We perform a weak-coupling renormalization-group analysis to show that a superconducting instability occurs. Frustration in the antiferromagnetic couplings, which arises from the underlying geometrical arrangement of the lattice, breaks the perfect nesting of the square lattice at half-filling. The spin-wave instability is suppressed and a superconducting instability predominates. For the isotropic triangular lattice, there are again signs of long-range magnetic order, in agreement with studies at strong-coupling.

I. INTRODUCTION

The family of \( \kappa - (BEDT - TTF)_2X \) layered organic crystals exhibit many fascinating electronic properties.\(^{1-3}\) There are similarities with the high-\( T_c \) cuprates.\(^{4}\) Competition between antiferromagnetic and superconducting instabilities, seen in the cuprates, also appears in the \( \kappa - (BEDT - TTF)_2X \) compounds.

The Hubbard model at half-filling on an anisotropic triangular lattice was proposed by McKenzie\(^3\) as a model of the conducting layers of \( \kappa - (BEDT - TTF)_2X \). It is a simplification of a model first introduced by Kino and Fukuyama.\(^5\) Two hopping matrix elements are considered, \( t_1 \) between sites on a square lattice and \( t_2 \) between next-nearest-neighbors along one of the two diagonal directions (see Fig. 1).

FIG. 1. Anisotropic triangular lattice with two hopping amplitudes \( t_1 \) and \( t_2 \).

The case \( t_1 = t_2 \) thus corresponds to the isotropic triangular lattice. The model interpolates between the square lattice \( (t_2 = 0) \), which has been the subject of many studies in the context of high-\( T_c \) cuprate superconductors and which has a spin-wave instability at half-filling, and completely decoupled chains \( (t_1 = 0) \) for which we have an exact Bethe ansatz solution by Lieb and Wu.\(^6\) The Hamiltonian is given by:

\[
H = -t_1 \sum_{<ij>} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) - t_2 \sum_{<<ij>>} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U_0 \sum_i (n_{i\uparrow} - \frac{1}{2}) (n_{i\downarrow} - \frac{1}{2}) + \mu \sum_i n_i, \tag{1}
\]

where \( <ij> \) denotes nearest-neighbor pairs of sites on the square lattice and \( <<ij>> \) are next-nearest-neighbor pairs along one of the two diagonal directions as shown in Fig. 1.

Values for the hopping amplitudes, obtained from several quantum chemistry calculations\(^7-9\) and for different anions \( X \), are in the range \( t_1 > t_2 \); that is, somewhere between the square and the isotropic triangular lattices. The non-interacting part of the Hamiltonian can be easily diagonalized and the corresponding Fermi surfaces at half-filling are depicted in Fig. 1.

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In the next section we present results from a weak-coupling renormalization-group analysis on this model. We use the approach of Zanchi and Schulz. The Fermi surface is divided into 16 patches. One-loop RG flow equations for all the possible scattering processes involving the patches on the Fermi surface are numerically integrated towards the low-energy limit. All subleading non-logarithmic contributions, arising from six-point functions generated during the mode elimination, are included. We conclude with a discussion of our results and make a comparison with results obtained in the strong-coupling limit.

II. RESULTS

The Hubbard model on the pure square lattice ($t_2 = 0$) and with repulsive on-site interaction $U_0 > 0$ has been extensively studied. Weak-coupling renormalization-group analyses of the couplings, resolved into discrete patches along the Fermi surface, have been performed. At half-filling, since the Fermi surface is fully nested (Fig. 2), an antiferromagnetic (AF) spin-wave instability develops. As the system is doped slightly away from half-filling, there is a crossover to a superconducting BCS regime and the dominant Cooper pairing symmetry is $d_{x^2-y^2}$. As shown in Fig. 3, we have reproduced these results.

Introducing non-zero $t_2$ offers a different way of favoring a BCS instability over AF. Perfect nesting is eliminated once the hopping $t_2$ between next-nearest-neighbors sites along one of the two diagonal directions is turned on (Fig. 1). In contrast to the square lattice where both AF spin-wave and BCS couplings increase during the renormalization-group transformations with $V^{AF}$ diverging faster than $V^{BCS}$, for sufficiently large $t_2$ there is a crossover to a regime where the BCS processes eventually dominate, signaling a superconducting instability. Furthermore, because the Fermi surface is imperfectly nested, the growth in both types of couplings weakens. Further increasing $t_2$ eventually destroys the nesting of the Fermi surface altogether and both types of divergences are suppressed. Three cases illustrating these crossovers are shown in Fig. 4.

FIG. 2. Fermi surface of non-interacting electrons for different values of the hopping amplitudes. The number on the top of each graph is the value of the quantity $t_2/(t_1 + t_2)$, ranging from 0 (square lattice) to 1 (decoupled chains). The chemical potential $\mu$ is varied to ensure that the system is at half-filling.

FIG. 3. Results for the square lattice ($t_2 = 0$). At half-filling ($\mu = 0$), spin-wave instability occurs, but as the system is doped away from half-filling, the perfect nesting is lost and there is a crossover to a BCS instability.

Introducing non-zero $t_2$ offers a different way of favoring a BCS instability over AF. Perfect nesting is eliminated once the hopping $t_2$ between next-nearest-neighbors sites along one of the two diagonal directions is turned on (Fig. 1). In contrast to the square lattice where both AF spin-wave and BCS couplings increase during the renormalization-group transformations with $V^{AF}$ diverging faster than $V^{BCS}$, for sufficiently large $t_2$ there is a crossover to a regime where the BCS processes eventually dominate, signaling a superconducting instability. Furthermore, because the Fermi surface is imperfectly nested, the growth in both types of couplings weakens. Further increasing $t_2$ eventually destroys the nesting of the Fermi surface altogether and both types of divergences are suppressed. Three cases illustrating these crossovers are shown in Fig. 4.
At half-filling, spin-wave instability occurs for $t_2 = 0$, but as $t_2$ increases, the BCS instability wins over. Finally, as $t_2$ is increased further, the nesting is destroyed and both divergences are suppressed.

Going further to the isotropic point, our weak-coupling RG analysis no longer shows BCS instabilities. In Fig. 5 the dominant AF and BCS channels are compared. Neither channel shows strong divergences, but the AF channel is significantly larger than the BCS channel. Slave-boson calculations find a Mott-Hubbard metal-insulator transition to occur at the relatively large value of $U_c = 7.23t$, but also find a metallic phase with incommensurate spiral order at $U < U_c$. Our weak-coupling results seem to indicate the onset of re-entrant long-range antiferromagnetic order.

AF ordering tendencies again disappear as $t_2$ is increased further, beyond $t_1$. This is as expected, since decoupled Hubbard chains do not exhibit AF order.

III. CONCLUSION

Hubbard models on the square lattice have been extensively studied in the context of high-$T_c$ superconductivity. We have reproduced the well-known result that there is an AF instability at half-filling. Also doping the system away from half filling induces a crossover to a BCS regime with $d_{x^2-y^2}$ pairing symmetry, as expected. More importantly, we have shown another way of triggering a BCS instability. Keeping the system at half-filling, but introducing the
diagonal hopping $t_2$ (as shown in Fig. 1), eliminates perfect nesting. Corresponding magnetic frustration kills the spin density wave, and Cooper pairing can dominate. This result suggests that superconductivity may occur in a model of strongly-correlated electrons that interpolates between the square and isotropic triangular lattices.

At the isotropic point, there are signs of re-entrant antiferromagnetic long range order. In the large-$U_0$ limit, at half-filling, the Hubbard model can of course be mapped to the spin-1/2 Heisenberg antiferromagnet which is known to have long-range AF order on the triangular lattice. Furthermore, away from the isotropic point, our weak-coupling RG analysis continues to agree qualitatively with results obtained for the pure Heisenberg system with two exchange couplings $J_1$ and $J_2$. The phase diagram of the Heisenberg model has been studied via a straight $1/S$ expansion, series expansion methods, and a large-N treatment. All three methods find two regions of long-range order: near the limit of a square lattice ($J_2 = 0$) and near the isotropic point ($J_2 = J_1$). It is remarkable that our weak-coupling analysis agrees with these results.

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