

## Gossamer Superconductivity near Antiferromagnetic Mott Insulator in Layered Organic Conductors

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Layered organic superconductors are on the verge of the Mott insulator. We use the Gutzwiller variational method to study a two-dimensional Hubbard model including a spin exchange coupling term as a minimal model for the compounds. The ground state is found to be a Gossamer superconductor at small on-site Coulomb repulsion  $U$  and an antiferromagnetic Mott insulator at large  $U$ , separated by a first order phase transition. Our theory is qualitatively consistent with major experiments reported in organic superconductors.

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There has been much interest recently in the novel physics of layered organic superconductors [1–4]. These compounds share most common physical properties with the high- $T_c$  superconductor but typically with much reduced temperature and energy scales.  $\kappa$ -(BEDT-TTF)<sub>2</sub>X ( $X$  = anion) is a family of the best characterized organic superconductors, where the quasi-two-dimensional (2D) Fermi surface has been observed and a direct first order transition between antiferromagnetic (AFM) insulator and superconductor can be tuned by applied pressure or magnetic fields [5–10]. The resemblance of its pressure-temperature phase diagram to that of the carrier-density-temperature phase diagram in cuprates and the fact of close proximity between the superconducting (SC) and AFM insulating phases have been taken as evidences for similar mechanisms governing high  $T_c$  superconductors. There has been strong evidence that the organic superconductors are at the verge of the Mott insulator [5,6], exhibiting the pseudogap phenomenon [7,8]. While an ongoing debate persists as to the precise symmetry of the singlet pairing, more recent NMR [11], angular dependent STM [12], and thermal conductivity measurements in the vortex state [13] indicate a  $d_{x^2-y^2}$  symmetry.

A schematic molecular structure for the  $\kappa$ -type organic conductors is shown in Fig. 1(a). The average electron density of the relevant band is  $3/2$  per molecule, or the  $1/4$  filled with holes. Since the intradimer hopping strength is much larger than that of interdimer hoppings, the carrier density of the compound is one hole per dimer, and the low energy electronic structure has been well approximated by a 2D Hubbard model at the half filling with each site representing a dimer [14].

Different from the cuprates, organic compounds can be SC at the half filling, which makes the  $t$ - $J$  model commonly used for the cuprates inappropriate to describe the organic SC state. Most theoretical works so far have taken a weak-coupling approach, in which a Hartree-Fock mean

field [14] and a fluctuation-exchange approximation [15] are used. The weak-coupling theory gives a phase diagram of the AFM and SC states qualitatively consistent with the experiments. However, the weak-coupling theory has difficulties to address the Mott insulator or the pseudogap phenomenon [5–8]. The transition between SC and AFM has also been investigated by using the renormalization group method [16].

Very recently, Laughlin has proposed a Gossamer Hamiltonian of which a partially Gutzwiller projected BCS state is an exact ground state with a tiny superfluid density at the half filling [17]. In that Hamiltonian, the SC state has an instability toward the AFM ordering [18]. Some of the present authors [19,20] have examined the Gossamer superconductor, the Mott insulator, and the resonating valence bond (RVB) state [21–23] in strongly correlated electron systems with the hope to unify the superconductivity in cuprates and in organic compounds [24]. In our previous study, we focused on the metallic/SC and insulating nature of the problem and neglected the antiferromagnetism. A related approach was recently taken by Baskaran [25], who introduced a two-species  $t$ - $J$  model

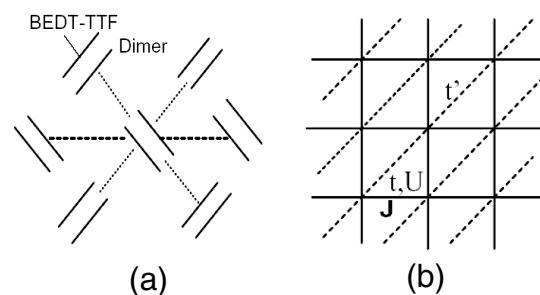


FIG. 1. (a)  $\kappa$ -type BEDT-TTF molecular arrangement within the conducting plane. (b) Triangular lattice structure of the 2D  $t$ - $t'$ - $J$ - $U$  model. We have  $t$ ,  $J$ , and  $U$  terms on the square lattice bonds with the  $t'$  term on the diagonal bonds.

to describe independent motions of empty sites and doubly occupied sites in an otherwise spin-1/2 background, and discussed the relevance of the model to the organic superconductors.

In this Letter, we use Gutzwiller's variational method to study the interplay between SC and AFM states in a modified Hubbard model in 2D given by Eq. (1) below. By using a renormalized mean field theory developed early for the  $t$ - $J$  model [22], we find that at the half filling the ground state is an AFM Mott insulator at large on-site repulsion  $U$  and a Gossamer superconductor at small  $U$ , followed by a normal metallic state at further smaller  $U$ . The transition between the AFM and SC phases is first order, and there is no coexistence of the two phases at the half filling. Our results are qualitatively consistent with major experiments in organic superconductors.

We consider a modified Hubbard model on a lattice as shown in Fig. 1(b),

$$H = U \sum_i n_{i\uparrow} n_{i\downarrow} - \sum_{(ij)\sigma} t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}) + J \sum_{(ij)} \vec{S}_i \cdot \vec{S}_j - \mu \sum_{i\sigma} n_{i\sigma}, \quad (1)$$

where we have chosen the vacuum to be the filled band near the Fermi level.  $c_{i\sigma}^\dagger$  is to create a hole with spin  $\sigma$  in the antibonding state of a  $(\text{ET})_2$  dimer site  $i$ ,  $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ , and  $U > 0$  is the on-site Coulomb repulsion. The nonzero hopping integrals are  $t_{ij} = t$  for the nearest neighbor (nn) pairs and  $t_{ij} = t'$  for the next nn pairs along the  $[1, 1]$  direction.  $\vec{S}_i$  is a spin-1/2 operator, and the summation in the spin exchange term is over all the nn pairs. This Hamiltonian contains an additional spin exchange term to the standard Hubbard model. We may expect the existence of such a spin exchange term at a moderately large value of the on-site Coulomb repulsion. In the limit  $U \gg t$ , the model is reduced to the  $t$ - $t'$ - $J$  model. There is another superexchange term  $J' \approx 4t'^2/U$  between the next nn spins. Since the NMR experiment [8] revealed the presence of a commensurate AFM, we expect that  $J'/J$  is small. The inclusion of the  $J'$  term is expected to further frustrate the commensurate AFM long range order, similar to the effect of the  $t'$  term. Ogata [26] has studied the  $t$ - $t'$ - $J$ - $J'$  model and concluded that the  $d$ -wave pairing symmetry is unchanged for  $J'/J < 0.65$ . Therefore, we expect the qualitative physics including the pairing symmetry will not be altered by the small value of  $J'$  and have neglected this term in our calculations. At the half filling, the large  $U$  limit of the model is reduced to the AFM Heisenberg model with an AFM ground state at small values of  $t'/t$ . At small  $U$ , we expect a metallic or a SC ground state. We believe that the model combined with the Gutzwiller trial wave function approach, Eqs. (2) and (3) below, is appropriate to study the phase transitions in organic superconductors. Note that the direct application of the Gutzwiller trial wave function to the Hubbard model is hardly to obtain the SC pairing because of the nonexplicit form of the spin-spin exchange interaction in the Hamiltonian.

To study the phase transition between the AFM and SC states, we consider a partially Gutzwiller projected spin density wave (SDW)-BCS wave function [27],

$$|\Psi_{\text{GS}}\rangle = \prod_i (1 - \alpha n_{i\uparrow} n_{i\downarrow}) |\Psi_0\rangle, \quad (2)$$

$$|\Psi_0\rangle = \prod_{\vec{k}} (u_{\vec{k}} + v_{\vec{k}} d_{\vec{k}\uparrow}^\dagger d_{-\vec{k}\downarrow}^\dagger) |0\rangle, \quad (3)$$

where  $d_{\vec{k}\sigma} = \cos(\frac{\theta_{\vec{k}}}{2}) c_{\vec{k}\sigma} - \sigma \sin(\frac{\theta_{\vec{k}}}{2}) c_{\vec{k}+\vec{Q},\sigma}$ , and  $\vec{Q} = (\pi, \pi)$  is the magnetic wave vector.  $\prod_i (1 - \alpha n_{i\uparrow} n_{i\downarrow})$  is a Gutzwiller projection operator, which partially projects out the doubly occupied electron states on every lattice site and  $0 \leq \alpha \leq 1$  measures the strength of the projection. Obviously,  $\alpha = 0$ , and  $\alpha = 1$  correspond to a nonprojected and a completely projected state, respectively. At  $\theta_{\vec{k}} = 0$ , we have  $d_{\vec{k}\sigma} = c_{\vec{k}\sigma}$ , and  $|\Psi_{\text{GS}}\rangle$  is reduced to a partially projected BCS state, which we loosely call "Gossamer SC state" [17,19]. In the limit  $u_{\vec{k}} v_{\vec{k}} = 0$ ,  $|\Psi_0\rangle$  is reduced to a SDW state. The variational parameters are  $u_{\vec{k}}$ ,  $v_{\vec{k}}$ ,  $\theta_{\vec{k}}$ , and  $\alpha$ . Such a wave function should enable us to study the phase transition between the AFM and SC states. The metallic or insulating phase can be determined by the continuity of the chemical potential.

To carry out the variation, we apply the Gutzwiller approximation to replace the effect of the projection operator by a set of renormalization factors, which are determined by statistical countings [22,28,29]. Let  $\langle O \rangle$  be the expectation value of the operator  $O$  in the state  $|\Psi_{\text{GS}}\rangle$ , and  $\langle O \rangle_0$  be that in the state  $|\Psi_0\rangle$ . The Gutzwiller approximation gives

$$\langle c_{i\sigma}^\dagger c_{j\sigma} \rangle = g_t^{ij} \langle c_{i\sigma}^\dagger c_{j\sigma} \rangle_0, \quad \langle \vec{S}_i \cdot \vec{S}_j \rangle = g_s \langle \vec{S}_i \cdot \vec{S}_j \rangle_0, \quad (4)$$

where  $g$ 's are determined by the ratio of the probability of the corresponding physical processes in the projected and unprojected states [22]. We introduce a staggered magnetization for sublattices  $A$  and  $B$ ,

$$m_0 = \frac{1}{2} \langle n_{A\uparrow} - n_{A\downarrow} \rangle_0 = -\frac{1}{2} \langle n_{B\uparrow} - n_{B\downarrow} \rangle_0. \quad (5)$$

$g$ 's are then functions of the electron density  $n$ ,  $m_0$ , and the double occupation number  $d = \langle n_{i\uparrow} n_{i\downarrow} \rangle$ ,

$$g_s = (n - 2d)^2 / (n - 2n_+ n_-)^2, \quad g_t^{ij} = G^i G^j, \quad (6)$$

$$G^A = g_s^{1/4} [s(1 - n_-) + \sqrt{n_- d / n_+}],$$

$$G^B = g_s^{1/4} [s(1 - n_+) + \sqrt{n_+ d / n_-}].$$

In the above equations,  $n_{\pm} = \frac{n}{2} \pm m_0$ , and  $s = \sqrt{\frac{1-n+d}{(1-n_+)(1-n_-)}}$ . The superindex in  $G$  refers to the sublattice of the site. Note that there is a one-to-one correspondence between  $d$  and  $\alpha$  given by

$$1 - \alpha = s^2 d / g_s n_+ n_-. \quad (7)$$

In the absence of the staggered magnetization,  $g_t$  and  $g_s$  in Eq. (6) are reduced to their values in the uniform state [20], which are further reduced, in the fully projected case

( $\alpha = 1$  or  $d = 0$ ), to the values in the RVB state [22]. Within the Gutzwiller approximation, the variation of the projected state for  $H$  in (1) is reduced to the variation of the unprojected state  $|\Psi_0\rangle$  for a renormalized Hamiltonian  $H_{\text{eff}}$ ,

$$H_{\text{eff}} = Ud - \sum_{(ij)\sigma} g_t^{ij} (c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}) + g_s J \sum_{(ij)} \tilde{S}_i \cdot \tilde{S}_j - \mu \sum_{i\sigma} n_{i\sigma}. \quad (8)$$

To proceed further, we introduce a self-energy  $\chi$  and a  $d$ -wave pairing amplitude  $\Delta$ ,

$$\chi = \sum_{\sigma} \langle c_{i\sigma}^\dagger c_{i+\hat{x}\sigma} \rangle_0 = \sum_{\sigma} \langle c_{i\sigma}^\dagger c_{i+\hat{y}\sigma} \rangle_0, \quad (9)$$

$$\Delta = \sum_{\sigma} \langle \sigma c_{i\sigma} c_{i+\hat{x}-\sigma} \rangle_0 = - \sum_{\sigma} \langle \sigma c_{i\sigma} c_{i+\hat{y}-\sigma} \rangle_0, \quad (10)$$

The singlet SC order parameter  $\Delta_{\text{SC}} \approx g_{\text{SC}} \Delta$ , with  $g_{\text{SC}} = (g_t^{AA} + g_t^{BB})/2$ . The pairing amplitude and the SDW state described below defines the variation of  $|\Psi_0\rangle$ . As in the usual SDW variation, we choose  $\cos\theta_{\vec{k}} = \epsilon_{\vec{k}}/\xi_{\vec{k}}$ , where  $\epsilon_{\vec{k}} = -(2tg_t^{AB} + 3Jg_s\chi/4)\gamma_{\vec{k},+}$  is the kinetic energy including a self-energy term of  $\chi$ , and  $\xi_{\vec{k}} = \sqrt{\epsilon_{\vec{k}}^2 + \tilde{\Delta}_{AF}^2(\vec{k})}$ , with  $\tilde{\Delta}_{AF}(\vec{k}) = \Delta_{af} + t'(g_t^{AA} - g_t^{BB})\zeta_{\vec{k}}$ .  $\Delta_{af}$  is a variational parameter to determine  $m_0$ . The second term in  $\tilde{\Delta}_{AF}$  arises from a spin-dependent hopping process along the  $[1, 1]$  direction in  $H_{\text{eff}}$ . In the above equations, we have denoted  $\gamma_{\vec{k},\pm} = \cos k_x \pm \cos k_y$ , and  $\zeta_{\vec{k}} = \cos(k_x + k_y)$ . With the above variational wave function, we calculate the expectation value of  $H_{\text{eff}}$  and find the ground state energy,

$$E = Ud - 4g_t\chi + g_t^{AA}\langle H_t^+ \rangle_0 + g_t^{BB}\langle H_t^- \rangle_0 - (3g_sJ/4)(\Delta^2 + \chi^2) - 2Jg_s m_0^2, \quad (11)$$

where  $m_0$ ,  $\chi$ ,  $n$ , and  $\Delta$  are the solutions of their corresponding self-consistent equations. The two additional variational parameters  $d$  and  $\Delta_{af}$  are to minimize the ground state energy. Note that  $0 \leq d \leq d_0$ , with  $d_0 = \langle n_{\uparrow} n_{\downarrow} \rangle_0$ . In Eq. (11),  $\langle H_t^\pm \rangle_0$  are given by

$$\langle H_t^\pm \rangle_0 = -\frac{2t'}{N} \sum_{\vec{k} \in A} \zeta_{\vec{k}} [v_{\vec{k}}^2 (1 \mp \sin 2\theta_{\vec{k}}) + v_{\vec{k}+\vec{Q}}^2 (1 \pm \sin 2\theta_{\vec{k}})],$$

where the summation of  $\vec{k}$  runs over the reduced Brillouin zone, and

$$v_{\vec{k}}^2 = \frac{1}{2} [1 - (\xi_{\vec{k}} - \tilde{\mu})/E_{\vec{k}}^-],$$

$$v_{\vec{k}+\vec{Q}}^2 = \frac{1}{2} [1 + (\xi_{\vec{k}} + \tilde{\mu})/E_{\vec{k}}^+],$$

with  $E_{\vec{k}}^\pm = \sqrt{(\xi_{\vec{k}} \mp \tilde{\mu})^2 + \Delta_{\vec{k}}^2}$ , and  $\Delta_{\vec{k}} = (3/4)Jg_s\Delta\gamma_{\vec{k},-}$ ,  $\tilde{\mu} = \mu + t'(g_t^{AA} + g_t^{BB})\zeta_{\vec{k}}$ .

We are now ready to discuss our results. We focus on the half filled case. There is a critical  $U_c$  to separate a metallic or SC state at a small  $U$  from an AFM insulator at a large  $U$ , and the transition is first order with no coexistence of

the two phases. These features are demonstrated in Fig. 2. There are two regimes in  $U$ . At  $U < U_c (\sim 5.5t)$ ,  $m = 0$  while  $\Delta$  and  $\Delta_{\text{SC}}$  increase monotonically as  $U$  increases.  $\Delta_{\text{SC}}$  is slightly smaller than  $\Delta$ . This is a SC state without AFM ordering. At  $U > U_c$ , but smaller than  $U_0$  with  $U_0 \approx 40t$  for  $J/t = 0.5$ ,  $\Delta = \Delta_{\text{SC}} = 0$ , while  $m = \sqrt{g_s}m_0$  changes abruptly from zero at  $U < U_c$  to a saturated value of 0.45. We have calculated the chemical potential at the half filling and found that it is discontinuous in the AFM state so that the state is an insulator. As we can see from the bottom panel of Fig. 2, as  $U$  increases,  $d$  decreases with a sudden drop at  $U_c$ , indicating the electron's localization in the insulating phase, and  $\alpha$  increases to its maximum in the SC phase followed by a discontinuous drop to zero at  $U = U_c$ . The latter indicates the absence of the projection in the AFM phase so that we have  $m = m_0$ . The phase transition in the parameter space  $U/t$  is a large first order. In addition to the sudden change of the magnetization, the energy derivative with respect to  $U$  also changes abruptly from 0.15 to 0.07 at  $U_c$  for the parameters in Fig. 2. This may suggest that the first order transition is robust. At  $U > U_0$  the  $J$  term dominates in energy; the ground state is a completely projected RVB state with the AFM long range order identical to that of the Heisenberg model [30]. We have also calculated these quantities with different values of  $J/t$  and  $t'/t$  and the results are qualitatively similar except that  $\Delta$  becomes very tiny at smaller  $J/t$ . As shown in the pressure experiments [6], the phase transition is first order and the phase boundary between the AFM and SC states merges with the phase boundary between the insulating and metallic states. In our theory, the AFM state is always a Mott insulator. Recent NMR experiments [5,8] show the proximity of pseudogapped superconductor and a commensurate AFM ordering with a finite moment of  $0.4\mu_B$  (or  $0.26\mu_B$ ) for  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Cl or Br at low temperatures, which suggests that the magnetic ordering is driven by the electron's strong correlation rather than by the Fermi surface nesting. In the Gossamer

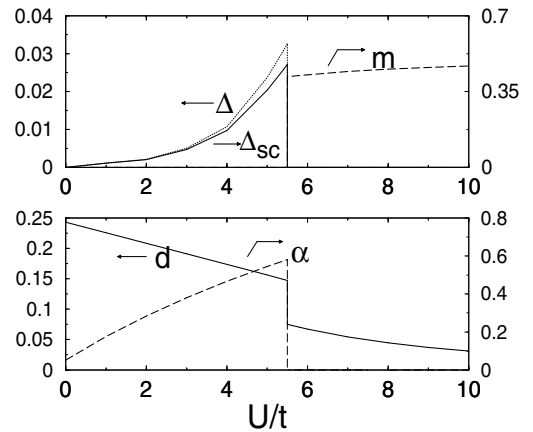


FIG. 2. Pairing amplitude  $\Delta$ , SC order parameter  $\Delta_{\text{SC}}$ , and AFM order parameter  $m$  (top panel), and electron double occupancy number  $d$  and the projection parameter  $\alpha$  (bottom panel), as functions of  $U$  for  $J/t = 0.5$  and  $t'/t = 0.8$ .

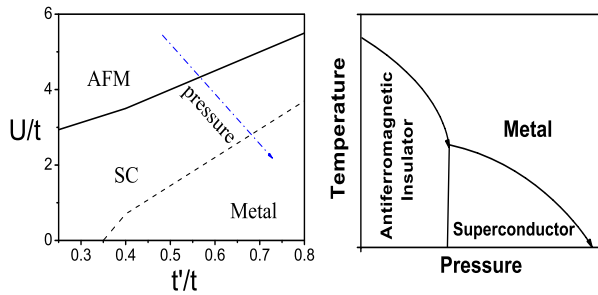


FIG. 3 (color online). Left panel: phase diagram of  $t'$  vs  $U$  for  $J/t = 0.5$  at the half filling. A schematic arrow indicates a possible flow of the parameters under the pressure. Right panel: schematic phase diagram of organic superconductors.

SC state, the quasiparticle energy is governed by  $\Delta$  [20–22], which is larger than the SC order parameter, implying a pseudogap phase. The smallness in the difference between  $\Delta_{SC}$  and  $\Delta$  in our theory is partly due to the phenomenological model we use, which favors the AFM state more than the Hubbard model does at moderate or large  $U$ . We expect the phase boundary in a more accurate theory will be shifted to the larger  $U$  and  $\Delta_{SC}/\Delta$  will be smaller.

Figure 3 displays the phase diagram in the parameter space of  $t'$  and  $U$  with fixed  $J/t = 0.5$  at the half filling [31]. There are three distinct phases. The system is in the AFM phase at large  $U$  and small  $t'$ , the paramagnetic metallic phase at small  $U$  and large  $t'$ , and the SC phase at the intermediate parameter region. Here we have defined a paramagnetic metallic phase if  $\Delta \leq 0.01$ . At this very small  $\Delta$ , the energy difference between a SC state and a normal metallic state is practically indistinguishable. The phase boundary between the SC and normal states thus obtained is indicated by a dashed line. For comparison, a schematic phase diagram abstracted from experimental measurements is shown in the right panel. Details of the pressure-temperature phase diagram of the AFM insulating salt have been reported [6,9,10]. The effect of pressure in the schematic phase diagram is to decrease  $U/t$  and/or to increase  $t'/t$ . Our theory is qualitatively consistent with the general features of this experimental phase diagram.

In summary, we have presented a strong coupling variational theory to examine the superconductivity near antiferromagnetic Mott insulator in layered organic conductors by using a Hubbard model including a spin-spin coupling term. The theory appears qualitatively consistent with a number of major experiments. The present model is still a minimal model. It will be interesting to develop a strong coupling theory based on a more realistic Hamiltonian to explain and predict experiments more quantitatively.

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- [31] At  $t' = 0$  and  $U = 0$ , the ground state of the model (1) is found to be SC with a small but finite value of  $\alpha$  with  $g_s > 1$ . The state prefers to be partially projected so that the SC instability is enhanced. We note that the quantum Monte Carlo calculations ( $J/t > 2$ ) [32] indicate the ground state to be AFM.
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