A Possible Mechanism of the Pseudogap in Organic Superconductor Based on the Superconducting Fluctuation

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The explanation of the anomalous behavior in κ-type (BEDT-TTF)$_2$X which was revealed by the nuclear magnetic resonance experiments is presented. We calculate the electronic properties by using the one-loop approximation for the superconducting fluctuation. The decrease of the density of states due to the large damping effect is obtained and the one-particle spectrum has a pseudogap structure around the Fermi level. It is found that these behaviors are peculiar to the quasi two-dimensional system and are enhanced in the incoherent metal. The similarity between the cuprates and this compounds is discussed and an experiment to check this proposal is suggested.

KEYWORDS: organic superconductor, superconducting fluctuation, pseudogap, one-particle spectrum, quasi two-dimensional system, incoherent metal

Organic compounds are known to show various phases including superconductivity under controlled temperature and pressure. In these compounds κ-type (BEDT-TTF)$_2$X (abbreviated as κ-(ET)$_2$X hereafter, and X=Cu(NCS)$_2$, Cu[N(CN)$_2$]Br, Cu[N(CN)$_2$]Cl) has attracted much attention because it has the highest superconducting transition temperature. The characteristic properties of κ-(ET)$_2$X are that the two-dimensional tight-binding approximation well explains the Shubnikov-de Haas experiments and the superconducting phase neighbors the antiferromagnetic phase under a controlled pressure. About the properties of the superconductivity of κ-(ET)$_2$X, the nuclear magnetic resonance (NMR) and the specific heat experiments of κ-(ET)$_2$X suggest that the superconducting gap has line nodes. Above the superconducting transition temperature ($T_c$), κ-(ET)$_2$X shows the anomalous properties, which were revealed by the NMR experiments. The striking character of the anomalous properties is that the uniform magnetic susceptibility and $1/T_1T$ ($T_1$ is the spin lattice relaxation rate and $T$ is temperature) are not independent of temperature dependence but decrease with decreasing temperature below $T \simeq 50\text{K}$. (This phenomenon may be called the spin gap or the pseudogap.)

Theoretically, the importance of the electron correlation in κ-(ET)$_2$X is suggested because its dimerized structure makes the system half-filled. The mechanism of superconductivity of κ-(ET)$_2$X is investigated using several approaches like the fluctuation exchange approximation (FLEX),$^{8,9}$ the third-order perturbation theory (TOPT)$^{10}$ and the quantum Monte Carlo simulation.$^{11}$ From these studies, it is found that this superconductivity is a spin fluctuation mediated one and the symmetry of the Cooper pair is $d$-wave in this compound. However, for the anomalous properties above $T_c$, there has been no theoretical proposal about the origin of these behaviors until now, and the aim of this paper is to propose a possible mechanism of the (pseudo-)spin gap in κ-(ET)$_2$X.

κ-(ET)$_2$X has a strong anisotropy in the conductivity, so that, it can be considered that this compound is a quasi two-dimensional system. From FLEX and TOPT, it is also known that $T_c$ of κ-(ET)$_2$X is rather high when $T_c$ is scaled with the bandwidth. Regarding the properties of κ-(ET)$_2$X stated above, it can be considered that the superconducting fluctuation plays an essential role in the appearance of the (pseudo-) spin gap.

There are many theories on the pseudogap in cuprates on the basis of the superconducting fluctuation, but these calculations are restricted to a two-dimensional (2D) case$^{13,14}$ and the effect of the dimensionality has not been considered yet.

In this Letter, we investigate the electronic properties for several values of the anisotropy parameter and for those of the attractive force by using the one-loop approximation for the superconducting fluctuation. From the nature of the one-loop calculation, our interest doesn’t lie in the effect of the fluctuation on $T_c$, but in the electronic properties above $T_c$. (Therefore $T_c$ is the mean field transition temperature in our discussion.) It is revealed that, owing to the superconducting, fluctuation the damping rate of quasiparticles increases and then the density of states (DOS) at the Fermi level decreases; this possibly explains the anomalous behavior. We also obtained the result that the one-particle spectrum has a pseudogap structure. This behavior is expected to be observed in the angle-resolved photoemission spectroscopy (ARPES) experiments. It is found that this pseudogap behavior is a characteristic property of a quasi 2D system such as κ-(ET)$_2$X. Furthermore, this behavior is enhanced in the incoherent metallic region where the spectrum is thermally broadened.

To observe the effect of the dimensionality, we consider...
the following Hamiltonian,
\[ \mathcal{H} = \sum_{\mathbf{k}, \sigma} \epsilon_{\mathbf{k}} \hat{c}_{\mathbf{k}, \sigma}^\dagger \hat{c}_{\mathbf{k}, \sigma} - \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} g_{\mathbf{k}, \mathbf{k}'} \hat{c}_{\mathbf{k}, \uparrow}^\dagger \hat{c}_{\mathbf{k}', \downarrow} \hat{c}_{\mathbf{k}', \uparrow} \hat{c}_{\mathbf{k}, \downarrow} \hat{c}_{\mathbf{k}, \uparrow}^\dagger \hat{c}_{\mathbf{k}', \downarrow}^\dagger \hat{c}_{\mathbf{k}', \uparrow}^\dagger \hat{c}_{\mathbf{k}, \downarrow}^\dagger. \]

Here \( \epsilon_{\mathbf{k}} \) is the noninteracting energy dispersion,
\[ \epsilon_{\mathbf{k}} = -2t(\cos k_x + \cos k_y) - 2t' \cos(k_x + k_y) - 2t_z \cos(2k_z) - \mu. \]
\( \mu \) is the chemical potential, and we adopt the dimer model. Therefore \( \mu \) is determined so as to fix the particle number per site to 1. We take \( t \) as the unit of energy, and fix the ratio \( t'/t \) to 0.70 as a realistic value. It should be noted that we include the inter-plane hopping term in the energy dispersion. The essential point to study the effect of the dimensionality, is not the form of this term but the ratio between \( t, t' \) (intra-plane hopping) and \( t_z \) (inter-plane hopping). We take the separable d-wave interaction as the attractive force, that is, \( g_{\mathbf{k}, \mathbf{k}'} = g \phi_{\mathbf{k}} \phi_{\mathbf{k}'} \).

On the basis of this model, we take the superconducting fluctuation into account by the one-loop approximation, which is taking T-matrix up to the first order in calculating the self-energy. The self-energy is written as the following form;
\[ \Sigma(k) = \frac{T}{N} \sum_q T_{\mathbf{k}, \mathbf{k}'}(q) G_0(q - k). \]

Here, \( N \) is the number of sites, \( G_0 \) is the bare Green’s function and \( T_{\mathbf{k}, \mathbf{k}'}(q) \) is the T-matrix;
\[ T_{\mathbf{k}, \mathbf{k}'}(q) = -\phi_{\mathbf{k}} D(q) \phi_{\mathbf{k}'} \]
where \( D(q) \) is the boson propagator;
\[ D(q) = \frac{1}{g} \frac{1}{1 - \frac{T}{g} \sum_{\mathbf{k}} \phi_{\mathbf{k}}^2 G_0(q - k)}. \]

Now, we show the results obtained by our calculation. First, we see the effect of the dimensionality by calculating the electronic properties for different values of \( t_z \). The calculated self-energies for the inter-plane transfer \( t_z = 0.10 \) and 1.00 are shown in Fig. 1. The momentum of the self-energies is fixed to the Fermi momentum situated on the line connecting \( \Gamma \) point \((0, 0, 0)\) and \( M \) point \((\pi, 0, 0)\). At this point called \( \Delta \) (see Fig. 5(a)), the attractive force takes the strongest value due to the \( d_{x^2-y^2} \)-wave symmetry. For the isotropic case \((t_z = 1.00)\), the behavior of the self-energy is the same as that of the conventional Fermi liquid. That is, \(-\text{Im} \Sigma(k, \omega)\) has a local minimum at \( \omega = 0 \) and the slope of \( \text{Re} \Sigma(k, \omega) \) around \( \omega = 0 \) is negative. These properties of the self-energy indicate the well-defined quasiparticle. However, for the quasi 2D case \((t_z = 0.10)\), the behavior of the self-energy is opposite to that of the isotropic case. The peak of \(-\text{Im} \Sigma(k, \omega)\) at \( \omega = 0 \) indicates that the quasiparticle is not well-defined, and the positive slope of \( \text{Re} \Sigma(k, \omega) \) around \( \omega = 0 \) indicates that the spectral weight of quasiparticles near the Fermi surface decreases. Therefore the DOS at the Fermi level is expected to be suppressed. The fact that these anomalous behaviors of the self-energy behind the precursor of the superconductivity was first pointed out by Janko, et al. In addition to this fact, it is pointed out here that these anomalous properties are seen only in the quasi 2D system.

To observe the relationship between the self-energy and the DOS, the temperature dependences of \( \rho(0) \) (the DOS at the Fermi level) and \(-\text{Im} \Sigma(k_F, 0)\) are shown in Fig. 2. It can be seen that for the quasi 2D case \((t_z = 1.00)\) \( \rho(0) \) begins to decrease far above \( T_c \) with the increase of the damping rate of quasiparticles near the Fermi level, \(-\text{Im} \Sigma(k_F, 0)\). On the other hand, for the isotropic case \((t_z = 1.00)\) \( \rho(0) \) increases down to \( T_c \) because the system acquires coherency as the temperature is decreased, which is supported by the decrease of the damping rate of quasiparticles, \(-\text{Im} \Sigma(k_F, 0)\). Although, as discussed below, two other factors are required for the quantitative comparison with experimental results, the decrease of \( \rho(0) \) over a wide temperature range suggests that the precursor of the superconductivity can be a candidate mechanism which gives rise to the anomalous behavior in \( \kappa-(ET)_2X \), because the quantities showing the anomalous behavior in the experiments are related to the DOS at the Fermi level.

The above discussion is described analytically by assuming that the boson propagator is written in the following form;
\[ D(q, \omega) \approx \frac{1}{M - i\alpha \omega + \xi^2 q^2}. \]  

Here, \( M \) is the mass term which describes how far the system is from the superconducting state and is written as \( M = \frac{1}{2} - \sum_k \phi_k^2 \frac{k^2}{2M}, \ M = 0 \) at \( T = T_c \).

With the use of this form for the boson propagator, \(-\text{Im}\Sigma(k_F, 0)\) can be written in the following form:

\[ -\text{Im}\Sigma(k_F, 0) \approx \frac{T \phi^2_{k_F}}{16\pi v_F \xi^2} \log(1 + \frac{\xi q_c)^2}{M}), \]  

for a 3D system, and

\[ -\text{Im}\Sigma(k_F, 0) \approx \frac{T \phi^2_{k_F}}{2\pi v_F \xi \sqrt{M}} \tan^{-1} \left( \frac{\xi q_c}{\sqrt{M}} \right), \]  

for a 2D system. Here, \( q_c \) is the cutoff momentum of the boson, and \( v_F \) is the velocity of the electron at the Fermi surface (FS).

From these equations, it can be seen that for a 2D system \(-\text{Im}\Sigma(k_F, 0)\) is large over a wide temperature region above \( T_c \). The interpolating formula between 3D and 2D cases can be written following the discussion by Takahashi \[14\].

\[ -\text{Im}\Sigma(k_F, 0) \approx \frac{T \phi^2_{k_F}}{16\pi v_F \xi^2} \log(1 + \frac{\xi q_c)^2}{M}), \]

\[ + \frac{T \phi^2_{k_F}}{2\pi v_F \xi \sqrt{M}} \tan^{-1} \left( \frac{\xi q_c}{\sqrt{M}} \right) - \tan^{-1} \left( \frac{\xi q_c}{\sqrt{M}} \right). \]  

Here, \( \epsilon \approx t_z/t \) is the parameter which characterizes the degree of the anisotropy.

We also calculated the one-particle spectrum \( (A(k, \omega) = -\text{Im}G(k, \omega)/\pi) \), and it is found that for the small inter-plane transfer \( t_z \) the spectrum has a pseudogap structure around \( \omega = 0 \). The relationship between \( t_z \) and the temperature \( (T_{\text{PS}}) \) at which the pseudogap opens at point \( A \), is shown in Fig. 3. \( T_{\text{PS}} \) is obtained in a manner such that the one-particle spectrum is calculated at intervals of 0.001 at a temperature which is less than one percent of \( T_c \), and \( T_{\text{PS}} \) is the temperature at which it is found to be suppressed around \( \omega = 0 \). It is seen that for the small

\[ \text{Im}\Sigma(k_F, 0) \propto \sqrt{T_F}(\frac{T}{T_F})^2 \frac{1}{\sqrt{M}} \]  

Here \( T_F \) is the Fermi degeneracy temperature. For large \( g, T_c \) is high and therefore the enhancement factor \( (T/T_F)^2 \) is large. Therefore, in the case of high \( T_c \), the damping effect is more enhanced even for rather large values of \( M \) than in the case of low \( T_c \). In other words the thermally broadened spectrum is more sensitive to the superconducting fluctuation.

This situation is considered to be realized because the superconductivity of \( \kappa-(ET)_2 X \) appears only near the Mott-insulating phase and it is likely that the \( T_F \) is very low owing to the strong electron correlation. From the above discussion, we propose that point \( A \) and its symmetry-related points of the Fermi surface must be destructed in this pseudogap region due to the strong fluctuation of the superconductivity. This will be checked by the ARPES experiments. The FS and the one-particle spectrum at point \( A \) are shown in Fig. 5. In Fig. 5(a), the FS of the single-band in our model is shown. The equivalence of this single band model and the two-band model (the hopping parameter \( t \) takes two kinds of values) is discussed in ref. 8. From Fig. 5(b), it can be seen that the one-particle spectrum has a pseudogap structure above \( T_c \).
Fig. 5. (a) The Fermi surface for \( t_z = 0.00 \). The meaning of point \( A \) is the same as above. (b) The temperature dependence of the one-particle spectrum. The values of \( g \) and \( t_z \) are shown in the figure. In this case \( T_c \) is 0.339.

This behavior disappears at the points of the FS away from point \( A \) and its symmetry-related points because of the \( d_{x^2-y^2} \)-wave symmetry of the attractive force.

Finally, we comment on the two factors which are not included in our discussion but which are required to be considered for an accurate quantitative description. One, is that we do not take into account the effect of the fluctuation in the determination of \( T_c \), i.e. the self-consistency requirement. For a quasi 2D system such as \( \kappa-(ET)_{2}X \), the fluctuation plays an essential role in determining \( T_c \). However, our conclusion that the pseudogap is peculiar to the quasi 2D system is not affected because our argument is basing on the mass term to the quasi 2D system is not affected because our argument is basing on the mass term \( M \) which measures the extent to which the system is away from the superconducting state. The other factor is the effect of the Coulomb repulsion. This is also important for obtaining accurate quantitative results in the system which neighbors the Mott insulating phase. However, if the adiabatic continuity is realized, the effect of the Coulomb repulsion is only the rescaling of various quantities, such as the Fermi degeneracy temperature. Justification of the adiabatic continuity in the metallic phase near the Mott-insulating phase is an important problem \[1\,13\] and will be discussed elsewhere.

In summary, we have calculated the electronic properties of the quasi 2D model by considering the superconducting fluctuation. The relationship between the DOS and the damping rate is shown and it is found that the decrease of the DOS is caused by the large damping rate which is enhanced by the superconducting fluctuation in the quasi 2D case. We have also calculated the one-particle spectrum, and the pseudogap behavior over a wide temperature range above \( T_c \) is obtained only for the quasi 2D case. It is also found that this behavior is characteristic of a rather incoherent metal which has a broad spectrum around the Fermi level and is sensitive to the fluctuation. These conditions for the appearance of the pseudogap can be applied to \( \kappa-(ET)_{2}X \). Therefore, the anomalous properties found by the NMR experiments are considered to be caused by the superconducting fluctuation. The strong two-dimensionality and the incoherent metallic properties are also the properties of cuprates in the under-doped region, so our argument can be applied to this material and supports the expectation that the anomalous behaviors of the cuprates and \( \kappa-(ET)_{2}X \) have the same origin.\[8\,10\] ARPES experiments are desired not only to check the validity of our proposal, but also to understand the property of pseudogap in \( \kappa-(ET)_{2}X \) more precisely.

Numerical computation in this work was carried out at the Yukawa Institute Computer Facility.

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\( \text{Re}\Sigma(k_F, \omega) \) vs \( \omega \) for different values of \( T \) and \( tz \):

- \( T=0.350 \) (tz=0.10)
- \( T=0.400 \) (tz=0.10)
- \( T=0.200 \) (tz=1.00)
- \( T=0.250 \) (tz=1.00)

\( g=2.00 \)
\[ \Im \Sigma(k_F, \omega) \]

- \( T = 0.350 \) (\( tz = 0.10 \))
- \( T = 0.400 \) (\( tz = 0.10 \))
- \( T = 0.200 \) (\( tz = 1.00 \))
- \( T = 0.250 \) (\( tz = 1.00 \))

\( g = 2.00 \)
$g = 2.00$

- $tz = 0.10$ (DOS)
- $tz = 1.00$ (DOS)
- $tz = 0.10$ (ise)
- $tz = 1.00$ (ise)
\[ \frac{(T_{pg} - T_c)}{T_c} \]

\[ g = 2.00 \]
$M = (T_{pg} - T_c)/T_c$

$t_z = 0.010$
(-π, π) 
(-π, 0) 
(-π, -π) 
(0, -π) 
(π, -π)
$A(k_F, \omega)$

- $T=0.520$ ($M=0.123$)
- $T=0.462$ ($M=0.090$)
- $T=0.390$ ($M=0.042$)
- $T=0.350$ ($M=0.010$)

$g=2.00$
$t_z=0.010$

(b)