Pseudogap formed from preformed pairs in the organic superconductors κ-(BEDT-TTF)$_2X$

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We study the pseudogap phenomenon of layered organic compounds κ-(BEDT-TTF)$_2X$ based on the scenario of preformed pairs. By solving self-consistently the closed equations of the T-matrix in the "pair approximation" scheme, we calculate the spectral function of electrons in a two-dimensional anisotropic triangular model at half-filling for $d_{x^2-y^2}$- and d_{xy}-wave attractive potentials, respectively. We find that a pseudogap occurs in the intermediate coupling region in the normal state and it opens up highly anisotropically in the momentum space. In particular, the momentum region where it appear first is quite different for both attractive potentials.

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I. INTRODUCTION

Recently, the mechanism for unconventional superconductivity is of considerable interest in condensed matter physics. The layered organic compounds κ-(ET)$_2X$, where ET represents the planar BEDT-TTF molecule and X anions, are examples of quasi-two-dimensional unconventional superconductors.[1] They have many similarities to high-T_c superconductors,[2] such as the proximity of superconductivity to a Mott insulator in the phase diagram.

Above the superconducting (SC) transition temperature (T_c), the normal metallic state of the underdoped high-T_c cuprates behaves in a way quite different from that we have encountered before. Especially, a suppression in the density of state has been observed or inferred in the measurements on some properties and it has been referred to as the pseudogap.[3] The pseudogap phenomenon is observed firstly in the NMR experiments, which manifests itself as a decrease in the Knight shift and the spin-lattice relaxation rate per temperature $1/T_1T$ (T the temperature) below the pseudogap opening temperature T^*.[4] Interestingly, κ-(ET)$_2X$ shows the similar anomalous properties.[5–8] As found by NMR experiments,[5–8] $1/T_1T$ increases with decreasing temperature below 150 K, which is a result of enhanced antiferromagnetic fluctuations. But it decreases with further decreasing temperature below $T\sim 50$ K. So, a similar pseudogap phenomenon has also been inferred from these experiments.[2,9]

The pseudogap phenomenon of high-T_c superconductors has been studied intensively and been taken as an important clue to the physics governing the high-T_c cuprates,[10] but little has been done theoretically on the similar phenomenon in layered organic superconductors. Recently, Jujo *et al.* have proposed a possible mechanism for the pseudogap in organic superconductor based on the SC fluctuation.[11] They used the one-loop approximation for the SC fluctuation and take the $d_{x^2-y^2}$-wave interaction as the attractive potential. However, the pairing symmetry in κ-(ET)$_2X$ is in fact still in debate both experimentally[8,12,13] and theoretically,[14–16] namely, whether it is a $d_{x^2-y^2}$-wave or a d_{xy}-wave pairing symmetry is controversial. On the other hand, for a strong coupling superconductivity, a feedback effect of the SC fluctuation should be considered. Therefore, in this paper we will study the pseudogap phenomena based on the "pairing approximation" scheme of the T-matrix formalism developed by Kadanoff and Martin.[17] In our studies, a self-consistent calculation based on the full closed equations is carried out, compared with the lowest-order approximation used by Jujo *et al.*[11] In this way, the feedback effect is expected to be included, and the BCS to Bose-Einstein crossover in the strong coupling limit can be tracked. This method has been used to discuss the pseudogap in high-T_c cuprates.[18,19]

We find that a pseudogap appears below the temperature T^*, which is identified as the temperature when the single peak of the electron spectral function splits into two peaks, and T^* is well above the SC transition temperature T_c which is determined by the Thouless criterion for superconductivity.[20] The dependence of the opening temperature of the pseudogap T^* on the coupling strength and the ratio of the next-nearest to the nearest hopping integral t'/t is presented. It suggests that the pseudogap is a result of the occurrence of preformed pairs in the intermediate coupling region between the BCS limit and Boson-Einstein limit. Its opening temperature increases with a reduced ratio t'/t. We also compare the pesudogap behavior for the $d_{x^2-y^2}$- and the d_{xy}-wave attractive interactions, and find that the magnitude and the opening temperature of the pseudogap for the d_{xy}-wave attractive interaction is smaller than those for the $d_{x^2-y^2}$-wave interaction. In particular, we find that the momentum region where the pseudogap opens up first is quite different for different attractive interactions.

The paper is organized as follows. In Sec. II, we will present the self-consistent T-matrix method. In Sec. III, we will carry out the numerical calculations of the single-particle spectral function and discuss the results. Finally a brief conclusion will be given in Sec. IV.

II. MODEL AND CALCULATION

We consider a two-dimensional model Hamiltonian with an attractive potential,

$$H = \sum_{k,\sigma} \epsilon_k C^+_{k,\sigma} C_{k,\sigma} + \sum_{k,k',q} V_{k,k'} C^+_{k+q/2,\uparrow} C^+_{-k+q/2,\downarrow} C_{-k'+q/2,\downarrow} C_{k'+q/2,\uparrow}, \quad (1)$$

where a separate interaction is assumed,

$$V_{k,k'} = g\varphi(k)\varphi(k'), \quad (2)$$

with $g(<0)$ the coupling strength, $\varphi(k)=\cos k_x - \cos k_y$ for the $d_{x^2-y^2}$ wave and $\varphi(k)=2\sin k_x \sin k_y$ for the d_{xy} wave. According to the arrangement of BEDT-TTF molecules in the conduction layer of κ-$(ET)_2X$[1] and the energy band calculation, Kino and Fukuyama[21] proposed a dimer model to describe its physics, in which a pair of ET molecules is considered as the basic structural unit of the conduction layer. Based on this simplest effective model, one can regard the conduction layer of κ-$(ET)_2X$ as the anisotropic triangular lattice with one site in the unit cell. It in fact corresponds to a square lattice with the transfer integral $-t$ between the nearest-neighbor sites and $-t'$ between a half of the next-nearest-neighbor sites along only [for example, along (1, 1) direction] one direction. So, its tight-binding energy band including the nearest- and the next-nearest-neighbor hopping terms is given by

$$\epsilon_k = -2t(\cos k_x + \cos k_y) - 2t' \cos(k_x + k_y) - \mu, \quad (3)$$

in which, μ is the chemical potential which is an input parameter and is adjusted to obtain the required density of electrons $n=1$. We note that the in-plane anisotropy is manifested in the term $\cos(k_x+k_y)$ of Eq. (3). For an isotropic energy band, this next-nearest-neighbor term should be $\cos(k_x)\cos(k_y)$. We can see that Eq. (3) is not symmetric under the exchange of k_x to $-k_x$, but the isotropic band is symmetric under this exchange. The Fermi surface calculated using Eq. (3) for $n=1$ is shown in the inset of Fig. 4.

According to the "pairing approximation" scheme,[18] the self-consistent T-matrix is given by[19]

$$T^{-1}(q,i\omega_n) = g^{-1} + \frac{1}{\beta N}\sum_{p,\xi_l} \varphi^2(p) G(p,i\xi_l) G^{(0)} \times (q-p, i\omega_n - i\xi_l) \quad (4)$$

and the self-energy of electrons is given by

$$\Sigma(k,i\xi_l) = \frac{1}{\beta N}\sum_{q,\omega_n} \varphi^2(k-q/2) T(q,i\omega_n) G^{(0)} \times (q-k, i\omega_n - i\xi_l). \quad (5)$$

The corresponding Green's function satisfies the Dyson equation,

$$G^{-1}(k,i\xi_l) = G^{(0)^{-1}}(k,i\xi_l) - \Sigma(k,i\xi_l), \quad (6)$$

where $G^{(0)}(k,i\xi_l)=(i\xi_l-\epsilon_k)^{-1}$ and $\beta^{-1}=T$. Equations (4)–(6) constitute a closed system of self-consistent equations. This set of equations will be solved iteratively. Because it is very difficult to get the convergence of G and T at all k points and Matsubara frequencies at the same time, we will take the density of electrons n as the criterion of convergence as we have done before,[19] n is given by

$$n = \frac{2}{\beta N}\sum_{k,\xi_l} G(k,i\xi_l) e^{i\xi_l 0^+}. \quad (7)$$

In the numerical calculations, the renormalized Green's function G is calculated as a function of complex (Matsubara) frequencies $i\xi_l$. However, the spectral function of electrons is related to the retarded Green's function as follows:

$$A(k,\omega) = -\frac{1}{\pi}\,\mathrm{Im}[G(k,i\xi_l \to \omega + i0^+)]. \quad (8)$$

Therefore, an analytic continuation to the real frequency is needed. We use the Padé approximants[22] to carry out the analytic continuation. The reliability of the analytic continuation using the Pade approximant was discussed extensively by Viderg and Serene[22] and also checked by us by comparing the spectral function of a quasiparticle Green's function and the spin susceptibility of a d-wave superconductor with and without using the Pade approximant. A fairly positive result has been obtained. The SC transition temperature T_c is obtained according to the Thouless criterion for a SC pairing instability,[20]

$$T^{-1}(q=0,\omega=0) = 0. \quad (9)$$

In the following numerical calculations, the summations over momentum $p(q)$ and Matsubara frequency $\xi_l(\omega_n)$ are preformed by dividing the momentum space into 32×32 lattices and by summing $\omega_n(\xi_l)$ from -128 to 128 in $\omega_n = 2n\pi T$ and $\xi_l=(2l+1)\pi T$, respectively. The stability of the results with respect to the number of the Matsubara frequencies and the dividing grids of the Brillouin zone has been checked and no qualitative difference has been found as will be discussed in the following section. The energy is in units of $2t$. In most of the paper, t'/t will be chosen to be 0.7,[21] except when we discuss the dependence of T_c and T^* on t'/t.

III. NUMERICAL RESULTS

A. Case of the $d_{x^2-y^2}$ wave

First, let us discuss the results with a $d_{x^2-y^2}$-wave attractive potential. In Fig. 1, the ω dependence of the electron spectral function $A(\mathbf{k},\omega)$ at $\mathbf{k}=(0,15\pi/16)$ for different temperatures $T=0.2, 0.18, 0.15$ is presented, with the coupling strength $g=-1.0$ which is in the intermediate coupling region as described below. At $T=0.2$, a single peak appears in the spectral function of electrons, which is a typical feature in the normal Fermi liquid state. When the temperature is decreased to $T=0.18$, a suppression of the spectral weight occurs near $\omega=0$, the Fermi energy. As a result, the single peak in $A(\mathbf{k},\omega)$ is split into two peaks. When decreasing the temperature further, one will find that the suppression becomes more and more obvious. The SC transition temperature T_c determined from the Thouless criterion Eq. (9) is 0.063, which is nearly one-third of 0.18. So, this suppression in the spectral weight is in the normal state and indicates the appearance of the pseudogap. We find that the opening tem-

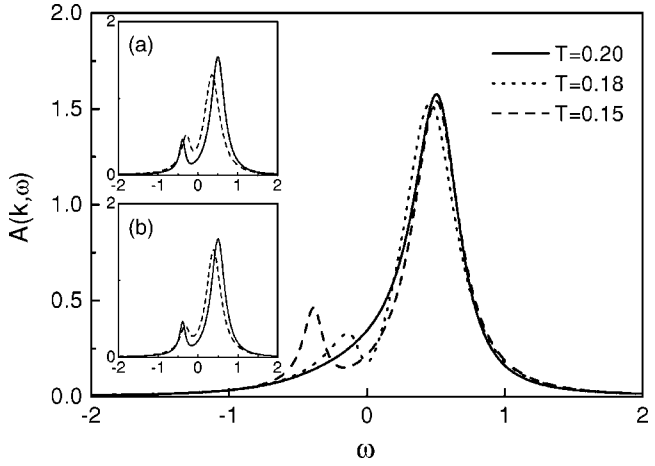

FIG. 1. Spectral function of electrons $A(\mathbf{k},\omega)$ vs ω (measured relative to the chemical potential) at different temperatures for the $d_{x^2-y^2}$-wave attractive potential with $\mathbf{k}=(0,15\pi/16)$ and the coupling strength $g=-1.0$. Inset (a), a comparison of the result at $T=0.15$ calculated by dividing the k space into 32×32 lattices (solid line) with that using 64×64 lattices (dashed line). The frequency summation is done over 256 Matsubara frequencies. Inset (b), a comparison of the result at $T=0.15$ calculated by summing over 256 Matsubara frequencies (solid line) with that using 512 Matsubara frequencies (dashed line). The k space is divided into 32×32 lattices.

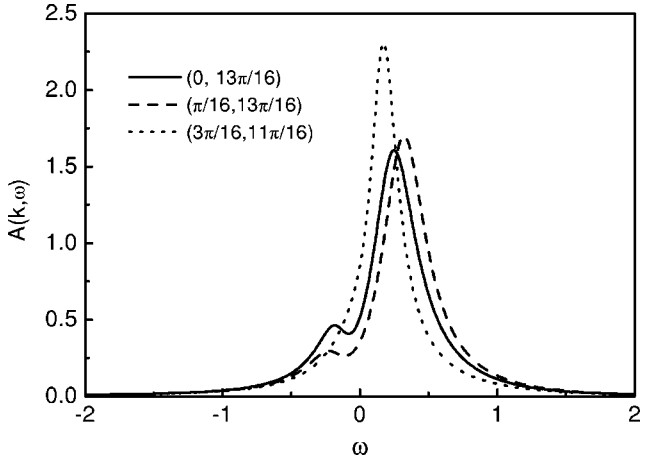

FIG. 2. Spectral function of electrons $A(\mathbf{k},\omega)$ vs ω (measured relative to the chemical potential) at different momenta for the $d_{x^2-y^2}$-wave attractive potential with $T=0.18$ and $g=-1.0$.

perature of the pesudogap T^*, which is identified as the starting temperature when a suppression of the spectral weight occurs, is approximately equal to 0.20.

The above calculations are carried out making use of 32×32 momentum grids in the Brillouin zone and 256 Matsubara frequencies. To check the stability of the results with respect to the number of the dividing grids and the Matsubara frequencies, we have repeated the calculations making use of 64×64 momentum grids and 512 Matsubara frequencies. The obtained results are shown in the inset (a) and (b) of Fig. 1 as dashed lines, respectively. The previous result is also shown as solid lines to make a comparison. One can see that there is no qualitative difference between them, though the magnitude of the pseudogap becomes smaller when more momentum grids or Matsubara frequencies are used.

To see the possible anisotropic distribution of the pseudogap, we show in Fig. 2 the spectral function for different k points near the Fermi surface with $g=-1.0$ at $T=0.18$. The positions of these k points in the Brillouin zone are represented by open circles in the inset of Fig. 4 (The three circles at first from the $k_x=0$ to the diagonal direction along the Fermi surface). It shows that the gaplike structure can be observed clearly around $(0,13\pi/16)$, but disappears gradually as the momentum moves away from it to the diagonal direction of the Brillouin zone. It means that the pseudogap opens up at different points in the momentum space. It appears first at the momentum where the attractive potential is maximum and gradualy opens up at other momenta along the Fermi surface, with the decrease of temperature. As a result, it will lead to the destruction of the Fermi surface in the normal state. This is different from the opening of the SC gap, in that case the gap opens up in all momenta along the Fermi surface simultaneously. This behavior is believed to arise from the $d_{x^2-y^2}$-wave form of the attractive potential. The similar anisotropic opening of the pseudogap in high-T_c superconductors has been observed by angle-resolved photoemission spectroscopy (ARPES) experiments,[23,24] so we propose to identify it by using the ARPES experiment.

In the weak coupling limit, the superconductivity occurs simultaneously with the pairing, this corresponds to the BCS regime. However, with the increase of the coupling strength, the chemical potential shifts downwards due to the strong renormalization of the Green's function of quasiparticles.[19,20] As a result, the creation of bosonic particles due to the preformed pairs of quasiparticles starts at certain temperature T^* which is above the SC transition temperature T_c. These bosonic particles are not infinitely long lived, because they can be damped by other nonpaired fermions. We think that it is the preformed pair that leads to the appearance of the pseudogap. The superconductivity happens when these preformed bosons become coherent via the Bose-Einstein condensation at T_c. In the very large coupling limit, the chemical potential will eventually shift down to the bottom of the fermionic energy band. Then, the Fermi surface is gone and the preformed pairs become bosons with infinite lifetime. This corresponds to an ideal gas Bose-Einstein asymptote.[20] Thus, the scheme we used here can lead to an interpolation between the BCS limit for small g and the Bose-Einstein condensation in the strong coupling (large g) limit. In view of this, the opening temperature of the pseudogap will decrease with the decrease of the coupling strength, and eventually disappears when the coupling strength is decreased to the weak coupling region. This is indeed obtained in our numerical calculations. In Fig. 3, we show the spectral function for several coupling constants g from -1.0 to -0.9, at the momentum $k=(0,15\pi/16)$ where the gap magnitude is maximum and the temperature $T=0.18$. One sees that the pseudogap weakens gradually with the decrease of g and closes up when the coupling strength reaches $g=-0.9$. So, the opening temperature T^* for the coupling $g=-0.9$ should be lower than $T=0.18$. When reducing the coupling strength

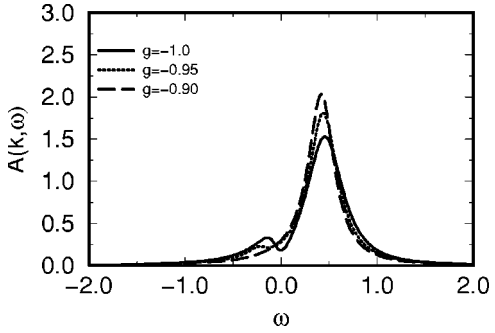

FIG. 3. Spectral function of electrons $A(\mathbf{k},\omega)$ vs ω (measured relative to the chemical potential) for the $d_{x^2-y^2}$-wave attractive potential with $T=0.18$ and $\mathbf{k}=(0,15\pi/16)$. The solid, dashed, and dotted lines are results for various coupling strengths.

further to the weak coupling region, one will find no pseudogap in the normal state.

Next, we discuss the dependence of the opening temperature T^* of the pseudogap on the ratio t'/t. For the κ-$(\mathrm{ET})_2X$ with a SC ground state, $t'/t=0.7$ is usually assumed.[21] However, it is shown[21,25] that this ratio may vary with the change of the anion X and the external pressure. In view of this, we present in Fig. 4 the opening temperature (T^*) (square points) of the pseudogap and the SC transition temperature (T_c) (triangle points) obtained using the Thouless criterion Eq. (9) for different ratios of t'/t, with $g=-1.0$ and at the momentum $\mathbf{k}=(0,15\pi/16)$. One can see that T^* increases with the decrease of t'/t. Meanwhile, T_c is several times smaller than T^* and shows a relatively flat dependence on t'/t. With the decrease of t'/t, the unperturbed Fermi surface obtained using Eq. (3) displays more and more strong nesting properties. The nesting of the Fermi surface will cause an enhanced density of states, therefore leading to higher pesudogap opening temperature.

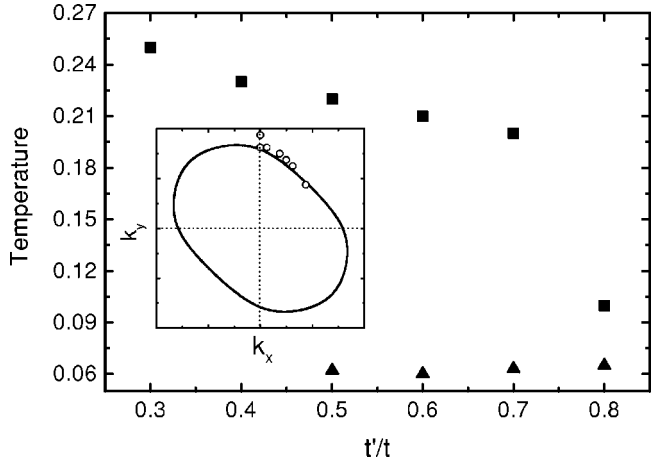

FIG. 4. Opening temperature of the pseudogap T^* (square) and the superconducting transition temperature T_c (triangle) vs t'/t, for the $d_{x^2-y^2}$-wave attractive potential with $\mathbf{k}=(0,15\pi/16)$ and the coupling strength $g=-1.0$. Inset, the bare Fermi surface (solid line). The open circles indicate the position of the k points at which our results are presented in Figs. 1–3 and 5–7, respectively.

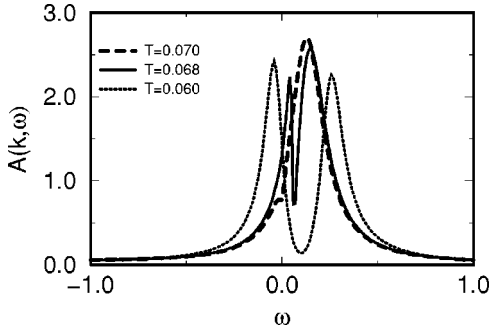

FIG. 5. Spectral function of electrons $A(\mathbf{k},\omega)$ vs ω (measured relative to the chemical potential) at different temperatures for the d_{xy}-wave attractive potential with $\mathbf{k}=(7\pi/16,7\pi/16)$ and the coupling strength $g=-1.0$.

B. Case of the d_{xy} wave

As noted in the introduction, whether the pairing symmetry in κ-$(\mathrm{ET})_2X$ is a $d_{x^2-y^2}$ wave or a d_{xy} wave is still controversial.[8,12–16] Therefore, we will discuss the possible pseudogap behavior for the d_{xy} wave in this section. Figure 5 shows the spectral functions for different temperatures with $\mathbf{k}=(7\pi/16,7\pi/16)$ and $g=-1.0$. One can see that, below the opening temperature $T^*=0.07$, a suppression of the spectral weight occurs and it leads to the double-peak structure, which suggests the opening of a pseudogap as discussed in Sec. III A. The suppression becomes more and more severe with the decrease of temperature, which is similar to the behavior observed for the $d_{x^2-y^2}$ wave as shown in Fig. 1. However, compared with the case for the $d_{x^2-y^2}$ wave, the opening temperature of the pseudogap is much lower and its magnitude is smaller.

In Fig. 6, we show the spectral function with $T=0.06$ and $g=-1.0$ for three different momenta near the Fermi surface. The positions of these k points in the Brillouin zone are represented by open circles in the inset of Fig. 4 (the three circles at first from the diagonal direction to $k_x=0$ along the Fermi surface). The same as the case of the $d_{x^2-y^2}$-wave interaction, the pseudogap appears highly anisotropically in the momentum space. However, the pseudogap opens up first around the momenta where the d_{xy}-wave interaction has maximum magnitude, i.e., that portion of the Fermi surface

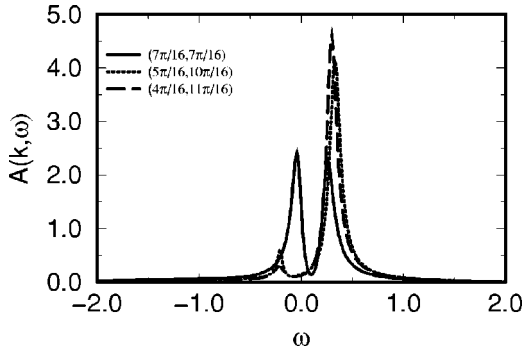

FIG. 6. Spectral function of electrons $A(\mathbf{k},\omega)$ vs ω (measured relative to the chemical potential) at different momenta for the d_{xy}-wave attractive potential with $T=0.06$ and $g=-1.0$.

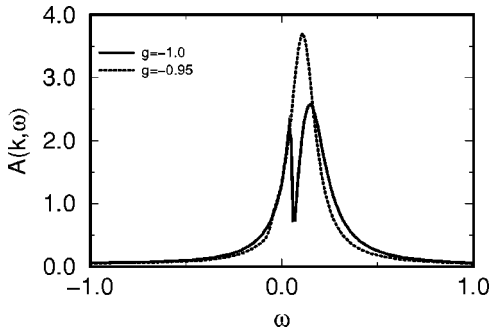

FIG. 7. Spectral function of electrons $A(\mathbf{k},\omega)$ vs ω (measured relative to the chemical potential) for the d_{xy}-wave attractive potential with $T=0.068$ and $k=(7\pi/16,7\pi/16)$. The solid and dashed lines are results for coupling strength $g=-1.0$ and $g=-0.95$, respectively.

along the diagonal direction, and gradually disappears when the momentum moves towards $(0,\pi)$ where the pseudogap for the $d_{x^2-y^2}$-wave interaction occurs first. This feature is quite different for both interaction potentials. In fact, this angular distribution of the opening of the pseudogap shares the similar momentum dependence with the magnitude of the SC gap and may suggest that the pseudogap is of the same symmetry with its SC gap. Finally, the result presented in Fig. 7 shows again that the pseudogap will close up with the decrease of the coupling strength.

From above comparison, one may expect that the $d_{x^2-y^2}$ wave is more stable than the d_{xy} wave based on the model Eq. (1), because the opening temperature of the pseudogap and consequently the SC transition temperature in the former is higher than the latter. But, we note that our model is in fact based on a rather general ground. The origin of the attractive potential and therefore the coupling strength for a possible $d_{x^2-y^2}$- and a d_{xy}-wave symmetry may be different. Therefore, we cannot determine which one is more stable here. However, the results presented here indicate that the pseudogap opens up at different points in the momentum space for both attractive potentials. This property may be taken as a possible way to probe the symmetry of the pseudogap and consequently the SC gap in κ-$(\mathrm{ET})_2X$.

IV. CONCLUSION

In summary, we have calculated the spectral function of electrons based on a two-dimensional anisotropic triangular model at half-filling for the $d_{x^2-y^2}$ and the d_{xy} attractive potentials, respectively, by solving self-consistently the closed equations of the T-matrix in the "pair approximation" scheme. We find that a pseudogap occurs in the intermediate coupling region below the opening temperature T^*, which is above the superconducting transition temperature determined by the Thouless criterion. It is ascribed to arising from the preformed pairs in the intermediate coupling region between the BCS limit and Boson-Einstein limit. We have studied the angular dependence of the pseudogap and find that the pseudogap opens up highly anisotropically in the momentum space. In particular, the momentum region at which it appears first is quite different for the $d_{x^2-y^2}$- and the d_{xy}-wave attractive potentials. The model studied is suggested to be related to the layered organic compounds κ-$(\mathrm{ET})_2X$, therefore our calculation provides a possible origin of the pseudogap behavior in this material.

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