Theory of the Hall Coefficient and the Resistivity on the Layered Organic Superconductors $\kappa$-(BEDT-TTF)$_2$X

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In the organic superconducting $\kappa$-(BEDT-TTF)$_2$X compounds, various transport phenomena exhibit striking non-Fermi liquid behaviors, which should be the important clues to understanding the electronic state of this system. Especially, the Hall coefficient ($R_H$) shows Curie-Weiss type temperature dependence, which is similar to that of high-$T_c$ cuprates. In this paper, we study a Hubbard model on an anisotropic triangular lattice at half filling, which is an effective model of $\kappa$-(BEDT-TTF)$_2$X compounds. Based on the fluctuation-exchange (FLEX) approximation, we calculate the resistivity ($\rho$) and $R_H$ by taking account of the vertex corrections for the current, which is necessary for satisfying the conservation laws. Our theoretical results $R_H \propto T^{-1}$ and $\cot \theta_H \propto T^2$ explain the experimental behaviors well, which are unable to be reproduced by the conventional Boltzmann transport approximation. Moreover, we extend the standard Eliashberg’s transport theory and derive the more precise formula for the conductivity, which becomes important at higher temperatures.

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I. INTRODUCTION

It is well-known that the superconducting organic compound $\kappa$-(BEDT-TTF)$_2$X systems exhibit rich variety of ground states, through the strong correlation effects between electrons. For example, X=Cu[N(CN)$_2$]Cl salt is in the antiferromagnetic (AF) insulating phase in a low pressure region, $P < 200$bar. With increasing the pressure, it changes to the superconducting (SC) phase at the transition temperature $T_c = 13$K through the weak first-order transition, and the SC phase disappears under $P > 10$kbar. Above $T_c$, $1/T_cT \propto T^{-1}$ is observed for a wider range of temperatures, which reflects the growth of the AF fluctuations as the temperature decreases. Thus, it is natural to consider that the AF fluctuation is the origin of superconductivity.

Recently, several theoretical works on $\kappa$-(BEDT-TTF)$_2$X system were done by using the fluctuation-exchange (FLEX) approximation, which is a kind of self-consistent spin-fluctuation theory. They showed that the $d$-wave like superconductivity is induced by the strong AF fluctuations. Moreover, characteristic features of the experimental pressure-temperature phase diagram were reproduced well.

In this system, various transport phenomena above $T_c$ also show interesting non-Fermi liquid behaviors. Recently, the temperature dependence of the resistivity ($\rho$) and the Hall coefficient ($R_H$) for X=Cu[N(CN)$_2$]Cl were measured precisely above $T_c$ under $P = 4.5 \sim 10$kbar in Ref.2. According to the measurement, the approximate relations $\rho \propto T$ and $R_H \propto T^{-1}$ are observed for $T = 30 \sim 100$K, and the Hall angle $\cot \theta_H = (\Delta \sigma_{xx}/\Delta \sigma_{xy})$ is proportional to $T^2$ well. In other measurement on X=Cu[NCS]$_2$, $R_H$ increases by a factor of three on cooling below 60K at ambient pressure ($T_c = 10$K). The mechanism of these interesting non-Fermi liquid behaviors, which are also observed in high-$T_c$ cuprates, should be understood consistently.

In this paper, we present the theoretical study on both $\rho$ and $R_H$ for $\kappa$-(BEDT-TTF)$_2$X by using the FLEX approximation. Based on the conserving approximation, all the vertex corrections (VC’s) for the current which is necessary to satisfy conserving laws are taken into account. We find that the Curie-Weiss-like behavior of $R_H$ is naturally reproduced by the VC’s when the AF fluctuations are dominant. On the other hand, the conventional Boltzmann approximation, which does not include any VC’s, fails to reproduce the temperature dependence of $R_H$. Experimentally, an intimate relation between the AF fluctuations and the transport phenomena is recognized.

Note that the effect of VC’s in nearly AF Fermi liquid was first studied in high-$T_c$ cuprates by refs.3,4 and the overall behavior of $R_H$ are naturally reproduced both for hole-doped compounds and for electron-doped compounds. The present study is based on them basically.
II. ELECTRONIC STATES GIVEN BY THE FLEX APPROXIMATION

We study the triangular lattice Hubbard model with anisotropic hopping parameters \( (t, t') \) as shown in the inset of Fig. 6, which is a simple effective model for \( \kappa \)-(BEDT-TTF)\(_2\)X system. The dispersion is given by

\[
e_k^0 = 2t(\cos(k_x) + \cos(k_y)) + 2t' \cos(k_x + k_y),
\]

where we put the lattice spacing 1. We analyze this model by using the FLEX method, which is a kind of self-consistent perturbation theory. This method had been applied to the study of high-\( T_c \) cuprates, and various non-Fermi liquid behaviors were reproduced well. It has also been applied to the superconductor ladder compound, \( \text{Sr}_{114-x}\text{Ca}_x\text{Cu}_{22}\text{O}_{4+\delta} \).

![FIG. 1. The Fermi surface of the present model \((t'/t = 0.7)\) for \( U = 9 \) (full line) and \( U = 0 \) (broken line) determined by \( \text{Re}(G_k^{-1}(0)) = 0 \). The long-dashed lines, connecting between X and Y for example, represent the AF-zone boundary. The hot spots locate on the AF-zone boundary. Inset: the lattice structure.](image1)

In the present study we put \( t = 1 \) and \( t' = 0.7 \), where a \( d \)-wave like superconductivity is realized at \( T_c > 0.02 \) for \( U \geq 7 \), and it is replaced by the AF phase for \( U \geq 10 \). Figure 6 shows the Fermi surfaces at half-filling for \( U = 0 \) and \( U = 9.0 \) at \( T = 0.02 \). The Fermi surface is hole-like because \( t, t' < 0 \). There are two reflection symmetries with respect to the \((\theta = \pi/4)\)-axis and the \((\theta = 3\pi/4)\)-axis in Fig. 6. We see that the nesting of the Fermi surface is strengthened by the deformation of the Fermi surface in the case of finite \( U \), which is caused by the real part of the self-energy.

The self-energy in the FLEX approximation is given by

\[
\Sigma_k(\epsilon_n) = T \sum_{q,l} G_{k-q}(\epsilon_n - \omega_l) \cdot V_q(\omega_l),
\]

\[
V_q(\omega_l) = U^2 \left( \frac{3}{2} \chi_q^s(\omega_l) + \frac{1}{2} \chi_q^s(\omega_l) - \chi_q^0(\omega_l) \right),
\]

\[
\chi_q^c(\omega_l) = \chi_q^0(\omega_l) \cdot \left( 1 - (+)U \chi_q^0(\omega_l) \right)^{-1},
\]

\[
\chi_q^0(\omega_l) = -T \sum_{k,n} G_{q+k}(\omega_l + \epsilon_n) G_k(\epsilon_n),
\]

where \( \epsilon_n = (2n + 1)\pi T, \omega_l = 2l \cdot \pi T \), respectively. By noticing the Dyson equation \( \{ G_k(\epsilon_n) \}^{-1} = \epsilon_n + \mu - \epsilon_k^0 - \Sigma_k(\epsilon_n) \), we solve the eqs. (2) \( \sim \) (5) self-consistently, choosing the chemical potential \( \mu \) so as to keep the system at half-filling. Here we use 4096 \( k \)-meshes and 256-Matsubara frequencies, respectively.

Here, \( \chi_q^s(0) \) gives the static spin susceptibility. Figure 2 shows the Curie-Weiss behavior of the maximum value of \( \chi_q^s(0) \), which is proportional to the square of the AF-correlation length \( \xi_{AF} \). The obtained relation \( \xi_{AF}^2 \propto T^{-1} \) is known to be caused by the renormalization of the self-energy. The FLEX approximation also gives the relation \( (T/T_0)^{-1} = \sum_k \text{Im} \chi_k^s(0)/\omega \propto T^{-1} \).

![FIG. 2. The maximum value of \( \chi_q^s(0) \) \((\propto \xi_{AF}^2)\) for \( U = 6 \sim 10 \).](image2)

 Nonetheless \( \chi_q^s(0) \) for \( U = 0 \) is incommensurate, it becomes commensurate in the case of \( U \geq 8 \) at \( T = 0.02 \), which is consistent with experiments. This change of the shape of \( \chi_q^s(0) \) is brought by the deformation of the interacting Fermi surface as shown in Fig. 6, which can not be reproduced by the simple renormalization of \( t'/t \).

We also note that the obtained \( \chi_q^s(0) \) will be slightly overestimated at low temperatures because its VC's are neglected here.

Next, Fig. 3 (a) shows the imaginary part of the self-energy, \( \gamma_k = \text{Im} \Sigma_k(-\text{i}0) > 0 \), along the Fermi surface for the region \( \pi/4 \leq \theta \leq 3\pi/4 \) for \( T = 0.02, 0.04, \ldots, 0.1 \).
The definition of the hot spots and the cold spots are given in Fig. 1. In this temperature region, $\gamma_k$ takes the minimum (maximum) value at the cold spot (hot spot). A hot spot is separated from its counterpart by $Q = (\pi, \pi)$ in the reciprocal space, and a cold spot is the most distant point from the AF-zone boundary. In the present study, the relations $\gamma_{\text{cold}} \propto T$ and $\gamma_{\text{hot}} \propto \sqrt{T}$ are satisfied because $\xi_{AF} \lesssim k^{-1}$ in the FLEX approximation, where $\Delta k$ is explained in Fig. 1. Note that the damping rate of the quasiparticle is given by

$$\gamma = \frac{\Delta k}{\xi_{AF}},$$

where $k_0 \equiv (1 - \frac{\partial}{\partial \omega} \text{Re} \Sigma_k(\omega))^{-1}$ is the renormalization factor. In the numerical calculation for $U = 9$, $\gamma_k \ll T$ around the cold spots at lower temperatures because $\gamma_k \ll 10$ there. Thus, quasiparticle can still be defined.

### A. Derivation of the Total Current $\mathbf{J}_k(\omega)$ Based on the Conservation Approximation

In this section, we calculate both $\sigma_{xx}$ and $\Delta \sigma_{xy}$ based on the conserving approximation. By using the Kubo formula, they are derived as

$$\sigma_{xx} = e^2 \sum_k \int_{-\infty}^{\infty} \frac{d\varepsilon}{\pi} \left( -\frac{\partial f}{\partial \varepsilon} \right) \left( |G_k(\varepsilon)|^2 \cdot v_k(\varepsilon) J_k(\varepsilon) \right. - \left. \text{Re} \{ G_k^{\dagger}(\varepsilon) \cdot v_k^{\dagger}(\varepsilon) \} \right),$$

$$\Delta \sigma_{xy} = B \cdot e^3 \sum_k \int_{-\infty}^{\infty} \frac{d\varepsilon}{2\pi} \left( -\frac{\partial f}{\partial \varepsilon} \right) S_{xy}(k, \varepsilon),$$

$$S_{xy}(k, \varepsilon) = |G_k(\varepsilon)|^2 : \text{Im} G_k(\varepsilon) \times v_k(\varepsilon) \left( J_{ky}(\varepsilon) \frac{\partial J_{xy}(\varepsilon)}{\partial k_y} - J_{xy}(\varepsilon) \frac{\partial J_{ky}(\varepsilon)}{\partial k_y} \right) + \langle x \leftrightarrow y \rangle,$$

where $f(\varepsilon) = (\exp((\varepsilon - \mu)/T) + 1)^{-1}$, and $G_k(\omega + i\delta)$ and $\Sigma_k(\omega + i\delta)$ are derived from $G_k(\omega_n)$ and $\Sigma_k(\omega_n)$ through the numerical analytic continuation. $B$ is the magnetic field parallel to the $z$-axis, $v_k(\omega) = \frac{\partial}{\partial \varepsilon} (\epsilon_k^{\dagger} + \text{Re} \Sigma_k(\omega))$ is the quasiparticle velocity, and $J_{k\mu}(\omega)$ is the total current which contains the vertex correction from $T_{22}$ in the notation of Ref. [4]. Later, we examine its importance in detail.

As for the resistivity, the second term of eq. (8) is neglected in the Eliashberg’s transport theory, whose derivation is given in §III C. We call it the incoherent part of the conductivity, $\sigma_{\text{inc}}$, because it is negligible in the case of $\gamma_k < T$. As a result, these formulæ (8)-(9) are valid even for $\gamma_k \sim T$.

In the present study, we solve the following Bethe-Salpeter equations for $J_{k\mu}(\omega)$ after the manner of the conserving approximation:

$$J_{k\mu}(\omega) = v_{k\mu}(\omega) + \sum_q \int_{-\infty}^{\infty} \frac{d\varepsilon}{2\pi} \left[ \text{cth} \frac{\varepsilon - \omega}{2T} - \frac{\varepsilon^2}{2T} \right] \times \text{Im} V_{k-q}(\epsilon - \omega + i\delta) \cdot |G_k(\varepsilon)|^2 \cdot J_{q\mu}(\varepsilon),$$

where we take only the Maki-Thompson (MT) type VC’s into account, and neglect the Aslamazov-Larkin (AL) terms because it has little contributions if $\chi_q(0)$ has a sharp peak around $q = (\pi, \pi)$; see Fig. 2(a). Figure 2(b) shows the numerical solution of eq. (8). We see that $\mathbf{J}_k$ is not parallel to $\mathbf{v}_k$.

On the other hand, in the Boltzmann theory within the relaxation time approximation (RTA), $J_{k\mu}(\varepsilon)$ is simply replaced by $v_{k\mu}(\varepsilon)$ in eqs. (8)-(9). It is an insufficient approximation for the Hall coefficient in nearly AF state as shown in Refs. [4].
Now we discuss why $\tilde{J}_k$ shows such an anomalous behavior. Here, we choose an arbitrary point on the Fermi surface, $k$, which locates between two hot spots in the region $\pi/2 < \theta < \pi$, and also define that $k' \equiv (k_y, k_z)$. (see Fig. 1). Then, $k - k' \approx (-\pi, \pi)$ is satisfied in the present system, as shown in Fig. 1. In the Bethe-Salpeter eq. (1), $\tilde{J}_k$ are strongly connected with $\tilde{J}_k'$ through $V_{k-k'}(\omega)$ in the presence of strong AF fluctuations. Taking this fact into account, the approximate solution of eq. (8) is given by

$$\tilde{J}_k = (\vec{v}_k + \alpha_k \vec{v}_{k'})(1 - \alpha_k^2)^{-1},$$  
(10)

where $\alpha_k = \cos(\theta_j(k) - \theta_j(q))|_{q-k}|< \xi_{AF}$, and $\theta_j(q)$ is the angle of $\vec{J}_q$. According to the definition, $\alpha_k < 1$ and $(1 - \alpha_k)^{-1} \propto \xi_{AF}^2$. Equation (10) means that $\tilde{J}_k$ at the hot spots will become parallel to the AF zone boundary when $\xi_{AF}$ approaches to infinity. In fact, such a tendency is recognized in Fig. 3(b). Thus, the anomalous behavior of $\tilde{J}_k$, which is the result of the multiple scattering between $\vec{v}_k$ and $\vec{v}_{k'}$, becomes quite singular when the AF fluctuations are dominant. As a result, the RTA is strongly violated when $\xi_{AF} \gg 1$.

Equation (9) is rewritten at low temperatures as

$$\Delta\sigma_{xy} = B \cdot \frac{e^3}{4} \int_{FS} dk_{||} S_{xy}(k_{||})$$

$$S_{xy}(k_{||}) = (\tilde{J}_k \times d\tilde{J}_k/dk_{||})z/\gamma_k^2$$

$$= |\tilde{J}_k|^2 (d\theta_j(k)/dk_{||})^2/\gamma_k^2,$$  
(11)

where $\int_{FS} dk_{||}$ represents the momentum integration along the Fermi surface, and $dk_{||}$ is parallel to the Fermi surface.

Because $d\alpha_k/dk_{||} = 0$ at the cold spot due to the symmetry of this model, $(\tilde{J}_k \times d\tilde{J}_k/dk_{||})z = (1 - \alpha_k^2)^{-1} \cdot (\vec{v}_k \times d\vec{v}_k/dk_{||})z$ at the cold spot by using eq. (10). Because $\Delta\sigma_{xy}$ is given mainly around the cold spots in the $k_{||}$-integration of eq. (11), we conclude the relation

$$\Delta\sigma_{xy}/\Delta\sigma_{xy}^0 \propto \xi_{AF}^2 \propto T^{-1}$$  
(12)

in the presence of the strong AF fluctuations. Here $\Delta\sigma_{xy}^0$ is given by replacing $\tilde{J}_k(\omega)$ with $\vec{v}_k(\omega)$ in eq. (7), which is equal to the result of the RTA. As for $\Delta\sigma_{xy}^0$, the conventional Kohler’s rule $\Delta\sigma_{xy} \propto (\sigma_{xx})^2$ is well satisfied in the present calculation because the anisotropy of $\vec{v}_k$ is not so extreme. ($\gamma_{hot}/\gamma_{cold}$ is at most 3.) As a result, eq. (12) leads the relation $R_H \equiv (\Delta\sigma_{xy}/B) \cdot \rho^2 \propto \xi_{AF}^2 \propto T^{-1}$, which is recognized in the present numerical calculations as shown below.

The above analysis is confirmed by the numerical results of $S_{xy}(k_{||})$ in Fig. 3(c). $S_{xy}(k_{||})$ is given by the RTA. The dominant contributions to $\Delta\sigma_{xy}$ come from the region around the cold spot, $S_{xy}(k_{||})$ for region $\pi/4 < \theta < \pi/2$ is considerably small because the curvature of the Fermi surface is very small there. In the present case, both $S_{xy}(k_{||})$ and $S_{xy}(k_{||})$ are positive everywhere. It is not the case for high-$T_c$ cuprates; for example, the change of the sign of $R_H$ is realized in Nd-compound.

**B. Numerical Results for Transport Phenomena**

Now, we study both $\rho$ and $R_H$ for various values of $U$, because the main effect of the applied pressure is expected to increases the bandwidth $W_k$, in other words, to reduce the value of $U/W_k$. At the same time, the value of $t'/t$ may be also modified by pressure. This effect is not discussed here although the electronic states are sensitive to $t'/t$ according to the FLEX approximation.

Figure 5(a) shows the temperature dependences of the resistivity $\rho = 1/\sigma_{xx}$. All the $\rho$’s show the approximate $T$-linear resistivity, reflecting the $T$-linear behavior of $\gamma_{cold}$. At the same temperature, $\rho$ increases monotonously as $U$ increases. Here, $\rho_0 = 1/\sigma_{xx}^0$ is the resistivity without the VC’s, which is given by replacing $\tilde{J}_k(\omega)$ with $\vec{v}_k(\omega)$ in eq. (7). We see that $\rho > \rho_0$ due to the VC’s for the current.

Here, we comment on the anisotropy of $\rho$. In the present model, $\sigma_{x\mu}$ depends on the angle of the $\mu$-axis because there is no four-fold rotational symmetry in this system. We find that $\rho$ takes its maximum (minimum) value along the $(\theta = \pi/4)$-axis ($(\theta = 3\pi/4)$-axis), and $\rho_{max}/\rho_{min} \approx 1.6$ with weak temperature dependence.

Next, we discuss on the Hall coefficient $R_H$. As shown in Fig. 5(b), $R_H$ follows the Curie-Weiss like temperature dependence, which is consistent with the relation (13). Actually, both $\xi_{AF} \propto \chi_{Q}(0)$ and $R_H$ increase monotonously as $U$ increases at the same temperatures. (see Fig. 2). As a result, the large temperature dependence of $R_H$ in $\kappa$-(BEDT-TTF)$_2$X salts is reproduced well by taking account of the VC’s for the current.

To see the importance of the VC’s, we also show $R_{H0}^0 = (\Delta\sigma_{xy}^0/B) \cdot \rho_0^2$, which is the result of the RTA: We see that $R_H^0$ is nearly constant, and $R_H \approx R_H^0$ at higher temperatures ($T \geq 0.1$) because of $\tilde{J}_k \approx \vec{v}_k$ in this case. We note that $dR_H^0/dT$ is slightly positive for $T > 0.05$, because the curvature of the Fermi surface around the cold spot decreases as $T$ decreases due to the
growth of the AF fluctuations. (see Fig. 1) Whereas $dR_H^0/dT < 0$ for $T < 0.05$ because of the rapid increase of the anisotropy of $\gamma_k$ at lower temperatures. However, this increase of $R_H^0$ is too small to explain experimental results.

Finally, we show $\cot \theta_H = \sigma_{xx}/\Delta \sigma_{xy}$ in Fig. 3 (c). All the $\cot \theta_H$’s are approximately proportional to $T^2$ below $T \lesssim 0.05$, where both $R_H \propto T^{-1}$ and $\rho \propto T$ are satisfied approximately. This result is highly consistent with experiments reported in Ref. 7. Similar behavior of $\cot \theta_H$ is also observed in high-$T_c$ cuprates. For high-$T_c$ cuprates, Anderson claimed that it suggests the non-Fermi liquid ground state which possesses two kinds of relaxation rates \cite{Anderson}. However, we stress that the relation $\cot \theta_H \propto T^2$ is naturally understood both for $\kappa$-(BEDT-TTF)$_2$X salts and for high-$T_c$ cuprates within the framework of the nearly AF Fermi liquid.

Finally, we discuss the $U$-dependence of $\rho$ and $R_H$ in the present calculations, and compare them with the experimental pressure dependences. According to the experiments on X=Cu[N(CN)$_2$]Cl, as the applied pressure increases, (i) $\rho$ decreases monotonously, (ii) $R_H$ is almost unchanged, and (iii) $\cot \theta_H$ decreases monotonously. The effect of the applied pressure is to increase $W_b$ while $U$ is unchanged, i.e., to decrease $U/W_b$. Moreover, on condition that $U/W_b$ is constant, it is easy to see that $\rho \propto W_b^{-1}$, $R_H \propto W_b^0$, and $\cot(\Theta_H) \propto W_b^{-1}$. As a result, the obtained $U$-dependence of $\rho$ and $\cot \theta_H$, which are shown in Fig. 3 (a) and (c), are consistent with experiments. On the other hand, the observed weak pressure dependence of $R_H$ will correspond to the behavior for $U \geq 8$ in Fig. 3 (b). In conclusion, experimentally observed pressure effects (i)-(iii) are reproduced in our study.

At last, we comment on the superconductivity: We find $T_c = 0.024$ for $U = 9$ and $T_c = 0.004$ for $U = 5$ by solving the Eliashberg equations. This is consistent with the decrease of $T_c$ under pressure observed experimentally. Roughly speaking, $T = 0.01$ corresponds to 10K because the band-width $W_{band} \sim 0.5$eV at ambient pressure. Although the obtained $T_c$’s is rather higher than experimental one’s, it decreases if we put $t'/t$ larger than 0.7\cite{H*.k}

C. Derivation of $\sigma_{inc}$

In this subsection, we give the derivation of the second term of eq. (6) based on the Fermi liquid theory. We call it the incoherent part of the conductivity $\sigma_{inc}$ because it gives negligible contribution when the quasiparticles are well-defined. We call the first term of eq. (6) the coherent part of the conductivity $\sigma_{coh}$, which was derived by Eliashberg under the assumption $\gamma_k \ll T$\cite{Eliashberg}.
According to the Kubo formula, the conductivity is expressed by the retarded two-particle Green function $K_R^R(\omega)$, whose explicit form within the Fermi liquid theory is given by eq. (9) of Ref. [4]. The first term of eq. (14), which was derived by Eliashberg, comes from the coherent terms of $K_R^R(\omega)$ which include at least one $g_2 = G^R G^A$. Here, we study the contribution from the the incoherent part without $g_2$, $K_{inc}^R(\omega)$, which has not been analyzed previously. Hereafter, we omit the momentum variables for simplicity. $K_{inc}^R(\omega)$ is given as follows:

$$K_{inc}^R(\omega) = -\sum_{i=1,3} \int \frac{d\epsilon}{4\pi i} \imath \lambda_0(\epsilon;\omega) g_1(\epsilon;\omega) \lambda_j(\epsilon;\omega), \quad (13)$$

$$\Lambda_j^\epsilon(\epsilon;\omega) = \left\{ \begin{array}{l}
\frac{d\epsilon}{4\pi i} \mathcal{T}_{i,j}(\epsilon,\epsilon';\omega) \\
\times g_j(\epsilon';\omega) \Lambda_j^\epsilon(\epsilon';\omega),
\end{array} \right. \quad (14)$$

which is shown in Fig. 6(a). Here, $g_1(\epsilon;\omega) = G_G(\epsilon + i\delta) G_G(\epsilon + i\delta)$, and $g_3(\epsilon;\omega) = \{g_1(\epsilon;\omega)\}^* \lambda_3(\epsilon;\omega) = \delta(\epsilon + \omega)/2\pi$, and $\Lambda_j^\epsilon(\epsilon;\omega)$ is given by the analytic continuation, whose definition is given by eq. (12) of Ref. [4]. (Hereafter, all the four-point vertices are 'irreducible' with respect to $g_1$.) We study the conductivity from the incoherent terms, given by $\sigma_{inc} = e^2 \lim_{\omega \to 0} \Re K_{inc}^R(\omega)/\omega$, and find that it gives a finite contribution when the life-time of the quasiparticles becomes shorter.

\[\text{FIG. 6. The diagrams for (a) } K_{inc}(\omega), \text{ and for (b) } \sigma_{inc}, \text{ respectively.}\]

Now, we derive the $\omega$-linear term of $\Re K_{inc}^R(\omega)$. $\mathcal{T}_{i,j}(\epsilon,\epsilon';\omega)$ (i = 1, 3) contains the thermal factors $\partial f/\partial \epsilon$ in eq. (12) of Ref. [4]. We can check that $K_{inc}^R(\omega)$ given by eqs. (13) and (14) becomes real quantity when we put $\omega = 0$ (i) in all the factors $\partial f/\partial \epsilon$ in eq. (14) and (ii) in all the irreducible vertices $\Gamma_{i,j}(\epsilon,\epsilon';\omega)$, by taking account of the relation $\Gamma_{i,j}^{(II)}(\epsilon,\epsilon';0) = \Gamma_{4-i,4-j}^{(II)}(\epsilon,\epsilon';0)$ for $i, j = 1, 3$. This means that the $\omega$-linear term of $\Im K_{inc}^R(\omega)$ comes only from one of (i) or (ii). Because $\Lambda_j^\epsilon(\epsilon;\omega)$ contains infinite number of $\mathcal{T}_{i,j}(\epsilon;\omega)$, we find that

$$\sigma_{inc} = -e^2 \int_{-\infty}^{\infty} \frac{d\epsilon}{\pi} \left\{ -\frac{\partial f}{\partial \epsilon} \right\} \Re \left\{ \Lambda_j^\epsilon(\epsilon;0) g_1(\epsilon;0) \Lambda_j^\epsilon(\epsilon;0) \right\} \quad (15)$$

where $\mathcal{T}_{i,j}(\epsilon,\epsilon')$ is defined as the $i\omega$-derivative of the vertex part of $\mathcal{T}_{i,j}(\epsilon,\epsilon';\omega)$, and put $\omega = 0$.

As for the second term of eq. (15), $\mathcal{T}_{i,j}(\epsilon,\epsilon')$ given by the MIT-term vanishes identically. On the other hand, the two AL-terms causes the finite contribution for $\Im K_{inc}^R(\omega)$. After the long calculation, $\mathcal{T}_{1,1}(\epsilon,\epsilon')$ in the FLEX approximation is given by

$$\mathcal{T}_{1,1}(\epsilon,\epsilon') = \mathcal{G}_{1,1}(\epsilon,\epsilon') \cdot \frac{\partial f}{\partial \epsilon}$$

where we have used the relation $\mathcal{G}_k = -\mathcal{V}_k$ in derivation, and $\mathcal{V}_k(\omega) = -\Im G_k(\omega + i\delta)/\pi$. Other terms which do not contribute to $\sigma_{inc}$ are dropped in eq. (16). In the same way of deriving eq. (17), we can show that $\mathcal{T}_{1,1}(\epsilon,\epsilon') = (-1)^{j-1}/2 \mathcal{T}_{1,1}(\epsilon,\epsilon')$ for $i, j = 1, 3$.

Now, we show that the second term of eq. (15) is negligible: It vanishes at $T = 0$ because Equation (17) vanishes in this case. (Note that $\Im G_k(0) = 0$.) It should be much smaller even at finite temperatures because of the cancellation caused by the factor $(\rho\epsilon + \rho\epsilon')$ in eq. (17). By this reason, we consider only the first term of eq. (15) hereafter.

As for $\Lambda_j^\epsilon(\epsilon;\omega)$ in eq. (13), we can show that

$$\Re \left\{ \Lambda_j^\epsilon(\epsilon;0) - \Lambda_j^\epsilon(\epsilon;0) \right\} = \frac{1}{2} \sum_{i,j=1,3} \int \frac{d\epsilon}{4\pi i} \left\{ \frac{\partial f}{\partial \epsilon} \right\} \left\{ \begin{array}{l}
\left( (\Gamma_{1,1}^{(II)}(\epsilon,\epsilon';0) \right. \\
- \left. (\Gamma_{2,2}^{(II)}(\epsilon,\epsilon';0) \right) \right) \mathcal{G}_k(\epsilon + i\delta) \mathcal{G}_k(\epsilon + i\delta)
\end{array} \right\}$$

which is proportional to $T^2$ at low temperatures. (Here, $\Gamma_{1,1}^{(II)}(\epsilon,\epsilon';0)$ is reducible with respect to $g_1$.) Thus, $\Lambda_j^\epsilon(\epsilon;0)$ approximately identical to $\Lambda_j^\epsilon(\epsilon;0)$. We have also estimated eq. (13) numerically by taking only the MIT terms into account, and find that the difference between them is at most a few percent.

In conclusion, we get the following expression for $\sigma_{inc}$:

$$\sigma_{inc} = -e^2 \sum_k \int_{-\infty}^{\infty} \frac{d\epsilon}{\pi} \left\{ -\frac{\partial f}{\partial \epsilon} \right\} \Re \left\{ \mathcal{G}_k(\epsilon + i\delta) \mathcal{V}_k(\epsilon + i\delta) \right\} \right\} \right\}$$

which is shown in Fig. 6(b). Here we have used the Ward identity $\mathcal{G}_k(\epsilon;0) = \mathcal{V}_k(\epsilon;0) + \delta(\epsilon) \Sigma_k(\epsilon + i\delta) = \mathcal{V}_k(\epsilon + i\delta)$. Note that the vertex correction, given by the momentum derivative of $\Sigma_k(\omega)$ appears twice in eq. (13). Thus the obtained $\sigma_{inc}$ gives the second term of eq. (1).
If we can put $G_k(\epsilon) = z_k/(\epsilon - \epsilon^* + i\gamma^*_k)$, eq. (19) becomes
\[\sigma_{\text{inc}} = e^2 \sum_k \int \frac{d\epsilon}{\pi} \left( -\frac{\partial f}{\partial \epsilon} \right) z_k^2 \gamma^*_k \left( (\epsilon - \epsilon^* + i\gamma^*_k)^2 + \gamma^*_k \right) \gamma^*_k, \]
where $\gamma^*_k = z_k \gamma^*_k$ and $\epsilon^*_k = z_k \epsilon^*_k$, respectively. In the case of $\gamma^*_k \ll T \ll W_b$, $\sigma_{\text{inc}} \approx 0$ is realized according to eq. (20), while $\sigma_{\text{coh}} \propto \gamma^*_k$. However, such a condition is not satisfied in the present calculation as shown in Fig. 3(a). At lower temperatures, $\gamma^*_k \approx T$ is satisfied because $z_k^{-1} \lesssim 10$ then, whereas $\gamma^*_k \gtrsim T$ at higher temperatures because $z_k^{-1}$ decreases as $T$ increases. Thus, $\sigma_{\text{inc}}$ is expected to be important at higher temperatures. In Appendix A, we show its importance numerically.

**IV. COMPARISON WITH EXPERIMENTS**

**A. Effect of Band-Splitting around the Hot-Spots**

In this section, we compare our theoretical results with experiments in more detail. Here, we discuss the validity of the present results based on the effective model shown in Fig. 4. Precisely speaking, in many (not all) real $\kappa$-(BEDT-TTF)$_2$X compounds, the Fermi surface splits slightly around the hot spots in Fig. 4 because a unit cell contains two dimers of the BEDT-TTF molecules (see Appendix B). In this sense, the present model may be too simplified for the quantitative studies.

However, the mechanism of the enhancement of $R_H$ due to the AF fluctuations proposed in this paper is surely valid because only the quasiparticles around the cold spots plays an important role for transport phenomena as shown in Fig. 5. On the other hand, $R_H$ at higher temperatures may be affected by the splitting of the Fermi surface at the hot spots. Thus, our calculation based on the dispersion, eq. (19), is comparable with experiments at least in the lower temperature region.

**B. Effect of Temperature Dependence of the Volume**

Next, we discuss the effect of the thermal contraction of the volume, which is known to be quite large in various organic metals. For example, (TMTSF)$_2$PF$_6$ at ambient pressure shows $\rho \propto T^n$ and $n \approx 2$, whereas $n \approx 1$ is concluded after the effect of the thermal contraction is compensated.

As for $X$={Cu[CN]}$_2$Cl, $\rho \propto T^n$ and $n \approx 2$ is observed below 100K at nearly ambient pressures. However, $n \approx 1$ is realized qualitatively under the constant-volume condition according to Ref. 4. In the article, the authors used the piston-cylinder clamped cell to make pressure, and the oil inside of the cell freezes at 200K, which make the volume of the sample constant.

**C. The Saturation of $R_H$ at Lower Temperatures**

Here, we comment on the saturation of $R_H$ below a characteristic temperature $T^*$ observed experimentally. Around $T \approx T^*$, the $1/T^2$ also saturates and begins to decrease below $T^*$, which is called the pseudo spin-gap behavior. This results suggests that $\xi_{AF}^2$, or $\chi_{gap}^2(0)$, will saturates below $T^*$. (We note that many experiments for $1/T^2$ are done at ambient pressure, so the volume contraction effect may play some quantitative effect on $1/T^2$ at low temperatures.) Thus, the analytical relation in our work, $R_H \propto \xi_{AF}^2$, is consistent with these experiments.

However, $R_H$ of our numerical calculation in Figs. 3 does not saturate: This is because the FLEX approximation does not reproduce the saturation of $\xi_{AF}^2$ below $T^*$, which is a significant future problem. One of the possible mechanism for it will be the precursor effect of superconductivity below $T^*$. Finally, we point out that $R_H$ begins to decrease on cooling below $T^*$ in under-doped high-$T_c$ cuprates, whereas it takes a saturate value at lower temperatures in $X$={Cu[CN]}$_2$Cl.

**V. CONCLUSIONS**

In this paper, we have presented the theoretical study for the resistivity and the Hall coefficient of the $\kappa$-(BEDT-TTF)$_2$X salts. Reference 2 point out some experimental evidences that there anomalous behaviors have close connection with the grows of the AF fluctuations. According to our theory, the Hall coefficient follows the relation $R_H \propto \xi_{AF}^2 \propto T^{-1}$ in nearly AF Fermi liquid state, which is consistent with the experiment under the constant volume condition. This anomaly of $R_H$, which can not be reproduced by the RTA, is found to come from the VC’s for the current which is indispensable to satisfy the conserving laws.

Moreover, based on the Kubo formula, we have derived the expression of the incoherent conductivity $\sigma_{\text{inc}}$ beyond the Eliashberg’s transport theory, and found that it give a qualitatively important contribution in $\kappa$-(BEDT-TTF)$_2$X and in high-$T_c$ cuprates at higher temperatures,
where \( \gamma_k \sim T \) is realized. It also give the appropriate temperature dependence of \( R_H \).

We have calculated both \( \rho \) and \( R_H \) based on the FLEX approximation for \( U = 6 \sim 10 \), without assuming any fitting parameters. The obtained \( U \)-dependences for \( \rho \), \( R_H \) and \( \cot \theta_H \) explain well the experimentally observed pressure dependences. In conclusion, many essential electronic properties of \( \kappa \)-(BEDT-TTF)\(_2\)X, especially both the anomalies of transport phenomena and the phase diagram, are explained well from the standpoint of the nearly AF Fermi liquid state. We stress that further observations under the constant volume condition are highly demanded for organic metals to make a meaningful comparison between theory.

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APPENDIX A: IMPORTANCE OF THE INCOHERENT CONDUCTIVITY

In this appendix, we numerically show the important role of \( \sigma_{\text{inc}} \) to get the reasonable behaviors of \( \rho \) and \( R_H \). Figure 7 shows the temperature dependence of the Hall coefficient and the resistivity. \( \rho = 1/(\sigma_{\text{coh}} + \sigma_{\text{inc}}) \), \( \rho' = 1/\sigma_{\text{coh}} \) and \( \rho_{\text{RTA}} = 1/\sigma_{\text{RTA}} \), respectively. Here, \( \sigma_{\text{RTA}} \) is given by replacing \( J_k \) with \( v_k \) in \( \sigma_{\text{coh}} \), which is equal to the result from the relaxation time approximation. We find that (i) \( \rho' > \rho_{\text{RTA}} \) because of the VC’s for the current, and (ii) \( \rho < \rho' \) because of \( \sigma_{\text{inc}} \), which becomes dominant especially at higher temperatures. As a result, \( \rho < \rho_{\text{RTA}} \) is realized at higher temperatures.

FIG. 7. (i) Comparison between \( \rho = 1/(\sigma_{\text{coh}} + \sigma_{\text{inc}}) \) and \( \rho' = 1/\sigma_{\text{coh}} \) for \( U = 10 \). \( \rho \) becomes smaller than \( \rho' \) because of the incoherent part of the resistivity. \( \rho_{\text{RTA}} \) is given by the RTA. (ii) Comparison between \( R_H = (\Delta\sigma_{xy}/B) \cdot \rho^2 \) and \( R_H' = (\Delta\sigma_{xy}/B) \cdot \rho'^2 \) for \( U = 10 \). We see that \( dR_H/dT \) becomes positive below \( T \approx 0.08 \), which is inconsistent with experiments.

As for Hall effect, \( R_H = (\Delta\sigma_{xy}/B) \cdot \rho^2 \) and \( R_H' = (\Delta\sigma_{xy}/B) \cdot \rho'^2 \), respectively. We see that \( R_H < R_H' \) because \( \rho < \rho' \). However, \( R_H(T = 0.02)/R_H(T = 0.2) \) is larger than that of \( R_H' \), so the incoherent conductivity make the temperature dependence of \( R_H \) larger. We stress that \( dR_H'/dT \) become positive at higher temperature, which contradicts with experiments. In conclusion, we find that \( \sigma_{\text{inc}} \) is necessary to reproduce the reasonable behavior of \( R_H \).

APPENDIX B: THE MORE PRECISE TIGHT-BINDING MODEL FOR \( \kappa \)-(BEDT-TTF)\(_2\)X

In real \( \kappa \)-(BEDT-TTF)\(_2\)X systems, there are two pairs of closely-packed BEDT-TTF molecules in a unit cell, and only the bonding-orbit of each closely-packed molecules contributes to make the Fermi surface. By taking the results of the band-calculations into account, we get the effective tight-binding model for \( \kappa \)-(BEDT-TTF)\(_2\)X as shown in Fig. 8, where there are two sites (a,b) in a unit cell. Each site corresponds to a closely-packed BEDT-TTF molecules. This model becomes equal to the anisotropic triangular lattice model given by Fig. 1 if we put \( t_1 = t_1' = t_2 = t_2' = t \) and \( t_3 = t_3' \).

FIG. 8. (a): The effective model for \( \kappa \)-(BEDT-TTF) salts with two-sites in a unit cell (a,b). The hopping parameters of this model are given by the parameters in Fig.1 of Ref. 1 as \( t_1 = (p + q)/2 \), \( t_1' = (p + q)/2 \), \( t_2 = (p' + q)/2 \), \( t_2' = (p' + q)/2 \), and \( t_3 = b_2/2 \), respectively. (b): The Fermi surface for \( t_1 = t_1' = t_2 = t_2' = 1 \) (full line), and for \( t_1 = t_1' = 1.1 \) and \( t_2 = t_2' = 0.9 \) (broken line), respectively. In both cases, \( t_3 = 0.7 \). Note that the former is equivalent to Fig.1 in the extended zone representation.

(a) (b)

Y

X
The dispersion of the tight-binding model given by Fig. 6 for \( U = 0 \) is derived as
\[
\epsilon_k = 2t_3 \cos k_y \left[ \left| t'_1 \right|^2 + t'_2^2 + t''_2^2 + 2(t_1 t'_1 + t_2 t'_2) \cos k_y + 2(t'_1 t_2 + t_1 t'_2) \cos k_x + 2t_1 t'_2 \cos(k_x + k_y) + 2t'_1 t_2 \cos(k_x - k_y) \right]^{1/2}.
\]
When \( |k_y| = \pi \), then \( \epsilon_k^+ - \epsilon_k^- = 2\left( (t_1 - t'_1)^2 + (t_2 - t'_2)^2 \right)^{1/2} \). This means that the Fermi surface splits around the hot spots when \( t_1 \neq t'_1 \) or \( t_2 \neq t'_2 \), which is realized in many systems. (However, both \( t_1 = t'_1 \) and \( t_2 = t'_2 \) are satisfied exactly in some compounds exceptionally, e.g., (28)). This splitting of the Fermi surface at the hot spots will not affect the temperature dependence of \( R_H \) at low temperatures, as discussed in §III.

1. K. Kanoda: Physica C 282-287 (1997) 299.
2. A. Kawano, K. Miyagawa, Y. Nakazawa and K. Kanoda: Phys. Rev. B 52 (1995) 15 522.
3. H. Kino, and H. Kontani: J. Phys. Soc. Jpn. 67 (1998).
4. H. Kondo and T. Moriya: J. Phys. Soc. Jpn. 67 3695 (1998).
5. J. Schmalian: Phys. Rev. Lett. 81 (1998) 4232.
6. H. Kondo and T. Moriya: J. Phys. Soc. Jpn. 68 (1999).
7. Yu. V. Sushko, N. Shirakawa, K. Murata, Y. Kubo, N.D. Kushch, E.B. Yagubskaï: Synth. Met. 85 (1997) 1541.
8. K. Murata, M. Ishibashi, Y. Honda, N.A. Fortune, M. Tokumoto, N. Kinoshita and H. Anzai: Solid State Comm. 76 (1990) 377.
9. H. Kontani, K. Kanki and K. Ueda: Phys. Rev. B 59 (1999) 14723.
10. K. Kanki and H. Kontani: J. Phys. Soc. Jpn. 68, (1999) 1614.
11. H. Kino and H. Fukuyama: J. Phys. Soc. Jpn. 65 (1996) 2158, and references therein.
12. N. E. Bickers et al.: Phys. Rev. Lett. 62 (1989) 961.
13. P. Monthoux and D. J. Scalapino: Phys. Rev. Lett. 72 (1994) 1874.
14. T. Dahm and L. Tewordt: Phys. Rev. B 52 (1995) 1297.
15. H. Kontani and K. Ueda: Phys. Rev. Lett. 80 (1998) 5619.
16. G. M. Eliashberg: Sov. Phys. JETP 14 (1962), 886.
17. H. Kohno and K. Yamada: Prog. Theor. Phys. 80 (1988) 623.
18. H. Fukuyama, H. Ebisawa and Y. Wada: Prog. Theor. Phys. 42 (1969) 494.
19. According to the analysis, \( \tilde{J}_0 \) becomes parallel to the magnetic Brillouin zone at the hot spot when \( \xi_{AF} \to \infty \), which may be interpreted as the precursor effect of the AF ordered state.
20. P.W. Anderson: Phys. Rev. Lett. 67 (1991) 2092.
21. M. Watanabe, Y. Nogami, K. Oshima, H. Ito, T. Ishiguro and G. Saito: accepted in Synth. Met.
22. C. Bourbonnais and D. Jérome: cond-mat/9903103.
23. M. Dressel eq al.: Synth. Metals. 85 (1997) 1503.
24. K. Murata: private communication.
25. When \( \rho \propto T^2 \) is observed in \( \kappa \)-(BEDT-TTF)\( _2 \)X compounds, in general, (i) the volume contraction is prominent, and (ii) \( \rho \) shows an insulating behavior at higher temperatures.
26. T. Jujo and K. Yamada: J. Phys. Soc. Jpn 68 (1999) 2198.
27. The experimentally observed ‘positive magnetoresistance’ is also reproduced by the VC’s theoretically if the AF fluctuations are dominant [H. Kontani: unpublished].
28. e.g., T. Komatsu, N. Matsukawa, T. Inoue and G. Saito: J. Phys. Soc. Jpn. 65 (1996) 1340: The overlap integrals are also listed for a number of \( \kappa \)-(BEDT-TTF)\( _2 \)X salts and related materials.