Seebeck coefficient of two-dimensional Dirac electrons in an organic conductor under pressure

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The Seebeck coefficient, which is proportional to a ratio of the thermoelectric conductivity to electrical conductivity has been examined for Dirac electrons in the organic conductor α-(BEDT-TTF)$_2$I$_3$ [BEDT-TTF denotes a molecule given by bis(ethylenedithio)tetrathiafulvalene] under a uniaxial pressure using a two-dimensional tight-binding model with both impurity and electron–phonon (e–p) scatterings. We calculate an anomalous temperature ($T$) dependence of the Seebeck coefficient $S_\nu$ with $\nu = x$ (perpendicular to the molecular stacking axis) and $y$, which shows $S_x > 0$ with a maximum at high temperatures and $S_y < 0$ with a minimum at low temperatures. The microscopic mechanism of such a sign change of $S_\nu$ is clarified in terms of the spectral conductivity. The result is compared with experiments on α-(BEDT-TTF)$_2$I$_3$.

I. INTRODUCTION

The two-dimensional massless Dirac fermions$^{1,2}$ which show a linear spectrum around the Dirac point have been studied extensively. Several properties as a bulk material are explored in an organic conductor$^{2}$ given by α-(BEDT-TTF)$_2$I$_3$, where α-(BEDT-TTF) denotes bis(ethylenedithio)tetrathiafulvalene. The conductor exhibits a zero-gap state (ZGS)$^3$ and the transport property is characterized by the density of states (DOS), which vanishes linearly at the Fermi energy$^4$. The explicit band structure of the Dirac cone is obtained using a tight-binding (TB) model, where transfer energies under pressures are estimated from the extended Hückel method$^{5,6}$. The Dirac cone was verified by the first-principles density functional theory (DFT) calculation. A further two-band model$^{7,8}$ has been proposed to examine Dirac electrons in an organic conductor.

Characteristic properties of the Dirac cone appear in the temperature ($T$) dependence of physical quantities. Magnetic susceptibility with a $T$ linear behavior at low temperatures shows a good correspondence between the theory and experiment$^{9,10}$. The chemical potential $\mu$, which also depends on $T$, takes a significant role in the transport. The reversal of the sign of the Hall coefficient occurs when $\mu$ becomes equal to the energy of the Dirac point. Such a sign reversal of the Hall coefficient was observed theoretically by assuming the extremely small magnitude of the impurity scattering$^{11,12}$. The sign reversal was also observed experimentally in the Hall conductivity of α-(BEDT-TTF)$_2$I$_3$.

Since the conductivity of Dirac electrons is fundamental as the transport, a two-band model with the conduction and valence bands has been studied, where the static conductivity at absolute zero temperature remains finite with a universal value, i.e., independent of the magnitude of impurity scattering owing to a quantum effect$^{13}$. At absolute zero temperature, the tilting of the Dirac cone provides the anisotropic conductivity and the deviation of the current from the applied electric field$^{14}$. At finite temperatures, the conductivity depends on the magnitude of the impurity scattering, $\Gamma$, which is proportional to the inverse of the life-time by the disorder. With increasing temperature, the conductivity increases for $\Gamma \ll T$.$^{15}$ Although a monotonic increase in the conductivity is expected for such a model, the measured conductivity (or resistivity) on the above organic conductor shows an almost constant behavior at high temperatures.$^{16,17}$ This is a noticeable transport of the Dirac electron in the presence of the electron-phonon (e–p) interaction, since the resistivity of the conventional metal at high temperatures increases linearly with respect to $T$ due to the e–p scattering. The resistivity showing a nearly constant behavior at high temperatures is explained by the acoustic phonon scatterings using a simple two-band model of the Dirac cone without tilting.$^{18}$ Although the effect of the e–p scattering at high temperatures is qualitatively understood, the model should be improved to explain the conductivity of the actual organic conductor, where the energy bands show deviations from the linear spectrum.$^{19,20}$ Thus, the TB model with transfer energies of α-(BEDT-TTF)$_2$I$_3$ is examined to show that the presence of acoustic phonons give rise to conductivity being nearly constant at high temperatures.$^{21,22}$

In addition to the electric conductivity, it is of interest to examine the thermoelectric (i.e., Seebeck) effect on the above model, where the $T$ dependence of $\mu$ takes a crucial role. The Seebeck coefficient can be obtained microscopically in terms of linear response theory$^{23,24}$. However, we have to be careful in treating the heat current, because there are several forms of the heat current depending on the Hamiltonian.$^{25,26}$ In the case with impurity potentials and electron-phonon interactions, Jonson and Mahan$^{27}$ showed that the heat current $J_Q$ can be expressed as

$$J_Q = J_{Q^{\text{kin}}} + J_{Q^{\text{pot}}} + J_{Q^{\text{p(I)}}} + J_{Q^{\text{p(II)}}} + J_{Q^{\text{ph}}},$$

(1)

(see, also Ref.$^{28}$), where $J_{Q^{\text{kin}}}$, $J_{Q^{\text{pot}}}$, and $J_{Q^{\text{ph}}}$ represent...
the heat current operators originating from the kinetic energy of electrons, from the (impurity) potentials, and from the phonon Hamiltonian, respectively. The heat current due to the electron-phonon interaction, $J_{Q}^{\text{ph}}$, is divided into two contributions. If one takes account of only $J_{Q}^{\text{ph}} = J_{Q}^{\text{phonon}} + J_{Q}^{\text{uniaxial}}$ as the heat current operator, one can show that Eqs. (14) and (15) below (called the Sommerfeld-Bethe relation) hold. However, $J_{Q}^{\text{uniaxial}}$ and $J_{Q}^{\text{multiaxial}}$ do not satisfy the Sommerfeld-Bethe relation and will give additional contributions in the electrothermal conductivity. For example, $J_{Q}^{\text{ph}}$ leads to the phonon drag effect. Jonson and Mahan also discussed that the contribution from $J_{Q}^{\text{uniaxial}}$ is small in nearly free electron systems. Thus, we do not consider this term and, in the following, we use the Sommerfeld-Bethe relation leaving the phonon-drag problem as a future problem.

So far, there are several theoretical studies on the Seebeck (and Nernst) effect in the Dirac electron systems, where the Seebeck coefficient exhibits the variety of the sign. In this paper, we study the Seebeck coefficient for the ZGS of Dirac electrons in the two-dimensional organic conductor, $\alpha$-(BEDT-TTF)$_2$I$_3$. There have been several experimental and theoretical studies on this material. As for the experiments, the ZGS has been obtained under both uniaxial pressures above $P_a = 5$ kbar and hydrostatic pressures above 1.5 GPa. Under the uniaxial pressures, the ZGS was found only for $P_a$ corresponding to the pressure along the $a$ direction. There are several measurements of resistivity suggesting the ZGS under the hydrostatic pressures. Regarding the Seebeck coefficient, the measurement has been performed only for hydrostatic pressures, where the sign change of the Seebeck coefficient with decreasing temperature occurs along the $b$ direction. However, another experiment exhibits the positive Seebeck coefficient without the sign change. It could be ascribed to the effect of the hole doping, since the latter material is a different sample from the former one.

As for the theory, there is a work discussing the sign reversal for the Seebeck coefficient of $\alpha$-(BEDT-TTF)$_2$I$_3$ under the hydrostatic pressure. However, the sign of the Seebeck coefficient for the $b$ direction obtained in this theory disagrees with that of the experiment. This issue remains as a future problem. In the present paper, we examine the Seebeck coefficient for uniaxial pressures, although the experiment has not yet been performed. We will show the sign change of the Seebeck coefficient in this case.

The present paper is organized as follows. First, the model and formulation to calculate the Seebeck coefficient for $\alpha$-(BEDT-TTF)$_2$I$_3$ with 3/4-filled band are given. Next, after calculating the $T$ dependence of the chemical potential, we show the Seebeck coefficient with the electric conductivity, which is analyzed in terms of the spectral conductivity. Finally, discussions, summary, and comparison with the experiment are given.

II. FORMULATION

We consider a two-dimensional Dirac electron system per spin, which is given by

$$H = H_0 + H_p + H_{e-p} + H_{\text{imp}}.$$  \hspace{1cm} (2)

$H_0$ describes a TB model of the organic conductor, $\alpha$-(BEDT-TTF)$_2$I$_3$. $H_p$ and $H_{e-p}$ describe an acoustic phonon and an electron-phonon (e-p) interaction, respectively. $H_{\text{imp}}$ is the impurity potential. The unit of the energy is taken as eV. Figure 1(a) shows the TB model for $H_0$ consisting of four BEDT-TTF molecules in the unit cell. $H_0$ is expressed as

$$H_0 = \sum_{i,j=1}^{N} \sum_{\alpha,\beta=1}^{4} \sum_{k} h_{\alpha\beta}(k)a_{i,\alpha}^\dagger a_{j,\beta}$$  \hspace{1cm} (3)

where $a_{i,\alpha}^\dagger$ denotes a creation operator of an electron of molecule $\alpha$ $= (\alpha(1), \alpha(2), \beta(3), \text{and} \beta(4))$ in the unit cell at the $i$-th lattice site. $N$ is the total number of square lattice sites and $a_{i,\alpha,\beta}$ denote the seven kinds of transfer energies $a_0, \cdots, a_3, b_1, \cdots, b_4$ between the nearest-neighbor (NN) sites as shown in Fig. 1(a). A Fourier transform for the operator $a_{i,\alpha,\beta}$ is given by

$$a_{i,\alpha,\beta} = 1/N^{1/2} \sum_{k} a_{\alpha,\beta}(k) \exp[i(k \cdot r_i)],$$

where $k = (k_x, k_y)$ and the lattice constant is taken as unity. $H_0$ is diagonalized by

$$\sum_{\beta} h_{\alpha\beta}(k)d_{\beta\gamma}(k) = E_{\gamma}(k)d_{\alpha\gamma}(k),$$  \hspace{1cm} (4)

where $E_{1}(k) > E_{2}(k) > E_{3}(k) > E_{4}(k)$.

The Dirac point $(k_D)$ is calculated from

$$E_{1}(k_D) = E_{2}(k_D) = \varepsilon_D.$$  \hspace{1cm} (5)

The ZGS is obtained when $\varepsilon_D$ becomes equal to the chemical potential at $T = 0$. The chemical potential $\mu$ is determined from the three-quarter-filled condition, which is given by

$$\frac{1}{N} \sum_{k} \sum_{\gamma} f(E_{\gamma}(k)) = 3,$$  \hspace{1cm} (6)

where $f(e) = 1/(\exp(e - \mu/k_B T) + 1)$ with $T$ being temperature and a Boltzmann constant taken as $k_B = 1$. Using the band energy $E_{\gamma}(k)$, the $T$ dependence of $\mu$ is examined in the next paragraph.

On the basis of four molecules in the unit cell of Fig. 1(a), the matrix element of $h_{\alpha\beta}$ in Eq. 13 is expressed as

$$h_{12}(k) = a_1 + a_2 Y, \quad h_{13}(k) = b_3 + b_2 X, \quad h_{14}(k) = b_1 + b_3 X, \quad h_{23}(k) = b_2 + b_3 X, \quad h_{24}(k) = b_1 + b_2 X, \quad h_{34}(k) = 2a_1, \quad h_{11} = h_{22} = h_{33} = h_{44} = 0,$$

and $h_{\alpha\beta}(k) = h_{\beta\alpha}(k)$, where $X = \exp[i(k_x + k_y)/2]$ and $Y = \exp[i(k_x - k_y)/2]$. Although this model is complicated,
when we use these transfer energies, we find the zero-
gap state (ZGS) composed of Dirac electrons as shown in
the inset of Fig. 1(b), which consistently explains sev-
eral experimental results. For the uniaxial pressure $P_a$
(kbar), which is applied to the $a$ direction, transfer ener-
gies with NN sites, $t = a_1, \ldots , b_4$ (eV), are estimated by
the extended Hückel method based on the crystal struc-
ture analyses with the x-ray diffraction measurement.
Using the overlap integrals estimated from the coordi-
nates of the BEDT-TTF molecules, transfer energies are
obtained by multiplying $-10$ eV corresponding to the
atomic energy. From an interpolation method between
$P_a = 0$ and 2 kbar, the transfer energies are given by
$t(P_a) = t(0)(1 + K_t P_a)$, where $t(0) = a_1(0), \ldots , b_4(0)$
$= -0.028, -0.048, 0.020, 0.123, 0.140, 0.062$, and 0.025,
and $K_t = 0.089, 0.167, -0.025, 0, 0.011$, and 0.032,
respectively. Note that the ZGS is obtained for $P_a > 3$
kbar.

In Fig. 1(b), the chemical potential $\mu$ is shown as a
function of $T$ with a fixed $P_a = 8$ kbar, which decreases
with increasing for $T < 0.01$. At $T = 0$, the chemical
potential is given by $\mu_0 = 0.185$, resulting in the ZGS
as shown on the plane of $\delta k = k - k_D$ (the inset), where
the conduction and valence bands touch at a Dirac point
$k_D = (0.55, 0.25)\pi$. The Dirac cone is tilted almost along
the $k_x$ axis, which gives rise to an anisotropy of the trans-
port property. With increasing $T$, $\mu$ decreases and takes
a slight minimum $\mu \approx 0.1824$ at $T \approx 0.01$. The decrease
of $\mu$ suggests that the hole exists in the valence band be-
low the Dirac point. The choice of $P_a = 8$ kbar is large
as the extrapolation, but could be used considering the
following facts. The Dirac point with increasing $P_a$ is
robust due to a small variation of $k_D$ compared with the
distance from the $\Gamma$ point ($k = 0$), where a pair of Dirac
points merges at $P_a \approx 40$ kbar. Furthermore, the ZGS
has been observed up to $P_a = 10$ kbar in the experiment
of the resistivity.

In Eq. (2), the second term denotes the harmonic
phonon given by $H_P = \sum_q \omega_q b_q^\dagger b_q$ with $\omega_q = v_s |q|$ and $h$
$= 1$. The third term is the e–p interaction with a coupling
constant $g_q$, where

$$H_{e-p} = \sum_{k, \gamma} \sum_q g_q c_\gamma(k + q)^\dagger c_\gamma(k)(b_q + b_q^\dagger), \quad (7)$$

with $c_\gamma(k) = \sum_\alpha d_\alpha\gamma a_\alpha(k)$. The e–p scattering is con-
dered within the same band (i.e., intraband) owing to the
energy conservation with $v \gg v_s$, where $v \approx 0.05 \pi$
denotes the averaged velocity of the Dirac cone. The
last term of Eq. (2), $H_{imp}$, denotes a normal impurity
scattering.

The spectral conductivity $\sigma_{\nu}(\epsilon, T)$ with $\nu = x$ and $y$
is calculated as

$$\sigma_{\nu}(\epsilon, T) = \frac{e^2}{\pi \hbar N} \sum_{k, \gamma} \sum_{\gamma'} v_{\nu, \gamma'}^\gamma(k)^* v_{\nu, \gamma'}(k) \times \frac{\Gamma_{\gamma'}}{(\epsilon - E_{\gamma'}(k))^2 + \Gamma_{\gamma'}^2} \times \frac{\Gamma_{\gamma}}{(\epsilon - E_{\gamma}(k))^2 + \Gamma_{\gamma}^2}, \quad (8)$$

$$v_{\nu, \gamma'}(k) = \sum_{\alpha, \beta} d_{\alpha, \gamma}(k) \frac{\partial h_{\alpha\beta}}{\partial \nu} \beta_{\beta, \gamma'}(k), \quad (9)$$

where $h = 2\pi \hbar$ denotes Planck’s constant. The spectral
conductivity depends on $T$ due to the e-p interaction.
In fact, $\Gamma_{\gamma}$ denotes the damping of the electron of the $\gamma$
band given by

$$\Gamma_{\gamma} = \Gamma + \Gamma_{ph}, \quad (10)$$
where the first term comes from the impurity scattering and the second term corresponding to the phonon scattering is given by Eq. (17).

\[ \Gamma_{\text{ph}}^\nu = C_0 R \times T |\xi_{\gamma,k}|, \quad (11a) \]

\[ R = \frac{\lambda}{\lambda_0}, \quad (11b) \]

where \( \lambda = |q|^{2}/\omega_{q}, \quad \xi_{\gamma,k} = E_\gamma(k) - \mu, \quad C_0 = 6.25\lambda_0/(2\pi^2) \) and \( \lambda_0/2\pi^2 = 0.1 \). \( \lambda_0 \) corresponds to \( \lambda \) for an organic conductor \(^{41,42} \) and \( \lambda \) becomes independent of \( |q| \) for small \( |q| \). \( R \) is taken as a parameter. We take \( \Gamma = 0.0005 \) and \( R=0.5 \) as in the previous paper \(^{41} \), where a choice of \( R=0.5 \) gives a reasonable suppression of the conductivity at high \( T \), and \( \Gamma = 0.0005 \) corresponds to a weak impurity scattering due to \( \Gamma \) being much smaller than \( T \).

In linear response theory, the electric current density \( j = (j_x,j_y) \) is obtained by the electric field \( \mathbf{E} = (E_x,E_y) \) and the temperature gradient \( \nabla T \), i.e., the \( v \) (= \( x \) and \( y \)) component of the current density, is expressed as

\[ j_v = L^\nu_{11}E_v - L^\nu_{12}\nabla_v T/T, \quad (12) \]

where \( L^\nu_{11} \) is the electrical conductivity \( \sigma_{\gamma,25} \) and \( L^\nu_{12} \) is the thermoelectric conductivity.

From Eq. (12), the Seebeck coefficient \( S_v \) is obtained by

\[ S_v(T) = \frac{L^\nu_{12}}{L^\nu_{11}}. \quad (13) \]

As discussed in the introductory part, in terms of Eq. (8), we calculate \( L^\nu_{11} \) and \( L^\nu_{12} \) from the Sommerfeld-Bethe relation,

\[ L^\nu_{11} = \sigma_v(T) = \int_{-\infty}^{\infty} d\epsilon \left( -\frac{\partial f(\epsilon)}{\partial \epsilon} \right) \times \sigma_v(\epsilon,T), \quad (14) \]

\[ L^\nu_{12} = -\frac{e}{\epsilon} \int_{-\infty}^{\infty} d\epsilon \left( -\frac{\partial f(\epsilon)}{\partial \epsilon} \right) \times (\epsilon - \mu)\sigma_v(\epsilon,T), \quad (15) \]

where \( e(>0) \) denotes the electric charge. Noting that \( -\partial f(\epsilon)/\partial \epsilon \) is the even function of \( \epsilon - \mu \), and \( \sigma_v(\epsilon,T) \) in Eq. (14) can be expanded as

\[ \sigma_v(\epsilon,T) = \sigma_v(\mu,T) + \sigma_v'(\mu,T)(\epsilon - \mu) + \frac{1}{2}\sigma_v''(\mu,T)(\epsilon - \mu)^2 + \cdots, \quad (16) \]

Eq. (15) is calculated as

\[ eL^\nu_{12}(T) = \frac{\pi^2}{3}\sigma_v'(\mu,T)T^2 - \frac{7\pi^4}{90}\sigma_v''(\mu,T)T^4 + \cdots, \quad (17) \]

at low temperatures. It is shown later that the sign change of \( S_v(T) \) with decreasing \( T \) comes from that of the first term of Eq. (17).

![Figure 2](image_url)

**Figure 2.** \( T \) dependence of the Seebeck coefficients \( S_x \) (red solid line) and \( S_y \) (red dashed line) for \( P_a = 8 \) kbar, which are compared with electric conductivities \( \sigma_x \) (solid line) and \( \sigma_y \) (dashed line). The e-p coupling is taken as \( R=0 \). The dotted line shows \( S_y \) for \( R=0 \). The inset denotes the magnified \( S_x \) and \( S_y \), which suggest \( S_y \to 0 \) at \( T \to 0 \).

### III. SEEBECK COEFFICIENT

Now we study \( S_v(T) \) using parameters of \( \alpha \)-(BEDT-TTF)\(_{2}\)I\(_3\). The Seebeck coefficient of \( \alpha \)-(BEDT-TTF)\(_{2}\)I\(_3\) under uniaxial pressures provides the following \( T \) dependence. In the present paper, we take \( P_a = 8 \), which shows a sign change, where \( S_x(T) > 0(<0) \) at high temperatures (at low temperatures). Figure 2 shows the \( T \) dependence of the Seebeck coefficient, \( S_x \) and \( S_y \), and the electrical conductivity \( \sigma_v \), where \( \sigma_y > \sigma_x \) for any \( T \) and \( S_x > S_y \) for \( T < 0.008 \). Note that both \( S_x \) and \( S_y \) exhibit the change of the sign at low temperatures. It is found that \( S_x = 0 \) at \( T \approx 0.0009 \) and that \( S_x \) takes a maximum \( \approx 17\mu V/K \) at \( T \approx 0.003 \). At low temperatures given by \( S_x < 0 \), \( S_x \) takes a minimum. Similar behavior is also obtained for \( S_y \), where \( S_y = 0 \) at \( T \approx 0.0015 \) and the temperatures corresponding to the maximum and minimum are almost the same as those of \( S_x \). The relation \( \sigma_y > \sigma_x \), which comes from the tilted Dirac cone [Fig. 1(b)] results in \( S_x > S_y > 0 \). The inset denotes the magnified \( S_x \) at low temperatures. A minimum exists at \( T \approx 0.0002 \) and the extrapolation to lower temperatures suggests \( S_x \approx -T \sigma_x'(\mu,T)/\sigma_x(\mu,T) \) [see Eqs. (16) and (17)]. The interband effect (\( \gamma \neq \gamma' \)) becomes small at low temperatures and the increase of \( \Gamma \) gives a slight reduction of \( S_y \). The decrease of the uniaxial pressure \( P \) reduces the temperature region for \( S_x > 0 \). Note that there is enough range of \( P_a \) for a sign change of \( S_x \), which is in general sensitive to parameters. In fact, \( S_x \) for \( P_a = 6 \) kbar (not shown here) also shows the sign change at \( T \approx 0.0005 \).

In order to comprehend the existence of \( S_k(T) = 0 \),
we examine the spectral conductivity. In Fig. 3, the spectral conductivity $\sigma_{\nu}(\epsilon, T)$ for $\nu = x$ is shown as a function of $\epsilon - \epsilon_{\text{min}}$, where $\sigma_{x}(\epsilon, T)$ takes a minimum at $\epsilon_{\text{min}} = 0.18447$. The minimum is close but lower than that of the Dirac point $\epsilon_D$ ($\epsilon_D - \epsilon_{\text{min}} \approx 0.0005$). $S_x(T) = 0$ occurs when $\mu(T) = \epsilon_{\text{min}}$ at some temperature. A similar minimum is obtained for $\sigma_y(T)(\epsilon, 0.001)$ (dashed line), which is larger than $\sigma_x(\epsilon, 0.001)$. $\sigma_x(\epsilon, T)$ is shown for the fixed $T = 0.0005$, 0.001, and 0.0015, where the width depends on $T$ due to $\Gamma_{ph}^x$ (Eq. 14). The vertical lines denote the corresponding $\mu(T)$, where $\mu(0.001) = \epsilon_{\text{min}}$ and $\epsilon_D = \mu(0)$. Since $\mu(0.0015) < \mu(0.001) < \mu(0.0005)$, $\sigma_x(T) > 0$ for $T < 0.001$ and $\sigma_y(T) < 0$ for $T > 0.001$. From Eq. (17), it turns out that $S_x > 0$ is obtained for $\mu(T) < \epsilon_{\text{min}}$ and $S_y < 0$ is obtained for $T \approx 0.00005 < 0.001$, i.e., for $\mu(T)$ being slightly lower than $\epsilon_{\text{min}}$ due to the second term of Eq. (17). Thus, with decreasing $T$, $S_x(T)$ changes the sign from a positive to a negative one at $\mu \approx \epsilon_{\text{min}}$ corresponding to $\sigma_x(\mu, T) = 0$. Note that the sign change of $S_y$ in Fig. 2 is obtained in the case of $\mu < \epsilon_D$. This fact is different from that of the Hall coefficient where the sign change occurs at $\mu = \epsilon_D$.

Here we note the minimum and maximum of $S_y$ in Fig. 2. Such a behavior is also obtained only for the impurity scattering, i.e., without the $e$-$p$ coupling ($R = 0$). Compared with the dotted line in Fig. 2, $S_x$ at high temperature is reduced by the $e$-$p$ coupling, while $S_x$ at low temperatures ($T < 0.001$) remains the same. We also examined $S_y$ at lower pressures. For $P_b = 6$, it is found that $S_x$ decreases and $S_y$ increases, while the maximum and minimum still exist. The spectral conductivity $\sigma_y(\epsilon)$ shows the existence of the minimum and the $T$ dependence of the chemical potential similar to Fig. 3.

IV. DISCUSSIONS AND SUMMARY

Here, we discuss the relevance of our result to experiments. The temperatures of the sign change and the maximum of $S_x$ ($> 0$) in Fig. 2 are similar to those obtained in the experiment under the hydrostatic pressures. Although this is suggestive, note that the experiment is carried out in the hydrostatic pressure, while our calculation is for the uniaxial pressure. As another aspect, a minimum of $S_x$ at low temperature, suggesting $S_x \rightarrow 0$ at $T = 0$, is an interesting piece of information from our calculation, that should be examined experimentally by decreasing the temperature.

Finally, let us comment on the Seebeck coefficient in the case of the hydrostatic pressure. The previous theory studied the effect of short-range repulsive interactions on the TB model with the transfer energies obtained from the first-principles calculation and showed that the decrease of $T$ leads to the sign change from $S_y > 0$ into $S_y < 0$ at $T \approx 0.0002$. Noting that the Seebeck coefficients are in general sensitive to parameters such as transfer energies and site potentials, we examined $S_x$ and $S_y$ for the following two cases. One is a model used in the previous calculation (but slightly different from that used in Ref.), in which the transfer energies obtained from the first-principles calculations are fixed at a low temperature, and the site potentials obtained from the mean field of the interaction are taken as those at $T = 0$. In this case, we obtained $S_y > 0$ at high temperatures followed by the sign change at low temperatures, while $S_x$ is negative at any temperature.

The other is a model in which the transfer energies are obtained by crystal structure analyses at $P = 1.76$ GPa. Using a choice of site potentials that gives a ZGS, we obtained $S_y > 0$ at high temperatures with the sign change at a temperature being slightly higher than that in the former model, while $S_x$ is negative at any temperature. Thus, we found $S_x < 0$ as a common feature of the above two models, which is inconsistent with the experiment. It remains a future problem to obtain a reliable TB model exhibiting the sign change of $S_x$ for hydrostatic pressures.

In summary, for the $T$ dependence of the Seebeck coefficient of $\alpha$- (BEDT-TTF)$_2$I$_3$, $S_x(T)$ under uniaxial pressures was calculated although there is no experiment at present. We obtained the sign change for both $S_x$ and $S_y$ and clarified the microscopic mechanism in terms of the spectral conductivity $\sigma_y(\mu, T)$. The correspondence of the present theory to the experiment awaits the future measurement of the Seebeck coefficient under uniaxial pressures.
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