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On physical properties of the organic metal (BEDT-TTF)₄Hg_{3- δ}Cl₈

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Résumé. — Nous présentons des études de résonance paramagnétique sur les électrons de conduction du $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$, complétées par des mesures de susceptibilité paramagnétique statique, de force thermoélectrique et d'absorption IR. Nous avons montré que les interactions d'échange fortes conduisent à un spectre RPE simple, élargi par le couplage spin-orbite. La bidimensionalité du système de spin a été également confirmée. Des anomalies ont été observées près de 80 K pour certaines propriétés physiques ; nous supposons qu'elles sont provoquées par une déformation du réseau des ions Hg. Nous avons démontré que l'anisotropie spectrale (RPE, IR) n'est pas toujours comparable à l'anisotropie de transport électrique du métal organique.

Abstract. — Conduction electron spin resonance study is presented together with static susceptibility, thermopower and IR absorption measurements for $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$. It is shown that the strong exchange interactions lead to a single ESR line broadened by the spin-orbit coupling; the role of two-dimensionality of the salt is emphasized. The two-dimensionality of the spin ordering is also stated. The anomalies of the ESR parameters near 80 K may be associated with a phase transition caused probably by the deformation of the incommensurate Hg sublattice. Our study confirms that the anisotropy of the spectral (IR, ESR) properties does not necessarily agree with that of the transport properties of the organic metal.

1. Introduction.

Molecular metals based on the donor bis (ethylenedithio) tetrathiafulvalene, BEDT-TTF, have recently attracted a great deal of interest. This family of organic conductors exhibits a considerable diversity in crystallographic phases and physical properties [1]. Some of the materials are metallic to low temperatures, some of the others were found to be superconductors under hydrostatic pressure [2] or under ambient conditions [3]. Apart form the superconductivity, these materials are interesting because they seem to be much more two-dimensional (2-D) than any other known organic conductors.

Recently, a crystal with a new type of acceptor is added to the $(BEDT-TTF)_n X$ family: the material named $(BEDT-TTF)_4Hg_3Cl_8$ was obtained by Lyubovskaya *et al.* [4]. The coexistence of two incommensurate lattices of BEDT-TTF and Hg [5] in this organic metal permits us to use a more correct formula of the salt: $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$.

In the present paper we report the study of the electron spin resonance (ESR) phenomena of the $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ crystals; the ESR investi-

gations were complemented by static magnetic susceptibility, thermopower and IR measurements.

The crystals of (BEDT-TTF)₄Hg_{3 - δ}Cl₈ were prepared electrochemically from tetrahydrofurane solution. The characteristic morphology is of a parallelogram in the *ab*-plane ; their diagonals are of the order of 0.5-2 mm. The BEDT-TTF cations form sheets in the *ab* plane. The most prominent intermolecular S-S contacts (~ 3.49 Å) are found between BEDT-TTF dimer units of the sheets. The molecular ordering and the intermolecular contacts give the compound a two-dimensional character which affects the physical properties. The salt becomes superconducting at about 1.8 K under a pressure greater than 12 kbars [4]. The 2-D structure as well as the knowledge of the electrical d.c. conductivity [4] and polarized reflectance [6] of the $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ crystals stimulate us to study the anisotropic behaviour of the ESR g-factor and the linewidth, and thus to calculate the basic ESR parameters. We intend to use the thermopower characteristics and IR absorption measurements to analyse the scattering and the coupling processes. Our final goal is to better understand the physical properties, especially the spin ordering, of the $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ salt.

2. Experimental.

All ESR and thermopower measurements were performed on single crystals. Under the assumption that two of the principal axes, a and b, are always located in the plane of the plate-like crystal, the axes were simply identified from the geometry. The first, called b, is close to the direction of molecular stacking and the short diagonal of the parallelogram; the second, called a, is perpendicular to the BEDT-TTF molecular stacks, and is close to the direction of the long diagonal line. The skin depth estimated for 9.1 GHz is between 1.3 and 1.7 mm at room temperature or between 0.7 and 0.9 mm at about 10 K, for the electromagnetic wave propagation in the plane direction, i.e. it is comparable with or slightly larger than the sizes of crystals employed here. Therefore, no skin effect is expected for this case. Because the high electrical anisotropy of the salt, the skin depth for wave propagation square with the plane is about 3 orders of magnitude larger than the crystal size in this direction; thus no skin effect is expected, either.

The ESR measurements were carried out with a SE/X 2544 X-band spectrometer made bv RADIOPAN (Poland). The temperature, within the range from about 4 to 400 K, was controlled by cooling a resonator with a flux of gaseous helium pumped via the heat exchanger. A symmetric and Lorentzian ESR line observed between about 50 K and 400 K permits us to estimate the paramagnetic susceptibility simply from the parameters of the line in this temperature range. Below 50 K the Dysonian signal is observed but a ratio A/B between the maximum and minimum of the derivative of absorption approximates 1.0; only at the lowest temperatures (T < 10 K) A/B comes up to 1.2. Thus the paramagnetic susceptibility estimated simply from the line parameters at this temperature range is loaded with a measuring error smaller than 20-25 %. The angular dependences of the g-factor and the peak-to-peak linewidth were measured at room temperature only, for three perpendicular crystal orientations.

The static magnetic susceptibility was measured by the Faraday method from 4 K to about 300 K whereas the thermoelectric power of the crystal was measured by the conventional gradient less than 1 K, from 370 K to about 100 K. The electrical contacts of silver wires were assured by wetting them with a silver paste-paint. The ends of the crystals were covered with aquadag to remove the chemical reaction. The thermopower was measured along orientations close to both the *a* and *b* axes. Spectral investigation of the powdered salt with KBr were performed in the intermediate IR region using SPECORD M80 spectrophotometer, at ambient temperature only.

3. Results and discussion.

3.1 ESR g-factor and the linewidth. — ESR observations have played a significant role in elucidating the electronic structure of the electrically conductive organic CT compounds. The electronic g-factor, the lineshape and/or the line-width as well as the dependence of these quantities on sample orientation and temperature are usually a main source of information. Unfortunately, the electronnuclear hyperfine splittings are lost in these salts to a combination of spatial averaging due to the delocalization of the electrons over many molecules and spin averaging due to spin - exchange interactions between the electrons. On the other hand intermolecular interactions and the resulting quasi 2-D character are observed in $(BEDT-TTF)_n X$ materials as being the fundamental mechanisms whereby spin-orbit scattering occurs, and broadens the ESR line.

Figure 1 shows the angular dependence of the gfactor for $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ at room temperature. The sample was rotated around the orthogonal axes a, b and c. A static field and an oscillating magnetic field, which are perpendicular to each other, were applied perpendicular to the rotating axes. The *g*-value is almost isotropic for the *ab* rotation (round c axis) and equals about 2.0017. The g-value anisotropy for rotations around a and b axes shows a maximum at 2.0130 and 2.0140, respectively, and a common minimum at about 2.0015. These results suggest that the g-tensor of (BEDT-TTF)₄Hg_{3- δ}Cl₈ is axially symmetric around the c axis of the crystal. The angular dependence of the gfactor is qualitatively in agreement with that reported by Sugano *et al.* [7] for α -(BEDT-TTF)₂I₃.



Fig. 1. — Angular dependence of the g-factor at room temperature. Calculated g-values for sample rotation around the a, b and c axes are shown by solid lines.

Usually, for the $(BEDT-TTF)_n X$ compounds, the angular dependence of the *g*-value of the crystal is well interpreted by taking into account the principal

values and axes of the g-tensor of the BEDT-TTF cation radical and their ordering in the crystal. To determine the principal values and axes of the g-tensor, we carried out a least-squares fitting to the equation

$$g^{2} = \sum_{i,j}^{n} g_{i,j}^{2} l_{i} l_{j}$$
(1)

where l is the direction cosine of the principal axis.

In the $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ system the molecule-packing modes have an intricate structure [4]; the molecule orientations as well as overlapings depend on the site. There are two typical molecular orientations in the crystal lattice, which are not described by the same direction cosines even in a very rough approximation. The existence of magnetically inequivalent chains of molecules due to different orientations of the molecular plane of BEDT-TTF would split the ESR absorption. However, only a single absorption line is observed in the whole temperature range examined here. This indicates that there is a strong exchange coupling between the molecules which average out the absorption. In this case, according to Sugano et al. [7] the variation of the g-factor as a function of rotation angle may be described by an average of the g-values of the BEDT-TTF species over the unit cell.

We assume that the molecular axes of the average BEDT-TTF molecule in $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ coincide with the monoclinic cell axes a, b and c. The solid lines in figure 1 denote the calculated g-values which are fitted to the experiments with the fitting parameters g_{ij} using equation (1). Since the g-tensor is axially symmetric, only $g_1 = 2.0135$ is obtained; the values of g_2 and g_3 are averaged at 2.0017 in the ab plane. The directions of the principal axes of the g-tensor are identical to the directions of the molecular axes of the average BEDT-TTF molecule in the crystal.

Figure 2 shows the angular dependence of the peak-to-peak linewidth ΔH observed at room temperature. In our crystal, a Lorentzian line-shape was



Fig. 2. — Angular dependence of the peak-to-peak linewidth at room temperature.

observed in the whole temperature range and for all orientations examined here. This angular dependence is qualitatively similar to that of the g value.

 $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ shows a broad ESR line, the maximum linewidth being as large as 11 mT. Similar linewidths were observed for α -(BEDT-TTF)₂I₃ (12.3 mT [7]). According to Sugano et al. [7] this would be due to the strong spinorbit coupling caused by the interaction of the triiodate with BEDT-TTF. Strong 2-D character of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ as well as the spin-orbit coupling caused by the interaction with $Hg_{3-\delta}Cl_8$ anion would be responsible for the broad ESR line in the salt. The large linewidth together with the gvalue indicates that there is considerable interaction of the $Hg_{3-\delta}Cl_8$ anion with BEDT-TTF.

Figure 3 shows the temperature dependence of the linewidth, ΔH and the *g*-factor, for randomly oriented single crystals of (BEDT-TTF)₄Hg_{3- δ}Cl₈.



Fig. 3. — Temperature dependence of the g-factor (•) and the linewidth, ΔH , (O), for the single crystal of (BEDT-TTF)₄Hg₃₋₈Cl₈.

The ΔH is constant for T > 80 K whereas it decreases nearly linearly with T, below about 80 K. A similar temperature dependence of the ΔH was observed e.g. by Samoano et al. [15] for $(TTF)_{12}(SCN)_7$, by Sugano et al. [7] and Rothaemel et al. [16] for α -(BEDT-TTF)₂I₃ and by Enoki *et al.* [17] for **BEDT-TTF** the range with ClO_4 . In 4 K < T < 80 K, ΔH of (BEDT-TTF)₄Hg_{3- δ}Cl₈ decreases more than 5 times. The beginning of the ΔH decrease is correlated with the maximum of the paramagnetic susceptibility (Fig. 4) and is close to the minimum of thermopower and an anomaly of the electrical conductivity observed by Lyubovskaya et al. [4]. We think that the abrupt change of the parameters describing the spin ordering, observed at about 80 K, is connected with a phase transition of the crystal.

At present it is difficult to characterize this anomaly more precisely. One of the possibilities is a change of organization of the anion sublattice,



Fig. 4. — Static paramagnetic susceptibility (O) and spin susceptibility (\bullet) of (BEDT-TTF)₄Hg₃₋₈Cl₈.

probably a deformation of the Hg chains. This hypothesis seems to be probable as the changes of the physical properties of the salt at the transition temperature are rather soft.

The peak-to-peak linewidth, ΔH , is almost constant for 80 K < T < 330 K; whereas a linebroadening effect is expected for organic metals. In our case it is possibly compensated by the linenarrowing due to the increase of the exchange interaction with temperature. The high value of ΔH is due mainly to the strong spin-orbit coupling; the role of the interstack interactions and the resulting partial 2-D character as well as incommensurate Hg lattice is significant, also.

Below the phase transition temperature, ΔH decreases because the spin-orbit coupling is weakened probably by the deformation of the Hg sublattice. It is possible that the Hg lattice becomes commensurate with the BEDT-TTF sublattice in the low T phase. This hypothesis will be studied.

The g-factor is temperature-independent in the whole T-region, within experimental accuracy. It suggests that the delocalized conduction electrons remain in the same local field above and below the phase transition.

3.2 THE SPIN SUSCEPTIBILITY. — The static paramagnetic susceptibility, corrected for core diamagnetism, and the normalized, at 250 K, spin susceptibility, found as an integrated ESR signal, are shown in figure 4. Nearly in the whole temperature range the spin susceptibility follows precisely the static one. For this reason it was concluded, similarly as Scott *et al.* [8] have shown for other organic metals, that the ESR signal corresponds to the entire conduction-electron susceptibility.

The paramagnetic ESR susceptibility of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ increases rather weakly with decreasing temperature, similar to the increase

observed by Venturini *et al.* [9] but contrary to Sugano *et al.* [7] for α -(BEDT-TTF)₂I₃. The temperature dependence of χ is not as in the metalliclike salt of BEDT-TTF cation. It resembles more a Bonner-Fisher [8] behaviour than a simple Pauli model. The thermal variations of the paramagnetic part of the susceptibility of (BEDT-TTF)₄Hg₃₋₈Cl₈ can be described by a Bonner-Fisher model with an exchange integral, $J = 6.4 \times 10^{-3}$ eV and a parameter, $\gamma \approx 0.7$. This unusual behaviour of the salt arises probably from the strong spin-orbit coupling caused by the interaction of the Hg₃₋₈Cl₈⁻ anion with BEDT-TTF⁺ cation.

On the other hand the spin susceptibility seems to characterize the Pauli spin susceptibility at high temperatures (T > 220 K). The Pauli spin susceptibility for a free electron gas is

$$\chi_{\rm p} = \mu_{\rm B}^2 N(E_{\rm F}) \tag{2}$$

where $N(E_{\rm F})$ is the spin density states at the Fermi level. For the case of noninteracting electrons (the simplest model of one-electron tight-binding band with a bandwidth 4 t), at T = 0 K, χ , for one kind of stack, is given by the simple Pauli susceptibility

$$\chi_{\rm p}(0) = \frac{N_0 \,\mu_{\rm B}^2}{\pi t \,\sin\,\left(1/2\,\pi\rho\right)}\,.\tag{3}$$

For the bandwidth about 0.27 eV (according to our thermopower measurements) and putting $\rho \approx 0.5$, the susceptibility of (BEDT-TTF)₄Hg₃₋₈Cl₈ is about 2.1×10^{-4} emu/mole. This value is smaller than the measured static magnetic susceptibility, at room temperature, which is 8.2×10^{-4} emu/mole. According to Cooper [10], at room temperature the susceptibility of most organic conductors is enhanced by a factor of 2-5 over the Pauli paramagnetism expected for noninteracting electrons using a reasonable value for the bandwidth.

The temperature and angular study of the ESR of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ confirms that the ESR signal corresponds to the conduction electrons. The single absorption line indicates a strong exchange coupling between the stacks. The strong and *T*-independent spin-orbit coupling caused by the interaction of the inorganic anions and organic cations is also stated. The 2-D of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ and the axial symmetry of its *g*-tensor are found ; then are expected from the crystal structure of the compound.

3.3 THERMOPOWER. — The experimental thermopower results of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ along both the *a*-axis (S_a) and *b*-axis (S_b) are shown in figure 5. In the *a*-direction negative values of *S* falling linearly with temperature at T > 180 K are found. We note a slight curvature at lower *T*. Small positive values of S_b are observed above about



Fig. 5. — Thermopower along the a- and b-axes vs. temperature.

330 K; in the lower *T*-region the thermopower is negative and decreases nonlinearly with temperature. A small increase of S_b is noticed below about 130 K. At high temperatures both components show a slope dS/dT not only qualitatively similar for the two compounds but also nearly equal in magnitude, about 0.17 μ V/K². However the thermopower tensor, represented by S_a and S_b is anisotropic ($S_a/S_b =$ -1.6 at 360 K) and highly *T*-dependent; the two components even have opposite signs above 330 K. The room temperature values of *S* are -20μ V/K and -6μ V/K along the *a* and *b* axes, respectively.

The observed thermopower of (BEDT-TTF)₄Hg_{3- δ}Cl₈ exhibits unusual characteristics of being nonlinearly *T*-dependent at low temperatures and reveals a distinct anisotropy. In the high *T*-region the temperature dependence represented by dS/dT is, however, nearly isotropic. The similarity of the thermopowers of (BEDT-TTF)₄Hg_{3- δ}Cl₈ and β -(BEDT-TTF)₂I₃ is noticed.

The thermopower of the ambient pressure organic superconductor β -(BEDT-TTF)₂I₃ has been studied by Mortenses *et al.* [11]. They suggested that it is physically meaningful to separate the *ab*-plane thermopower into

$$S(T) = S_1(T) + S_0$$
(4)

where S_1 is *T*-dependent and nearly isotropic within the plane and S_0 is anisotropic and basically *T*independent. Starting from Mott's formula it was shown that it is possible to have an isotropic bandcontribution to *S* and an anisotropic scattering-contribution, namely if the Fermi surface is nearly spherical, in D dimension, and a relaxation rate, τ , is anisotropic. The separation of the experimental S into an isotropic term, S_1 , and an anisotropic term, S_0 (Eq. (4)) seems accordingly to reflect the bandterm and the scattering-term, respectively. The former term along an axis α becomes :

$$S_{B\alpha} = \frac{\pi^2 K_B k_B T}{6 e t_{\alpha}}, \quad \alpha = a, b$$
 (5)

where t_{α} is a transfer integral. The above equation is valid for crystals with 2-D free electron gas and with a 3/4-filled tight binding band.

The crystal structure of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ [4] as well as the polarized reflectance [6] and our ESR studies show its 2-D character. This is why we can decompose the experimental S_a and S_b values according to equation (4) and evaluate t_a from equation (5). The $S_1(T)$ and S_0 terms are 0.19 T and -78, respectively, for the *a*-axis and T > 180 K; the same for the *b*-axis and T > 250 K are : 0.16 T and -54. The band-term thermopower results in transfer integrals which are : $t_a = 0.062 \text{ eV}$ and $t_b = 0.074 \text{ eV}$. The good agreement between the above integrals and the t-values found by Jacobsen et al. [12] for $(BEDT-TTF)_2I_3$ type salts and by Kuroda et al. [13] for α -(BEDT-TTF)₂I₃ and salts with ClO_4^- and ReO_4^- anions confirms the validity of the model used above.

A comparison of the scattering terms suggests an anisotropy of the scattering. Below T = 180 K alone *a*-axis and below T = 250 K alone *b*-axis, additional positive contributions to *S* appear. These contributions are highly anisotropic and it is likely that they reflect changes in the scattering mechanism. It is not understood why these changes appear at temperatures as high as 250 K in the *b* direction.

3.4 IR ABSORPTION. — The IR absorption spectrum is the superposition of a broad contribution of intraband transitions which is possibly nearly isotropic, and a relatively sharp bands due to the inter-band transitions strongly polarized. Kaplunov's investiof the polarized reflectivity gation of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ [6] has shown that IR spectra are nearly isotropic in the *ab*-plane. In both *a* and b directions, the bands due to the interaction of molecular vibrations with conducting electrons are observed. The electron-vibrational interaction was described in terms of the phase phonon model; values of the electron-phonon coupling constants are typical for the case of weak electron-phonon interactions [6].

According to Tajima *et al.* [14] the anisotropy of the IR band does not necessarily agree with that of d.c. conductivity. This is the case of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$.

From our absorption IR measurements (Fig. 6) it is possible to determine the frequency of CT band and the normal vibrations of the salt. The former is $3\,450\,\mathrm{cm}^{-1}$, at room temperature and the latter are : 2 965 (w), 2 925 (m), 2 855 (w), 1 445 (s), 1 410 (s), 1 390 (w), 1 330 (vs), 1 270 (s), 1 260 (m), 1 175 (m), 1 125 (w), 1 010 (w), 1 020 (w), 975 (vw), 960 (vw), 940 (vw), 925 (w), 895 (m), 845 (vw), $800 \text{ (vw)}, 685 \text{ (w)}, 490 \text{ (w)}, 465 \text{ (m)}, 440 \text{ (s) } \text{cm}^{-1}$ relative intensities are denoted : vs (very strong), s (strong), m (medium), w (weak), vw (very weak). The frequencies of molecular vibrations of the salt are nearly the same as the frequencies found by Kaplunov et al. in the spectrum of BEDT-TFF; some of the bands can be identified as the bands seen the reflectivity spectrum in of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ [6].



Fig. 6. — IR absorption spectrum of the powdered $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ salt with KBr at room temperature.

The strong intraband transition at about 3450 cm^{-1} permits us to estimate the width of the conducting band to be less than 0.43 eV. This estimate is in rough agreement with 4t = 0.27 eV found from the thermopower analysis.

4. Conclusions.

The studies of the ESR, the static paramagnetic susceptibility, the thermopower and the IR spectra of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ have provided some new insights into this material. It was shown that the physical properties of this organic metal are determined mainly by the delocated electrons. The parameters of the ESR line testify that the signal corresponds to the conduction electrons; it is confirmed by static susceptibility study.

The delocation of the electrons over many molecules and the spin averaging due to spin-exchange interactions between the electrons eliminate the electron-nuclear hyperfine splittings. Similarly, the strong exchange coupling between the molecules averages out the absorption and leads to the single absorption line. The broad ESR line, about 11 mT at room temperature, is probably due to the strong spin-orbit coupling caused by the interaction of $Hg_{3-\delta}Cl_8^{2-}$ anion with BEDT-TTF⁺ cation; the role of two-dimensionality as well as incommensurability of the lattices of cation and Hg is also noticed. The spin-orbit coupling is approximately *T*independent at T > 80 K. Below about 80 K the decrease of ΔH is observed. Its beginning is correlated with anomalies of other physical properties of the salt. We understand these changes as a result of the phase transition caused probably by the deformation of the Hg sublattice.

The angular study of the ESR of the salt shows a strong 2-D character of the spin ordering. The gtensor is axially symmetric with $g_1 = 2.0135$ and with $g_2 = g_3 = 2.0017$; the latter values were determined in the ab plane. Small anisotropy of the IR reflection spectra confirms the 2-D character of the compound. The isotropy in the plane *ab* does not occur in the electric transport measurements. The anisotropy of the thermopower stated on the ground of the results along both the a- and b-axes is large and the components S_a and S_b even have opposite signs above 330 K. The analysis of the data revealed two components of the Seebeck coefficient : $S_1 - T$ -dependent and nearly isotropic within the ab plane, and S_0 , anisotropic and basically T-independent. The former represents the band contribution while the latter, the scattering one. It is found that the band structure of $(BEDT-TTF)_4Hg_{3-\delta}Cl_8$ is nearly isotropic in two dimensions with effective transfer integrals of 0.062 eV-0.074 eV.

The results mentioned above show that the anisotropy of the transport properties of the compound does not necessarily agree with that of the spectral (IR, ESR) properties. The prominent bands found in the IR region are mainly determined by the conduction electrons-intramolecular vibrations coupling. The ESR spectrum is defined by the spinexchange interactions. The isotropic band contribution to the thermopower has the properties determined by the band structure of the compound. On the contrary, the anisotropic contribution to S is due to the scattering process. This explains why the anisotropy of the spectral and transport properties is not the same.

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