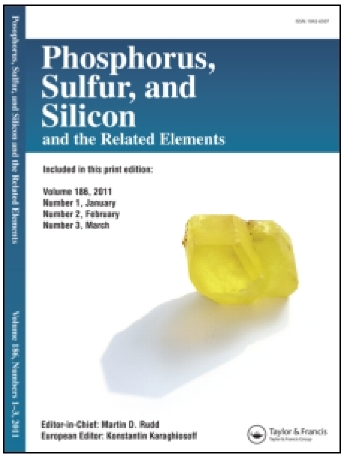

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Fermi Surface Topology and Electronic Structures of Two- Dimensional Organic Conductors Based on Bedt-TTF and Mdt-TTF

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FERMI SURFACE TOPOLOGY AND ELECTRONIC STRUCTURES OF TWO-DIMENSIONAL ORGANIC CONDUCTORS BASED ON BEDT-TTF AND MDT-TTF

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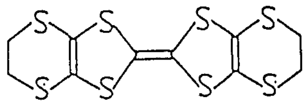
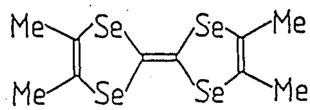
Abstract The electronic structures of two series of quasi-2D organic metals, α-$(ET)_2MHg(SCN)_4$ (M=K,NH_4) and θ-$(MDT\text{-}TTF)_2Au(CN)_2$, are discussed based on the resistivity measurements using high magnetic field up to 35T or an application of pressure.

INTRODUCTION

A variety of organic radical cation salts like those derived from TTF and selenium and tellurium analogs has extended the field of chemistry and physics. Recently, new BEDT-TTF (ET,Fig.1) based conductors, κ-$(ET)_2Cu[N(CN)_2]X$ (X=Cl,Br) were synthesized with the superconducting transition temperatures above 11K. The manifested feature of ET superconductors is the strong two-dimensionality (2D) in their electronic structures compared to the quasi-1D of TMTSF (Fig.2) superconductors. For conventional low-dimensional organic conductors, the strong correlation among conduction electrons causes the antiferromagnetic spin fluctuation, and the spin-density-wave (SDW) ground state is stabilized. Therefore, high-dimensionality is necessary to obtain superconductor. However, for many organic superconductors, the superconducting state is just situated in the proximity of the SDW state, because both states are attributed to high electron density of states. So it is interesting to explore the organic conductors in the region of the dimensional crossover, and the dimensionality examination (Fermi surface topology) is of special importance. In this study, we discuss the Fermi surfaces of α-$(ET)_2MHg(SCN)_4$ (M=K,NH_4) and θ-$(MDT\text{-}TTF)_2Au(CN)_2$ (MDT-TTF, Fig.3).





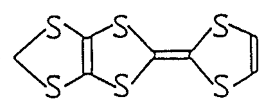


FIG. 1 BEDT-TTF FIG. 2 TMTSF FIG. 3 MDT-TTF

THE α-$(ET)_2MHg(SCN)_4$ (M=K,NH_4) SERIES

We have carried out the magnetoresistance measurements on the novel organic conductor α-$(ET)_2MHg(SCN)_4$ (M=K,NH_4) under high magnetic fields up to 35T. The experimental details are described in references [1][2]. These salts show metallic behavior in conductivity down to 0.5 K and the NH_4 salt shows superconducting transition below about 1 K.[3] The tight-binding band calculations by T. Mori have indicated the presence of both one-dimensional open Fermi surfaces and two-dimensional closed cylindrical one for these salts.[4] We observed the large Shubnikov-de Haas (SdH) oscillations (Fig.4), which indicate the existence of two-dimensional Fermi surfaces. The cross-sectional areas of the two-dimensional Fermi surfaces are estimated as 16% for the K salt and 13% for the NH_4 salt of the first Brillouin zone.

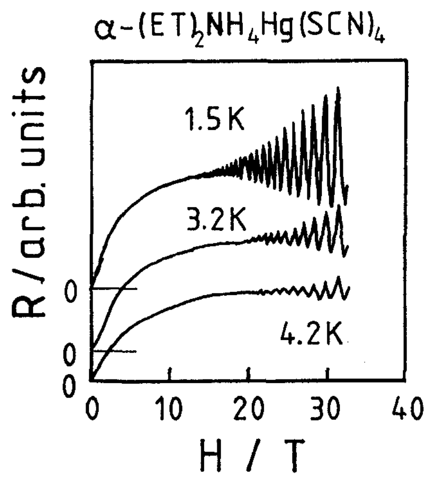


FIG. 4 The Shubnikov-de Haas (SdH) oscillations of organic superconductor α-$(ET)_2NH_4Hg(SCN)_4$.

THE θ-$(MDT\text{-}TTF)_2Au(CN)_2$ SALT

MDT-TTF is an asymmetric donor, of which the salt is capable to form a variety of structures. A superconductivity has been observed in κ-$(MDT\text{-}TTF)_2AuI_2$ which has a two-dimensional electronic structure.[5] The MDT-TTF molecule has partial structures of TTF and BMDT-TTF, which suggests that the polarization of MDT-TTF is lower than that of BEDT-TTF. The low degree of molecular polarizability increases the on-site Coulomb repulsion; U, which is proportional to the enhancement factor of the spin susceptibility attributed to exchange interaction. And the large U enhances the k-dependent magnetic response function, which causes the spin fluctuation. So the competition between the superconducting state and SDW states are expected. However no SDW instability has been detected in the MDT-TTF salts so far.

Thin plate-like red-brown crystals of θ-$(MDT\text{-}TTF)_2Au(CN)_2$ were prepared by a conventional electrochemical method. The experimental details are described in reference [6]. MDT-TTF molecules form a segregated column in which the molecules are ordered in one direction. The donor columns construct a

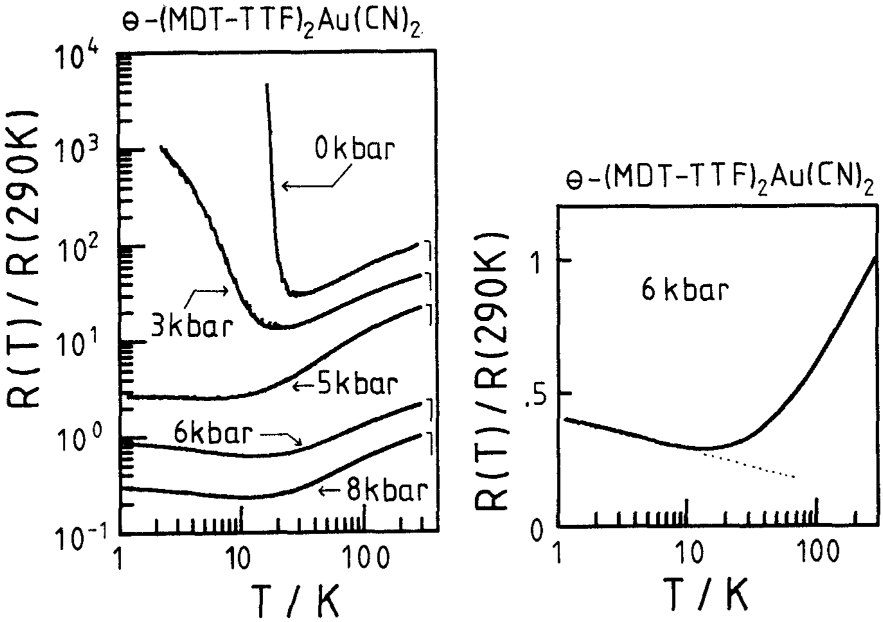


FIG. 5 Temperature dependence of the normalized resistivity of θ-$(MDT\text{-}TTF)_2Au(CN)_2$ under pressure.

conducting layer which is sandwiched between the insulating anion layers. However, the tight intermolecular atomic contacts within and between stacks indicate the substantial two-dimensional character of this salt. The temperature dependence of the electronic resistivity under pressure is shown in Fig. 5. This salt is metallic down to 50 K at ambient pressure followed by a metal-insulator transition. Above 5 kbar a metallic state is stabilized down to 1.2 K but no superconducting transition was detected so far.

The temperature dependence of the spin susceptibility (χ_{spin}) and that of the peak-to-peak width ΔH are shown in Fig. 6. The weak temperature dependence of the spin susceptibility from room temperature down to 20 K is consistent with the metallic behavior. The magnitude of spin susceptibility at room temperature, 5 x 10^{-4} emu/mole, is very high but close to that of a superconductor κ-$(MDT\text{-}TTF)_2AuI_2$,[7] indicating that the electron density of states is almost the same between them. The weak decrease of ΔH from room temperature down to 100 K is explained by Elliott mechanism (ΔH ∝ Resistivity) which describes the conduction electron relaxation. The remarkable ΔH broadening and χ_{spin} decrease observed below 60 K suggest the presence of antiferromagnetic spin fluctuation. At 20 K, at which metal-

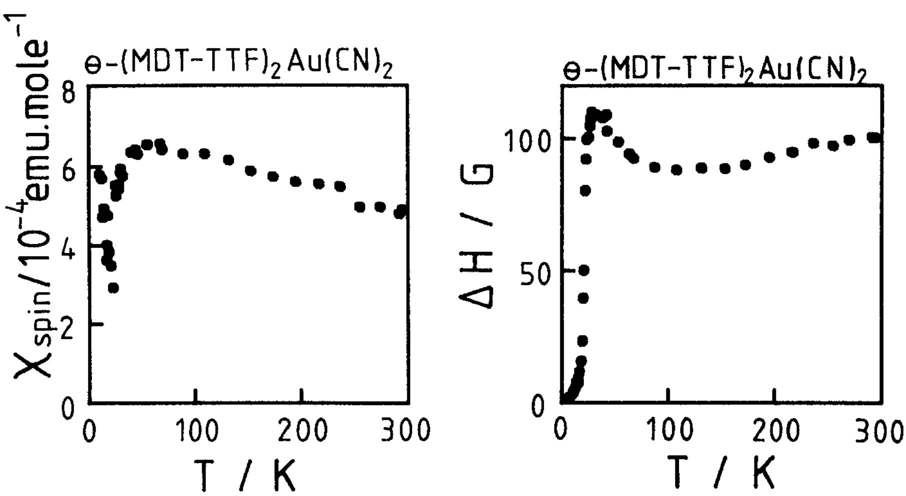


FIG. 6 Temperature dependence of the spin susceptibility; χ_{spin} and peak-to-peak linewidth; ΔH for θ-$(MDT\text{-}TTF)_2Au(CN)_2$.

insulator transition is observed in the resistivity measurements, the EPR signal disappears suddenly and is replaced by a new signal with very narrow linewidth (< 10 G), which is ascribed to localized spin.

The disappearance of main signal suggests the occurrence of either CDW or SDW state. The divergence of the nuclear spin relaxation rate and the broadening of the line width in the proton NMR measurements show that the nature of the transition is magnetic. Therefore the NMR measurements have proved that the ground state is SDW state.[8]

DISCUSSION

Recent proton NMR spin-lattice relaxation measurements of θ-(MDT-TTF)$_2$Au(CN)$_2$ under pressure indicate that a spin-fluctuation exists even in the metallic state. The same behavior has also been observed for α-(ET)$_2$KHg(SCN)$_4$. The KHg(SCN)$_4$ salt shows anomalous positive magnetoresistance and static magnetic susceptibility anisotropy below 8 K, suggesting the presence of the antiferromagnetic spin correlation.[9] The fact that θ-(MDT-TTF)$_2$Au(CN)$_2$ undergoes an SDW state in spite of strong two-dimensionality suggests that the Fermi surface of this salt is opening with fairly warping. As mentioned above, α-(ET)$_2$KHg(SCN)$_4$ salt possesses an open Fermi surface, which is fairly warped. It seems that a fairly warped Fermi surface plays a crucial role in the antiferromagnetic instability.

The resistivity of θ-(MDT-TTF)$_2$Au(CN)$_2$ salt under pressure increases slightly below about 10 K, which seems log(T) dependence. A similar behavior has been observed in the (ET)$_2$Cu$_5$I$_6$ salt[10] and (TSeT)$_2$Cl[11] [where TSeT denotes tetraselenotetracene]. The band calculations of the Cu$_5$I$_6$ salt[12] and (TSeT)$_2$Cl[13] have indicated the presence of warped open Fermi surfaces for these salts. The imperfect nesting of the Fermi surfaces in quasi-two-dimensional conductors and the strong correlation of electrons due to the high density of states may cause the resistivity increase at low temperatures.

In conclusion, α-(ET)$_2$MHg(SCN)$_4$, θ-(MDT-TTF)$_2$Au(CN)$_2$, and related salts, which are the organic conductors in the region of the dimensional crossover, show anomalous physical properties associated with the warped open Fermi surfaces, such as the magnetoresistance. Furthermore, the high density of states and the rather high-dimensionality indicate that these salts are in the neighborhood of superconductors.

ACKNOWLEDGMENT

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REFERENCES

1. T. Osada, R. Yagi, A. Kawasumi, S. Kagoshima, N. Miura, M. Oshima, and G. Saito, Phys. Rev., B41, 5428(1990).
2. T. Osada, A. Kawasumi, R. Yagi, S. Kagoshima, N. Miura, M. Oshima, H. Mori, T. Nakamura, and G. Saito, Solid State Commun., 75, 901(1990).
3. a. H. H. Wang, K. D. Carlson, U. Geiser, W. K. Kwok, M. D. Vashon, J. E. Thompson, N. F. Larsen, G. D. McCabe, R. S. Hulscher, and J. M. Williams, Physica C, 166, 57(1990).
   b. H. Mori, S. Tanaka, K. Oshima, M. Oshima, G. Saito, T. Mori, Y. Maruyama, and H. Inokuchi, Solid State Commun., 74, 1261(1990).
4. T. Mori, H. Inokuchi, H. Mori, S. Tanaka, M. Oshima, and G. Saito, J. Phys. Soc. Jpn., 59, 2624(1990).
5. G. C. Papavassiliou, G. A. Mousdis, J. S. Zambounis, A. Terzis, A. Hountas, B. Hilti, C. W. Mayer, and J. Pfeiffer, Synth. Metals, 27B, 379(1988).
6. T. Nakamura, G. Saito, T. Inukai, T. Sugano, M. Kinoshita, and M. Konno, Solid State Commun., 75, 583(1990).
7. P. Delhaes, J. Amiell, S. Flandrois, L. Ducasse, A. Fritsch, B. Hilti, C. W. Mayer, J. Zambounis, and G. C. Papavassiliou, J. Phys. (Paris), 51, 1179(1990).
8. K. Kanoda, Y. Kobayashi, T. Takahashi, T. Inukai, and G. Saito, Phys. Rev., B42, 8678(1990).
9. T. Sasaki, N. Toyota, M. Tokumoto, N. Kinoshita, and H. Anzai, Solid State Commun., 75, 93(1990).
10. L. I. Buravov, A. V. Zvarykina, M. V. Kartsovnik, N. D. Kushch, V. N. Laukin, R. M. Lobkovskaya, V. A. Merzhanov, L. N. Fedutin, R. P. Shibaeva, and E. B. Yagubskii, Zh. Eksp. Teor. Fiz., 92, 594(1987).
11. I. F. Schegolev, P. A. Kononovich, M. V. Kartsovnik, V. N. Laukhin, S. I. Pesotskii, B. Hilti, and C. W. Mayer, Synth. Metals., to be submitted.
12. T. Mori and H. Inokuchi, in The Physics and Chemistry of Organic Superconductors, edited by G. Saito and S. Kagoshima (Springer, Berlin, 1990), p 204.
13. I. F. Schegolev and E. B. Yagubskii, in Extended Linear Chain Compounds, edited by J. P. Miller (Plenum Press, New York and London, 1982), vol 2, p 385.