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Antiferromagnetic ordering of the incommensurate organic superconductor (MDT-TS)$(AuI_2)_{0.441}$ with a high spin-flop field

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The magnetic torque and high-pressure transport properties of the incommensurate organic superconductor (MDT-TS)$(AuI_2)_{0.441}$ are investigated, where MDT-TS is 5*H*-2-(1,3-diselenol-2-ylidene)-1,3,4,6-tetrathiapentalene. The magnetic torque provides unquestionable evidence of an antiferromagnetic ordered state with a high spin-flop field of 6.9 T at ambient pressure. Hydrostatic pressure suppresses this insulating state and induces superconductivity, where the superconducting transition temperature rises to the maximum, 4.9 K at 1.27 GPa; the superconducting phase exists between 1.0 and 1.8 GPa above 1.5 K.

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I. INTRODUCTION

In the materials with strongly correlated electrons such as the high-T_c superconductors,[1] heavy fermion compounds,[2] and organic molecular crystals,[3–6] the superconducting (SC) phases appear in the vicinity of the antiferromagnetic (AF) state. In view of the current theoretical models, magnetism bordering on an SC phase is quite important for the superconductivity in many of these materials.[7–13] Organic superconductors such as $(TM)_2X$ (TM=TMTTF: tetramethyl-tetrathiafulvalene or TMTSF: tetramethyl-tetraselenafulvalene, X: anions), κ-$(ET)_2X$ [ET: bis(ethylenedithio)tetrathiafulvalene], and β'-$(ET)_2ICl_2$ have superconducting phases bordering on spin density wave (SDW) or AF insulating states in the temperature-pressure (T-P) phase diagrams.[3,4,14] It is a characteristic of these organic superconductors that the ratio of the donor molecules to anions is stoichiometric 2:1, and the carrier number is fixed to one hole per two donor molecules. Since the dimerization of $(TM)_2X$ is small, the electronic states are explained by the physics based on 3/4-filled band.[15,16] On the other hand, κ- and β'-type $(ET)_2X$ have strongly dimerized structures, and the resulting 3/4-filled band in the presence of the dimerization gap corresponds to effective half filling.[4] The ground state of these organic superconductors is basically regulated by U/W except for a highly frustrated triangular lattice system κ-$(ET)_2Cu_2(CN)_3$,[17] where U is the on-site Coulomb repulsion and W is the bandwidth. Accordingly, the ground state depends not only on physical pressure but also on the change of the anion X (chemical pressure).

By contrast, an ambient-pressure organic superconductor (MDT-TSF)$(AuI_2)_{0.436}$, where MDT-TSF is methylenedithio-tetraselenafulvalene [Fig. 1(a)], has uniform donor stacking without dimerization.[18] This superconductor shows an incommensurate anion structure and unique Fermi surface reconstruction by the incommensurate anion potential.[19–21] The incommensurate anion potential effect on the electronic states appears in the heat capacity at the superconducting transition.[22]

The AuI_2 salt of the sulfur analog MDT-TS [Fig. 1(a)] is represented as (MDT-TS)$(AuI_2)_{0.441}$, and has basically the same incommensurate crystal structure as the selenium ana-

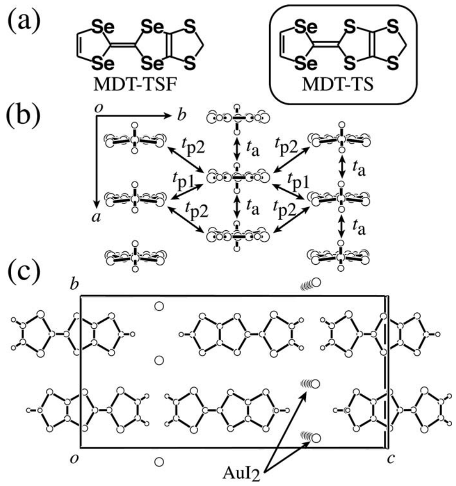

FIG. 1. (a) MDT-TS molecule. (b) Crystal structure projected along the molecular long axis and (c) projection onto the *bc* plane of (MDT-TS)$(AuI_2)_{0.441}$. t_a, t_{p1}, and t_{p2} in (b) are crystallographically independent transfer integrals.

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log. This compound has a uniform donor stacking along the *a* axis and the donors form a conducting sheet on the *ab* plane [Figs. 1(b) and 1(c)]. The anions form chains running parallel to the donor stacks, and the anion periodicity is incommensurate to the donor spacing. Consequently, the non-integer composition, (MDT-TS)$(\mathrm{AuI}_2)_{0.441}$, has been determined from the ratio of the donor lattice (a=3.992(3) Å) and the anion lattice (a'=9.045(6) Å) as a/a'=0.4413(3).[23] As a result, the charge transfer degree from the donor to anion is 0.4413(3) electrons per a donor molecule. The resulting energy band filling is 0.7764(2), and different from the usual 3/4- and 1/2-filling. The Fermi surface is two-dimensional owing to the considerable interchain interaction. This compound, however, shows a metal-insulator (MI) transition at low temperatures, and shows superconductivity under high pressure.[23,24] The magnetic susceptibility of the present compound gradually increases with decreasing temperature in the metallic region. The magnetic susceptibility exhibits anisotropic behavior below 50 K, which suggests an AF ordering, but a spin-flop transition is not observed for magnetic fields up to 5 T.[24] It should be mentioned that α-$(\mathrm{ET})_2\mathrm{KHg(SCN)}_4$, whose ground state is a charge density wave state, shows anisotropic behavior of the magnetic susceptibility.[25–27] Therefore, it is important to observe a spin-flop transition in (MDT-TS)$(\mathrm{AuI}_2)_{0.441}$ to verify the antiferromagnetic ground state.

The present paper reports magnetic torque measurements, which demonstrate the antiferromagnetic state with the high spin-flop field $\mu_0 H_{\mathrm{sf}}$=6.9 T and the high Néel temperature T_N=50 K, where μ_0 is the vacuum magnetic permeability. The phase diagram is extended to higher pressures using the high-pressure transport results, and maximum T_c is determined.

II. EXPERIMENT

Single crystals were prepared by electrocrystallization.[23] For the magnetic torque measurements, the sample (~30 μg/crystal) placed on a microcantilever for atomic force microscopy was mounted on a one-axis rotator in a cryostat in a 15 T superconducting magnet, and was cooled to 1.7 K.[28] Uncertainty in the alignment of the field relative to the crystal axes is a few degrees. The high-pressure resistivity measurements were carried out by a clamped piston-cylinder cell consisting of a NiCrAl alloy inner cylinder and a BeCu outer shell with Daphne 7373 oil as a pressure-transmitting medium, and the sample was cooled to 1.5 K.[29] The room-temperature-pressure value was determined by the resistance measurement of a Manganin wire with the pressure coefficient of 2.4%/GPa.[30] Because the pressure is released by about 0.15 GPa between 300 K and 50 K, this value is subtracted from room-temperature values.[31,32] The resistivity measurements were performed by the four-probe method along the *a* (intralayer) axis with ac current. Magnetic torque measurements were carried out at Tsukuba Magnet Laboratories, NIMS.

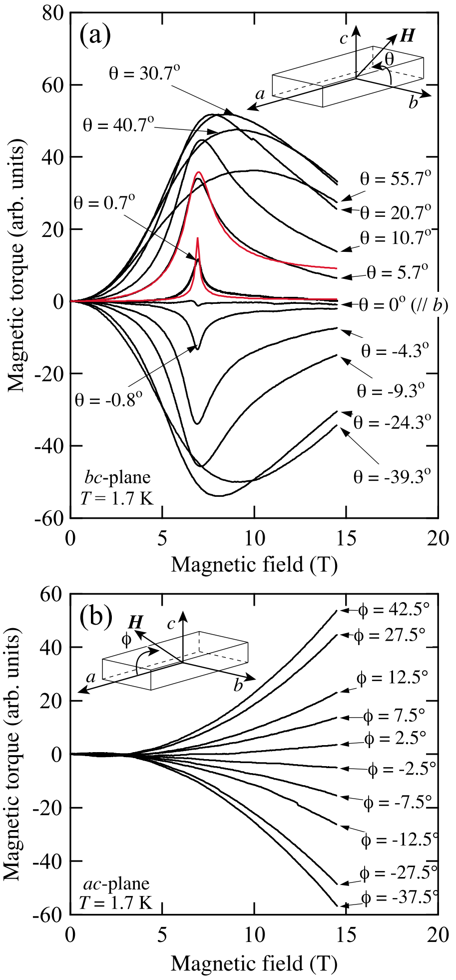

FIG. 2. (Color online) Magnetic torque curves of (MDT-TS)$(\mathrm{AuI}_2)_{0.441}$ at 1.7 K under various magnetic field directions of θ and ϕ, which are the angles tilted (a) from *b* to *c* and (b) from *a* to *c* axes, respectively. The red lines in (a) are the fitted results.

III. RESULTS AND DISCUSSION

Figures 2(a) and 2(b) show the magnetic torque curves of a piece of the single crystal at 1.7 K in various magnetic field directions. The field orientation as shown in the insets is defined by tilt angles θ and ϕ, which are the angles tilted from *b* to *c* and *a* to *c* axes, respectively. The magnetic torque curves have a peak at 6.9 T under $|\theta|$=0.7° in the *bc*-plane, coming from the spin-flop transition from the low-field AF state to the high-field canted AF state. The peak broadens with increasing the angle $|\theta|$. Above $|\theta| \gtrsim 30°$, the magnetic torque curve changes to a gradual spin rotation. The magnetic torque curves do not show peak structure in the *ac*-plane [Fig. 2(b)], indicating that the ground state is a uniaxial AF ordered state with the easy axis parallel to the *b* axis. This is in good agreement with the anisotropic behavior of the spin susceptibility.[24] Although several organic antiferromagnetic compounds show a step-like anomaly above the spin-flop field owing to the transition from the canted AF state to the field induced ferromagnetic state,[33,34] the present compound does not show such a signal up to 14.5 T.

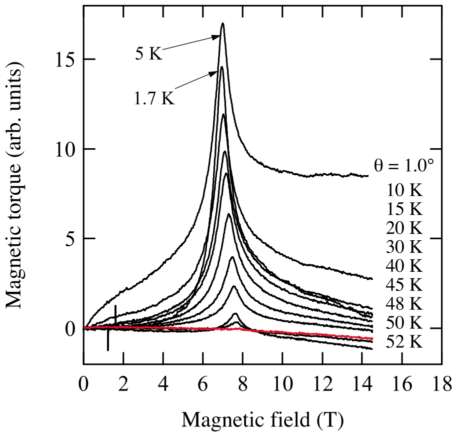

FIG. 3. (Color online) Magnetic torque curves at $\theta = 1.0°$ under various temperatures.

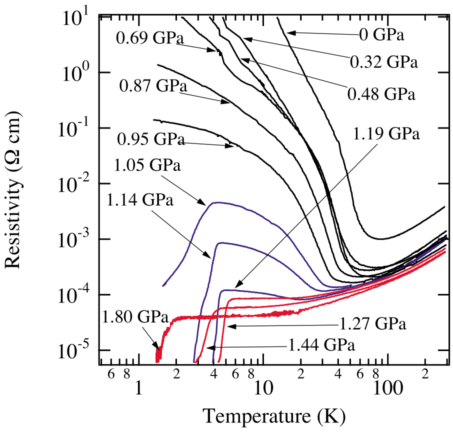

FIG. 4. (Color online) Temperature dependence of the resistivities under various pressures. The results below 1.19 GPa are the previous work.[24]

The magnetic torque τ for a uniaxial antiferromagnet is expressed by the following equations.[35–38]

$$\tau = \frac{1}{2}(\chi_\perp - \chi_\parallel)\mu_0 H^2 \frac{\sin 2\theta}{\sqrt{\lambda^2 - 2\lambda \cos 2\theta + 1}} \tag{1}$$

$$\lambda = \left(\frac{H}{H_{\mathrm{sf}}}\right)^2 \tag{2}$$

$$H_{\mathrm{sf}} = \sqrt{\frac{2K_u}{\mu_0(\chi_\perp - \chi_\parallel)}}, \tag{3}$$

where $\chi_\parallel$ and $\chi_\perp$ are the spin susceptibilities parallel and perpendicular to the easy axis, respectively, H is the external magnetic field, θ is the angle between the applied field and the easy axis, H_{sf} is the spin-flop field, and K_u is the anisotropy energy. By fitting to these equations, we obtain the spin-flop field as $\mu_0 H_{\mathrm{sf}} = 6.9$ T. We can estimate $K_u = 1.4 \times 10^{-1}$ J/mol at 1.7 K, where we define 1 mol as $(\mathrm{MDT-TS})_{2.27}(\mathrm{AuI}_2)$, and use $\chi_\parallel = 0$ and $\chi_\perp = 6.0 \times 10^{-4}$ emu/mol based on our previous work.[24]

Figure 3 shows magnetic torque curves under various temperatures at $|\theta| = 1.0°$. The spin-flop peak intensity decreases with increasing temperature, and the spin-flop field slightly increases with increasing temperature. The peak structure vanishes at 52 K. This is in agreement with the Néel temperature ($T_N = 50$ K) defined from the anisotropic behavior of the spin susceptibility. Although we have measured temperature dependence of the magnetic torque under various magnetic field to estimate the field dependence of T_N, our data have not shown any anomaly around 50 K.

The resistivities under various pressures are shown in Fig. 4. At ambient pressure, the resistivity decreases with decreasing temperature and increases below $T_\rho = 85$ K. The metal-insulator transition temperature T_{MI} is determined from the peak of $d(\ln \rho)/d(1/T)$ as $T_{\mathrm{MI}} = 50$ K at ambient pressure. T_ρ and T_{MI} shift to lower temperatures as the pressure increases, and the resistivities drop to the noise level above $P_c = 1.05$ GPa below onset $T_c = 4.3$ K (midpoint $T_c = 3.2$ K). The resistance peak observed above 1.05 GPa corresponds to the superconducting onset T_c. The resistance peak is suppressed with increasing pressure, and T_c increases with increasing pressure. At 1.27 GPa, the resistance peak vanishes and the normal Fermi liquid-like behavior appears. The maximum midpoint (onset) T_c is 4.9 K (6.2 K) at 1.27 GPa, and T_c decreases with $dT_c/dP \sim -6$ K/GPa above this pressure value. This decreasing rate is significantly smaller than those of other organic superconductors; β''-$(\mathrm{ET})_2\mathrm{SF}_5\mathrm{CH}_2\mathrm{CF}_2\mathrm{SO}_3$ has -13.4 K/GPa, and κ-$(\mathrm{ET})_2\mathrm{Cu(NCS)}_2$ shows -38.4 K/GPa.[39]

Figure 5 shows the T-P-H phase diagram of $(\mathrm{MDT-TS})(\mathrm{AuI}_2)_{0.441}$ based on the transport and the magnetic measurements. The triple point bordering on insulator, metal, and superconductor, exists at about $P = 1.22$ GPa and $T_c = 4.5$ K in the T-P plane. Below this pressure, the superconducting phase exists under the insulating phase.[40] This is the same as those of κ-$(\mathrm{ET})_2\mathrm{Cu[N(CN)_2]Cl}$,[43] β'-$(\mathrm{ET})_2\mathrm{ICl}_2$,[14] and $(\mathrm{TMTSF})_2\mathrm{AsF}_6$.[44] For κ-$(\mathrm{ET})_2\mathrm{Cu[N(CN)_2]Cl}$, the NMR and ac susceptibility measurements under helium gas pressure have shown that the AF phase coexists with the SC phase in such a pressure region.[43] The present MDT-TS salt seems to have a coexistence region of the AF and SC phases, as well. The highest T_c appears

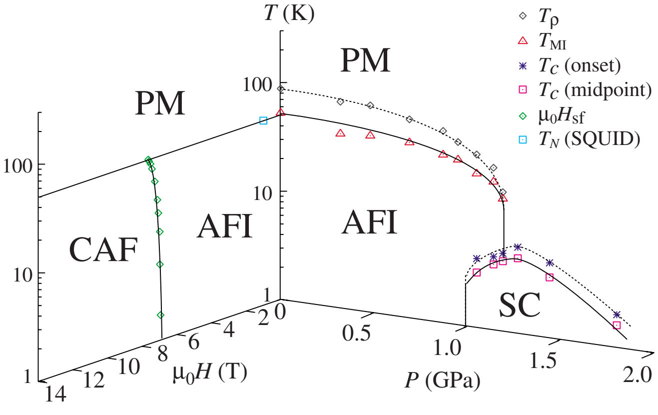

FIG. 5. (Color online) Phase diagram of $(\mathrm{MDT-TS})(\mathrm{AuI}_2)_{0.441}$ (PM: paramagnetic metal, AFI: antiferromagnetic insulator, SC: superconductor, CAF: canted antiferromagnetic state). The solid and dotted lines are guides to the eye.

TABLE I. The Néel temperature T_N, the spin-flop field $\mu_0 H_{\mathrm{sf}}$, the anisotropy energy K_u, the maximum midpoint superconducting transition temperature T_c^{max} under the pressure P, the energy band filling, and the ground state for (MDT-TS)(AuI$_2$)$_{0.441}$, β'-(ET)$_2$ICl$_2$,[14,47] (TMTSF)$_2$AsF$_6$,[44,48] and (TMTTF)$_2$Br.[49,50]

	(MDT-TS)(AuI$_2$)$_{0.441}$	β'-(ET)$_2$ICl$_2$	(TMTSF)$_2$AsF6	(TMTTF)$_2$Br
T_N (K)	50	22	12	13
$\mu_0 H_{\mathrm{sf}}$ (T)	6.9	1.1	0.45	0.42
K_u (J/mol)	1.4×10^{-1}	5.4×10^{-3}	1.5×10^{-4}	–
T_c^{max} (K)	4.9	13.4	1.26[a]	0.8
P (GPa)	1.27	8.2	1.2	2.6
Band filling	0.7764(2)	effective 0.5	0.75	0.75
Ground state	AFI	AF Mott	IC-SDW	C-SDW

[a] T_c is defined as the intersection of the extrapolated normal resistivity curve with the tangent drawn through the inflexion point in Ref. 44.

slightly above the triple point pressure not only in the MDT-TS salt but also in the above ET and TMTSF salts.

Although the present phase diagram resembles that of (TM)$_2X$,[3] T_{MI} and T_c of the present compound are about four times higher than those of (TM)$_2X$. The magnetic insulating state below $T_{\mathrm{MI}}=50$ K is not a simple antiferromagnetic state with alternate spins such as β'-(ET)$_2$ICl$_2$, because each molecule or dimer does not have one electron. In this sense, the insulating state is regarded as a kind of SDW state. In contrast to (TM)$_2X$, however, the electronic state of the present compound is two dimensional,[24] and the Fermi surface does not nest. Yoshioka *et al.*[45] have shown the possibility of MI transition in organic conductors with noninteger carrier number based on the one-dimensional extended Hubbard model, where donor carriers are slightly deviated from quarter filling and under an incommensurate periodic potential from anions. They have pointed out that the incommensurate Mott insulating state can be generated by the anion potential, δ, and the ratio between the on-site Coulomb repulsion and the nearest-neighbor transfer integral, U/t, without the nearest-neighbor Coulomb repulsion, i.e., $V=0$. The present experimental results do not give any information about the microscopic magnetic structure, i.e., the periodicity of the spin arrangement. Therefore, the mechanism of the MI transition of the present compound is unclear. Neutron diffraction and x-ray magnetic diffraction measurements are potential tools to clarify this important point, if the small sample size is overcome. For the heavy fermion compounds, CeRhIn$_5$ shows an incommensurate AF ordering, in which the periodicity of the spin arrangement is determined by a neutron diffraction, and an SC phase appears under high pressure.[46] The incommensurability of the spin arrangement slightly changes with increasing pressure. Although the SC phase exists under the AF phase in CeRhIn$_5$, as well, the AF phase is not an insulator but a metal.

Table I lists the characteristic parameters of (MDT-TS)(AuI$_2$)$_{0.441}$, β'-(ET)$_2$ICl$_2$,[14,47] (TMTSF)$_2$AsF$_6$,[44,48] and (TMTTF)$_2$Br.[49,50] The later three compounds show three kinds of AF ordered states, AF Mott insulating state, incommensurate SDW (IC-SDW) state, and commensurate SDW (C-SDW) state. The TMTSF and TMTTF salts have almost the same AF parameters and superconducting transition temperatures. On the other hand, the β'-type ET salt has much larger parameters than those of (TM)$_2X$. This means some relation between the AF parameters and T_c. Although all AF parameters of the present compound are much larger than those of the other compounds listed in Table I, the highest T_c is much lower than that of the ET salt.

There are two important differences between the MDT-TS and β'-ET superconductors in Table I. The first one is the energy band filling. The β'-ET salt has an effective half filled band because of the strong dimerized structure. On the other hand, the band filling of the MDT-TS salt is 0.7764(2). The electronic correlation of the β'-ET salt should be stronger than that of the MDT-TS salt. In the layered organic superconductors, the κ-type ET superconductors and the MDT-TSF ones, we have found that the strength of the many-body effect is the major factor that determines T_c.[51,52] Therefore, the difference of the band-filling is a possible reason of the difference of T_c. The second point is the incommensurate anion potential. Although Yoshioka *et al.*[45] expect that the incommensurate potential is the origin of the metal-insulator transition, they have not shown the relation between the incommensurate potential and the appearance of the SC phase. Our results might suggest another reason for the difference of T_c, that the incommensurate potential suppresses the SC phase.

IV. CONCLUSION

In summary, the incommensurate organic conductor (MDT-TS)(AuI$_2$)$_{0.441}$ shows an antiferromagnetic ordering with a high Néel temperature ($T_N=50$ K) and a high spin-flop field ($\mu_0 H_{\mathrm{sf}}=6.9$ T) at ambient pressure. Although the highest T_c (midpoint $T_c=4.9$ K) appears at $P=1.27$ GPa, T_c is much lower than that of β'-(ET)$_2$ICl$_2$ in spite of the high Néel temperature, a large spin-flop field, and anisotropy energy.

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[1] G. Burns, *High-Temperature Superconductivity* (Academic, New York, 1992).

[2] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. W. Haselwimmer, and G. G. Lonzarich, Nature (London) **394**, 39 (1998).

[3] D. Jérome, Science **252**, 1509 (1991).

[4] K. Kanoda, Hyperfine Interact. **104**, 235 (1997); J. Phys. Soc. Jpn. **75**, 051007 (2006).

[5] T. Ishiguro, K. Yamaji, and G. Saito, *Organic Superconductors*, 2nd ed. (Springer, Berlin, 1998).

[6] B. J. Powell and R. H. McKenzie, J. Phys.: Condens. Matter **18**, R827 (2006).

[7] S.-C. Zhang, Science **275**, 1089 (1997).

[8] J. Schmalian, Phys. Rev. Lett. **81**, 4232 (1998).

[9] H. Kino and H. Kontani, J. Phys. Soc. Jpn. **67**, 3691 (1998).

[10] T. Moriya and K. Ueda, Adv. Phys. **49**, 555 (2000).

[11] B. J. Powell and R. H. McKenzie, Phys. Rev. Lett. **94**, 047004 (2005).

[12] J. Liu, J. Schmalian, and N. Trivedi, Phys. Rev. Lett. **94**, 127003 (2005).

[13] J. Y. Gan, Y. Chen, Z. B. Su, and F. C. Zhang, Phys. Rev. Lett. **94**, 067005 (2005).

[14] H. Taniguchi, M. Miyashita, K. Uchiyama, K. Satoh, N. Môri, H. Okamoto, K. Miyagawa, K. Kanoda, M. Hedo, and Y. Uwatoko, J. Phys. Soc. Jpn. **72**, 468 (2003).

[15] H. Seo and H. Fukuyama, J. Phys. Soc. Jpn. **66**, 1249 (1997).

[16] H. Seo, J. Merino, H. Yoshioka, and M. Ogata, J. Phys. Soc. Jpn. **75**, 051009 (2006).

[17] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, Phys. Rev. Lett. **91**, 107001 (2003).

[18] K. Takimiya, Y. Kataoka, Y. Aso, T. Otsubo, H. Fukuoka, and S. Yamanaka, Angew. Chem., Int. Ed. **40**, 1122 (2001).

[19] T. Kawamoto, T. Mori, K. Takimiya, Y. Kataoka, Y. Aso, and T. Otsubo, Phys. Rev. B **65**, 140508(R) (2002).

[20] T. Kawamoto, T. Mori, C. Terakura, T. Terashima, S. Uji, K. Takimiya, Y. Aso, and T. Otsubo, Phys. Rev. B **67**, 020508(R) (2003).

[21] T. Kawamoto, T. Mori, C. Terakura, T. Terashima, S. Uji, H. Tajima, K. Takimiya, Y. Aso, and T. Otsubo, Eur. Phys. J. B **36**, 161 (2003).

[22] T. Ishikawa, Y. Nakazawa, S. Yamashita, M. Oguni, K. Saito, K. Takimiya, and T. Otsubo, J. Phys. Soc. Jpn. **75**, 074606 (2006).

[23] K. Takimiya, M. Kodani, N. Niihara, Y. Aso, T. Otsubo, Y. Bando, T. Kawamoto, and T. Mori, Chem. Mater. **16**, 5120 (2004).

[24] T. Kawamoto, Y. Bando, T. Mori, K. Takimiya, and T. Otsubo, Phys. Rev. B **71**, 052501 (2005).

[25] T. Sasaki, H. Sato, and N. Toyota, Synth. Met. **42**, 2211 (1991).

[26] K. Kanoda, A. Kawamoto, K. Miyagawa, and Y. Nakazawa, Synth. Met. **70**, 973 (1995).

[27] P. Foury-Leylekian, S. Ravy, J.-P. Pouget, and H. Müller, Synth. Met. **137**, 1271 (2003).

[28] E. Ohmichi and T. Osada, Rev. Sci. Instrum. **73**, 3022 (2002).

[29] T. Terashima, T. Matsumoto, C. Terakura, S. Uji, N. Kimura, M. Endo, T. Komatsubara, and H. Aoki, Phys. Rev. Lett. **87**, 166401 (2001).

[30] H. Fujiwara, H. Kadomatsu, and K. Tohma, Rev. Sci. Instrum. **51**, 1345 (1980).

[31] K. Murata, H. Yoshino, H. O. Yadav, Y. Honda, and N. Shirakawa, Rev. Sci. Instrum. **68**, 2490 (1997).

[32] K. Yokogawa, K. Murata, H. Yoshino, and S. Aoyama, Jpn. J. Appl. Phys., Part 1 **46**, 3636 (2007).

[33] T. Sasaki, H. Uozaki, S. Endo, and N. Toyota, Synth. Met. **120**, 759 (2001).

[34] K. Kajiyoshi, T. Kambe, M. Tamura, and K. Oshima, J. Phys. Soc. Jpn. **75**, 074702 (2006).

[35] K. Yosida, Prog. Theor. Phys. **6**, 691 (1951).

[36] T. Nagamiya, K. Yosida, and R. Kubo, Adv. Phys. **4**, 1 (1955).

[37] J. Kanamori, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1963), Vol. I, Chap. 4.

[38] H. Uozaki, T. Sasaki, S. Endo, and N. Toyota, J. Phys. Soc. Jpn. **69**, 2759 (2000).

[39] S. Sadewasser, C. Looney, J. S. Schilling, J. A. Schlueter, J. M. Williams, P. G. Nixon, R. W. Winter, and G. L. Gard, Solid State Commun. **104**, 571 (1997).

[40] We use the word *insulator* when the resistance increases with decreasing temperature. However, such an *insulating* phase in the present compound under $T>T_c$ and $1.0<P<1.3$ GPa might be categorized in a *bad metal* introduced by Emery and Kivelson for a high-T_c cuprate (Ref. 41). The transport properties of a *bad metal* in organic conductors have been investigated in Ref. 42.

[41] V. J. Emery and S. A. Kivelson, Phys. Rev. Lett. **74**, 3253 (1995).

[42] J. Merino and R. H. McKenzie, Phys. Rev. B **61**, 7996 (2000).

[43] S. Lefebvre, P. Wzietek, S. Brown, C. Bourbonnais, D. Jérome, C. Mézière, M. Fourmigué, and P. Batail, Phys. Rev. Lett. **85**, 5420 (2000).

[44] R. Brusetti, M. Ribault, D. Jérome, and K. Bechgaard, J. Phys. (Paris) **43**, 801 (1982).

[45] H. Yoshioka, H. Seo, and H. Fukuyama, J. Phys. Soc. Jpn. **74**, 1922 (2005).

[46] A. Llobet, J. S. Gardner, E. G. Moshopoulou, J.-M. Mignot, M. Nicklas, W. Bao, N. O. Moreno, P. G. Pagliuso, I. N. Goncharenko, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B **69**, 024403 (2004).

[47] N. Yoneyama, A. Miyazaki, T. Enoki, and G. Saito, Bull. Chem. Soc. Jpn. **72**, 639 (1999).

[48] K. Mortensen, Y. Tomkiewicz, and K. Bechgaard, Phys. Rev. B **25**, 3319 (1982).

[49] S. S. P. Parkin, J. C. Scott, J. B. Torrance, and E. M. Engler, Phys. Rev. B **26**, 6319 (1982).

[50] L. Balicas, K. Behnia, W. Kang, E. Canadell, P. Auban-Senzier, D. Jérome, M. Ribault, and J. M. Fabre, J. Phys. I **4**, 1539 (1994).

[51] T. Kawamoto, T. Mori, T. Konoike, K. Enomoto, T. Terashima, S. Uji, H. Kitagawa, K. Takimiya, and T. Otsubo, Phys. Rev. B **73**, 094513 (2006).

[52] T. Kawamoto and T. Mori, Phys. Rev. B **74**, 212502 (2006).