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Optical, spin-resonance, and magnetoresistance studies of (tetrathiatetracene)$_2$(iodide)$_3$. The nature of the ground state

R. B. Somoano, S. P. S. Yen, V. Hadek, and S. K. Khanna
Jet Propulsion Laboratory, Pasadena, California 91103

M. Novotny
Physics Department, Stanford University, Stanford, California 94305

T. Datta and A. M. Hermann
Physics Department, Tulane University, New Orleans, Louisiana 70118

John A. Woollam
NASA Lewis Research Center, Cleveland, Ohio 44135

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Optical, spin resonance, and high-field magnetoresistance studies of the organic charge-transfer salt (tetrathiatetracene)$_2$(iodide)$_3$ [$(\mathrm{TTT})_2\mathrm{I}_3$] have been carried out. This material is a single-carrier system consisting of segregated linear chains of TTT cation radicals and tri-iodide ions which are incommensurate with respect to each other. The I_3^- chains exhibit considerable disorder perpendicular to the chain axis. $(\mathrm{TTT})_2\mathrm{I}_3$ has a high room-temperature conductivity $\sigma(300\,^\circ\mathrm{K}) \sim 1000\ \Omega^{-1}\,\mathrm{cm}^{-1}$ along the chain axis and is metallic down to $\sim 100\,^\circ\mathrm{K}$ where a possible phase transition occurs. The nature of this transition is discussed. Below 30 °K, $(\mathrm{TTT})_2\mathrm{I}_3$ exhibits semiconducting properties indicating that the low-temperature ground state is nonmetallic.

I. INTRODUCTION

One of the fundamental problems in the study of quasi-one-dimensional (1D) organic metals is the stabilization of the metallic state against electronically driven instabilities which may lead to nonmetallic behavior. Some of the factors which affect the stability of the metallic state include interchain coupling, disorder, and commensurability energy. Most organic compounds which exhibit metal-like properties to low temperatures ($T \sim 30-100\,^\circ\mathrm{K}$) contain both electrons and holes as charge carriers. Thus, the recent report[1] that (tetrathiatetracene)$_2$(iodine)$_3$ [$(\mathrm{TTT})_2\mathrm{I}_3$] is a stable organic metal down to 3.3 °K is of considerable interest. This material is of additional interest because: (i) unlike other organic metals, TTT is not a derivative of tetrathiafulvalene (TTF), (ii) there is only one type of charge carrier (holes), associated with the TTT cation radical stacks, (iii) the TTT and iodine lattices are incommensurate and the iodine chains exhibit considerable disorder,[2] and (iv) the conductivity peaks in the vicinity of 50–100 °K,[1,3] and then drops to a value of the order of $(0.1-0.01)\sigma_{\mathrm{RT}}$ at 4 °K, a relatively high value for this temperature.

Recent studies of this compound have suggested that $(\mathrm{TTT})_2\mathrm{I}_3$ is: (i) a stable organic metal down to 3.3 °K,[1] (ii) a metal down to $\sim 50-90\,^\circ\mathrm{K}$ where a phase transition occurs leading to nonmetallic behavior with disorder reducing the interchain coupling, thereby, making the system more one dimensional,[3] and (iii) a disordered semiconductor with strong interchain coupling at all temperatures with no phase transitions.[4] In view of the diversity of these suggestions, we have measured the electrical, magnetic, and optical properties of $(\mathrm{TTT})_2\mathrm{I}_3$. We wish to present evidence supporting a metallic state at high temperatures, but also supporting a nonmetallic state at temperatures below 30 °K.

II. EXPERIMENTAL

The crystals studied in this investigation have an orthorhombic crystal structure with lattice constants of $a = 18.424$ Å, $b = 4.957$ Å, $c = 18.323$ Å, and $\alpha = \beta = \gamma = 90°$,[5] in agreement with the structure determined by Smith and Luss.[2] The b axis is the stacking axis. The TTT and iodine sublattices are incommensurate with respect to each other and their lattice spacings yield a stoichiometry of $(\mathrm{TTT})_2(\mathrm{I}_3)_{1.04}$ or $\mathrm{TTT\text{-}I}_{1.56}$. This is similar to that reported by Isett[1] and Mihaly.[4] However, Kiminskii *et al.*[5] report a homogeneity range of TTT-I , where $1.50 \leq x \leq 1.55$. The diffuse lines due to the disordered iodine chains seen for compositions with $x > 1.50$, revert to spots for $x = 1.50$, indicative of ordered iodine chains. All of the crystals examined in this study exhibited diffuse lines due to disorder in the iodine lattice.

We have measured the electrical conductivity of single crystals of $(\mathrm{TTT})_2\mathrm{I}_3$ using the conventional four-probe technique with aquadag contacts. The room-temperature conductivity along the needle axis $\sigma_{\mathrm{dc}}^{b}(\mathrm{RT})$ is $\sim 1000\ \Omega^{-1}\ \mathrm{cm}^{-1}$ giving a mean-free-path λ of the order of two lattice constants. We have also measured the anisotropy in the conductivity at room temperature by the Montgomery technique and find that $\sigma_b/\sigma_a \sim 160$. This value is low compared to the ratio $\sigma_b/\sigma_a \sim 300-500$ observed[6] in (tetrathiafulvalene) (tetracyanoquinodimethane) TTF-TCNQ (the *a* axis in TTF-TCNQ is perpendicular to the donor and acceptor sheets). Thus, there appears to be significant interchain coupling in $(\mathrm{TTT})_2\mathrm{I}_3$ although not as much as is observed in the stable organic semimetal hexamethyltetraselenafulvalene (HMTSeF)-TCNQ, where $\sigma_b/\sigma_a \sim 30 \pm 10$.[6]

The *b*-axis conductivity increases with decreasing temperature in a metal-like fashion similar to that reported by Isett.[1] The resistivity can best be described in the temperature range $100 < T < 300\,^{\circ}\mathrm{K}$ by $\rho(T) = \rho_0 + \rho_1 T^{\gamma}$ with $\gamma = 2.3-2.4$ (compare to the previously reported[1] value of 2.0). At $\sim 100\,^{\circ}\mathrm{K}$, the conductivity peaks with $\sigma_{\mathrm{max}}^{b}/\sigma_{\mathrm{RT}}^{b} \sim 2-3$. We have found that most of our crystals tend to fracture at temperatures slightly less than 90 °K, in spite of very careful crystal mounting and very slow cooling. The conductivity decreases with temperatures below 90 °K to a value at 4 °K of the order of (0.01) σ_{RT}^{b}. The temperature dependence is not purely exponential in this temperature range. At 4 °K, the mean free path deduced from the conductivity is much less than the lattice constant and coupled with the temperature dependence, indicates a nonmetallic state at low temperatures.

The room-temperature polarized reflectivity of a single crystal of $(\mathrm{TTT})_2\mathrm{I}_3$ is shown in Fig. 1. The difference between $R_{\parallel}$ and $R_{\perp}$ is quite pronounced, reflecting the anisotropic nature of these crystals. Other important features of $R_{\parallel}$ are the sharp increase in the reflectivity near 0.6 eV, indicative of a plasma edge, and the prominent structure centered around 1.85 eV which is due to interband transitions. The minimum in the reflectivity near 0.6 eV also suggests the importance of nearby interband transitions. Therefore, we have analyzed the reflectivity data using a Drude-Lorentz model. The dielectric constant is given by

$$\tilde{\epsilon}_{\mathrm{DL}} = \tilde{\epsilon}_f + \tilde{\epsilon}_b \ , \qquad (1)$$

where

$$\tilde{\epsilon}_f = -\omega_p^2/(\omega^2 - i\omega/\tau) \qquad (2)$$

and

$$\tilde{\epsilon}_b = \epsilon_c + \sum_i \frac{\omega_{p_i}^2}{(\omega_i^2 - \omega^2) - i\omega/\tau_i} \ . \qquad (3)$$

$\tilde{\epsilon}_f$ and $\tilde{\epsilon}_b$ are the free and bound electron contributions to the dielectric constant, respectively. $\omega_p^2 = 4\pi N_h e^2/m^*$ is the free-electron plasma frequency, N_h is the density of holes, m^* the optical effective mass, $\tau = \Gamma^{-1}$ the relaxation time, ϵ_c the contribution to the dielectric constant from the core electrons, and ω_i is the frequency of the interband transition. A fit of this model to the data using one Lorentzian oscillator yields the following parameters: $\epsilon_c = 2.61 \pm 0.27$, $\hbar\omega_p = 1.33 \pm 0.15$ eV, $\hbar\Gamma = 0.076 \pm 0.056$ eV, $\hbar\omega_i = 1.87 \pm 0.18$ eV, $\hbar\Gamma_i = 0.81 \pm 0.04$ eV, and $\hbar\omega_{p_i} = 2.94 \pm 0.42$ eV. Note that the minimum in the reflectivity occurs approximately at

$$\hbar[\omega_p^2/\epsilon_1^b(\omega_{\mathrm{min}})]^{1/2} \ ,$$

since the free-electron plasma frequency is screened by the nearby interband transitions. The inset shows the real and imaginary parts of the total dielectric constant, ϵ_1 and ϵ_2, respectively, as computed from Eq. (1) where $\tilde{\epsilon}_{\mathrm{DL}} = \epsilon_1 + i\epsilon_2$. From the plasma energy 1.33 eV and the density of carriers, one can estimate the optical effective mass. Using the x-ray data and assuming complete charge transfer, $N_h \sim 1.2 \times 10^{21}$ holes/cm^3, one finds $m^* \sim 0.93\ m_0$. For a three quarter-filled electron band, using the tight-binding approximation, this gives a crude estimate of the bandwidth of ~ 1.5 eV. A similar value was obtained by Mihaly[4] using the room-temperature thermoelectric power (TEP). Measurements of the magnitude and temperature dependence of the longitudinal TEP of single crystals of our samples[7] yielded results in agreement with Mihaly's work.[4] The assignment of the absorption bands centered at 1.85 eV and polarized along the *b* axis, to particular interband transitions has not been made. It is possible that there is more than one transition comprising the structure centered around 1.97 eV, but the uncertainty in the experiment precludes such identification. A slightly better fit to the data may be obtained by using the Drude-Lorentz model with additional Lorentzian oscillators. However, in view of the uncertainty in the data, we attached no particular physical significance to these additional oscillators. The interpretation of this structure in terms of excitations of $(\mathrm{TTT})^0$, $(\mathrm{TTT})^+$, or I_3^- is complicated since these molecules absorb in similar regions. Solid-state diffuse reflectance studies by Mihaly[8] and by Matsunaga[9] indicate that $(\mathrm{TTT})^0$ absorbs at 1.61, 2.17, and 2.6 eV while $(\mathrm{TTT})^+$ has bands at 0.78, 1.18, 2.28 eV, and a shoulder at 2.60 eV. Diffuse reflectance studies of symmetrical tri-iodide ions in different solids[10] show absorption bands for I_3^- at 2.16, 2.71, and 3.38 eV. Mihaly[8] has also measured the diffuse reflectance of $(\mathrm{TTT})_2\mathrm{I}_3$ for $\hbar\omega \gtrsim 1.6$ eV and finds an absorption band at 2.23 eV and a shoulder at 2.73 eV. Thus, he finds no sign of the $(\mathrm{TTT})^0$ absorption at 1.61 eV and concludes that only $(\mathrm{TTT})^+$ and/or I_3^- are absorbing. Our broad band at 1.87 eV could reflect contributions from $(\mathrm{TTT})^0$ (1.61 and 2.17 eV), $(\mathrm{TTT})^+$ (0.78, 1.18, and 2.28 eV), and I_3^- (2.16 eV). However, the absence of the $(\mathrm{TTT})^0$ and I_3^- bands at 2.6 and 2.7 eV, respectively, cannot be accounted for. Intriguingly, Gabes and Stufkens[10] measured the diffuse reflectance of tetrabutylammonium tri-iodide $[(\mathrm{butyl})\mathrm{NH_4I_3}]$ and observed an absorption band at 1.85 eV which they assign as a charge transfer transition. Thus, the optical absorption of $(\mathrm{TTT})_2I_3$ is not quantitatively understood at present. The optical reflectivity, however, provides additional evidence for metallic behavior at room temperature. Far-ir measurements would be useful for confirming the metallic nature of the reflectivity.

In order to gain further insight into the charge transport process, we have studied the electron spin resonance (ESR) and high-field magnetoresistance of $(\mathrm{TTT})_2I_3$. The ESR measurements were carried out on six oriented single crystals using a conventional Varian (E-12) X-band spectrometer with a rectangular cavity operating in the TE_{102} mode. The temperature was measured using an Fe chromel-Au thermocouple and controlled with an Air Products cryotip and Heli-Trans (Model ADP-B) temperature controller. The crystals were mounted on a quartz fiber using vacuum grease which was then inserted into an ESR tube. At room temperature, for the orientation $(\vec{H}_0 \parallel \vec{c},\ \vec{H}_1 \parallel \vec{b})$, a relatively broad line having a Lorentzian line shape is observed with a linewidth of 155 ± 5 G. This large linewidth is what one would expect in view of the large spin-orbit coupling constant of iodine ($\Lambda \sim 5060\,\mathrm{cm}^{-1}$). The g value was measured by using the hyperfine lines of Mn^{2+} in ZnS as a marker. The g value for this orientation at 4.2 °K is 2.0052 ± 0.0006) and is independent of temperature up to 300 °K. The temperature dependence of the linewidth is shown in Fig. 2. The linewidth decreases linearly with decreasing temperature down to ~105 °K. This temperature dependence, as well as the Lorentzian line shape, is suggestive of spin-lattice relaxation processes in a metal, and thus, corroborates the conclusions deduced from the reflectivity and the temperature dependence of the conductivity. A sharp drop in linewidth occurs near 105 °K, the same region where the conductivity peaks. Note that this behavior is roughly similar to that observed in TTF-X,[11] X is a halide or pseudohalide, but quite different from that found in TTF-TCNQ.[12] The linewidth continues to decrease linearly with decreasing temperature down to ~30 °K. The linear nature of the temperature dependence in this temperature range (30–110 °K) is puzzling in view of the positive temperature coefficient of the conductivity. If a transition has occurred at ~100 °K, it is obviously not of the metal-to-insulator type, but may be a metal-to-metal or metal-to-semimetal transition. Below 30 °K, the linewidth becomes temperature independent suggesting additional changes occurring in the electronic structure.

The g value for the orientation $\vec{H}_0 \parallel \vec{a},\ \vec{H}_1 \parallel \vec{b}$ has also been measured. It is 2.0002 ± 0.0006 and independent of temperature from 4–300 °K. The linewidth is isotropic for H_0 in the a-c plane $(\vec{H}_1 \parallel \vec{b})$ at 4.2 °K and room temperature. The orientation from $\vec{H}_0 \parallel \vec{c}$ to $\vec{H}_0 \parallel \vec{a}$ was done manually while the crystals were at 4.2 °K and, therefore, is approximate. More accurate studies of the orientation dependence of the g value are scheduled.

Karimov *et al.*[13] have made magnetoresistance

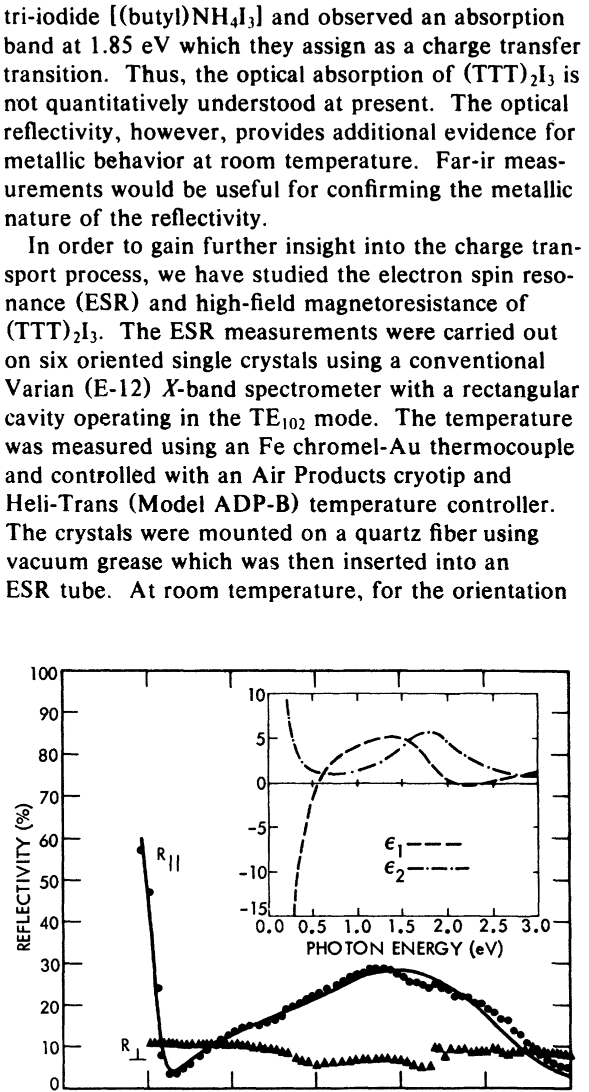

FIG. 1. Optical reflectivity of a single crystal of $(\mathrm{TTT})_2I_3$. $R_{\parallel}$ and $R_{\perp}$ are the reflectivities for light polarized parallel and perpendicular to the conducting needle b axis, respectively. The solid line represents a fit of Eq. (1) to the $R_{\parallel}$ data. The inset shows the real and imaginary parts of the total dielectric constant ϵ_1 and ϵ_2, respectively, as computed from Eq. (1) where $\tilde{\epsilon}_{\mathrm{DL}} = \epsilon_1 + i\epsilon_2$.

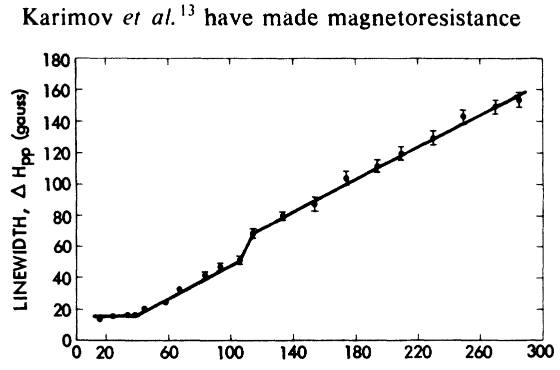

FIG. 2. Temperature dependence of the ESR linewidth of oriented single crystals of $(\mathrm{TTT})_2I_3$, $(\vec{H}_0 \parallel \vec{c},\ \vec{H}_1 \parallel \vec{b})$. The solid-line is to facilitate viewing and is of no physical significance.

measurements on single crystals of $(TTT)_2I_3$ in fields up to 55 kG. We have carried out similar measurements in fields up to 102 kG throughout the temperature range $4 \leq T \leq 160\,^\circ K$. Four-probe magnetoresistance measurements with evaporated gold contacts were performed with the current i along the conducting axis, both parallel and perpendicular to the magnetic field. We find no measurable magnetoresistance down to ~22 °K (sensitivity was ±0.5% of the zero-field resistance). This is consistent with the expected magnitude of the magnetoresistance of a metal where $\Delta R/R$ is of the order of $(\omega_c \tau)^2$. Using the values of m^* and τ obtained from the analysis of the reflectivity data, we find $(\omega_c \tau)^2 \sim 10^{-3}$, in agreement with the lack of observation of a measurable magnetoresistance in the metallic state. At $T < 22\,^\circ K$, a large and positive magnetoresistance is observed which varies with temperature and magnetic field as $\sim (H/T)^2$ (see Fig. 3), in agreement with the results of Karimov *et al.*[13] The magnitude of the magnetoresistance is independent of crystal (and current) orientation with respect to the magnetic field, and does not obey Kohler's rule. The increase in $\Delta R/R$ with decreasing temperature is not simply a reflection of the zero-field resistivity as in the metallic system $HgAsF_6$,[14] since the resistivity increases with decreasing temperature for $(TTT)_2I_3$ below 22 °K.

The magnetoresistive behavior may be contrasted with that observed in TTF-TCNQ.[15] In TTF-TCNQ, the magnetoresistance is also independent of orientation, but is small and negative. Also, a sharp peak in $\Delta R/R$ is observed at the onset of magnetoresistance, a feature which is believed[15] to reflect the mean-field temperature dependence of the Peierls gap. No such peak was observed in $(TTT)_2I_3$.

There are some similarities between the resistive behavior of $(TTT)_2I_3$ and that observed in semimetallic HMTSeF-TCNQ.[16] The magnetoresistance of HMTSeF-TCNQ is positive and of the order of 3% at zero-pressure and 28 kG, the maximum field for which measurements were reported, and it does not follow Kohler's rule. There are, however, some significant differences in the resistive behavior of $(TTT)_2I_3$ and that of HMTSeF-TCNQ. The zero-field conductivity ratio $\sigma_{4.2}/\sigma_{RT}$, in $(TTT)_2I_3$, is at least a factor or two smaller. Moreover, the tendency toward leveling off of the low-temperature conductivity (at zero pressure) in HMTSeF-TCNQ is not evident in $(TTT)_2I_3$. Furthermore, the magnetoresistance of HMTSeF-TCNQ does not follow the expected H^2 law as it does in $(TTT)_2I_3$. This latter feature in HMTSeF-TCNQ and the lack of adherence to Kohler's law have both been attributed[16] to the presence of holes and electrons along the two-chain system. There is, however, only one set of conducting chains in $(TTT)_2I_3$.

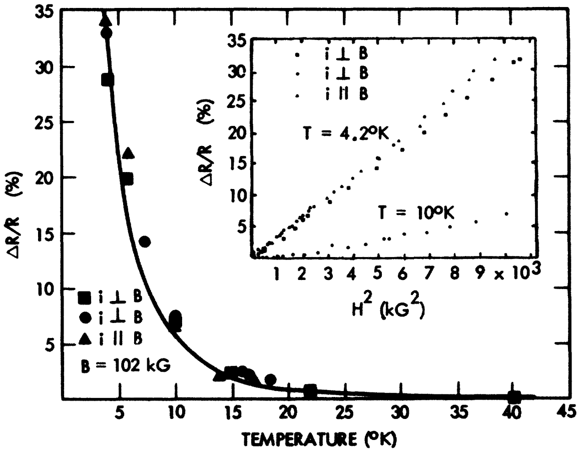

FIG. 3. Magnetoresistance of a single crystal of $(TTT)_2I_3$ as a function of temperature. The solid line is a fit to the equation $\Delta R/R = 600/T^2$. Inset shows the magnetoresistance as a function of applied field.

Additionally we call attention to the large magnitude of the zero-pressure magnetoresistance in $(TTT)_2I_3$, ~35% at 4.2 °K (at 102 kG). In summary, the lack of adherence to Kohler's law in the single-chain system of $(TTT)_2I_3$ argues against the existence of a simple metallic state below 30 °K. Furthermore, while there are some similarities in the magnetoresistance behavior of $(TTT)_2I_3$ and that of semimetallic HMTSeF-TCNQ, there are significant differences which we believe are reflections of different modes of charge transport.

III. CONCLUSIONS

We have measured the electrical, magnetic, and optical properties of $(TTT)_2I_3$. The crystals examined in this study exhibited disorder in the iodine chains and were characterized by a stoichiometry of $(TTT)_2(I_3)_{1.04}$. We believe that $(TTT)_2I_3$ is metallic at high temperatures ($100 < T < 300\,^\circ K$). Evidence supporting this conclusion includes the high value of the room-temperature conductivity, the optical reflectivity, the temperature dependences of the conductivity and ESR line width, and the absence of a measurable magnetoresistance. In addition, the TEP is small, positive, and weakly dependent upon temperature.[4,7] The behavior is similar to what one would expect for hole conduction along the TTT chain in a three quarter-filled electron band.

At 100 °K, the temperature dependences of the conductivity, ESR line width, and TEP all show abrupt changes. We believe that this indicates a possible phase transition. However, the nature of the transition is unclear. The ESR linewidth continues to decrease linearly with decreasing temperature below 100 °K and there is no observable magnetoresistance. The conducitivity decreases with decreasing temperature, but not exponentially. Consequently, we believe that if a phase transition has occurred, it may be of the metal-to-metal or metal-to-semimetal type.

At 20–30 °K, a metal-to-insulator phase transition occurs. This is indicated by the onset of a large and positive magnetoresistance, the leveling off in the temperature dependence of the ESR linewidth, and by recent derivative analysis of the electrical conductivity in this region.[1,17] Even though the magnitude of the conductivity is still relatively high, $\sim 0.01\sigma_{\mathrm{RT}}$ for these temperatures, it suggests a mean free path of the order of 10^{-2} lattice constants, and thus, is inconsistent with metallic behavior. Thus, we believe that the low-temperature ground state ($T < 30\,^\circ\mathrm{K}$) of $(\mathrm{TTT})_2\mathrm{I}_3$ is nonmetallic.

Finally, a comment should be made concerning the role of disorder in this material. As mentioned earlier, it has been suggested that the disorder observed in x-ray studies of $(\mathrm{TTT})_2\mathrm{I}_3$ may be responsible for stabilizing the metallic state down to 3.3 °K or, when combined with interchain coupling, may provide delocalized states in the band gap of a semiconductor[4] such that metal-like charge transport occurs. Certainly the pronounced disorder observed in the x-ray studies should affect the transport properties. The temperature dependence of the conductivity at low temperatures can provide a useful signature for the influence of disorder. However, there is no evidence for a $\ln\sigma \sim -(T_0/T)^{1/2}$ behavior, indicative of hopping in disordered one-dimensional systems,[18] in $(\mathrm{TTT})_2\mathrm{I}_3$. The reported temperature dependences at low temperatures,[1,13] vary as $\ln\sigma \sim -(T_0/T)^{1/3}$ and $\ln\sigma \sim \alpha T$. We have not been able to fit our data to a law of the form $\ln\sigma \sim -(A/T)^n$ over any physically significant temperature range. Similarly, Kaminskii *et al.*[3] report large variations in $\sigma(T)$ at low temperatures. Thus, more extensive low-temperature conductivity measurements would be useful. Preliminary x-ray studies[5] of the temperature dependence of the structure reveal that the iodine chains are becoming more ordered as the temperature is lowered (i.e., the diffuse layer lines gradually collapse into diffuse spots with decreasing temperature). Similar results have been noted in diffuse x-ray scattering of $(\mathrm{TTT})_2\mathrm{I}_3$.[19] Thus, the degree of disorder is decreasing at low temperatures. The important parameters are the ratios of the random potential to the electronic bandwidth ($4t_{\parallel}$) and the interchain coupling, and the competition between electron localization and suppression of a Peierls-type instability by disorder in $(\mathrm{TTT})_2\mathrm{I}_3$ may be quite subtle. The elucidation of the nature of the transition near 100° K (if it exists) would shed light on this question. X-ray studies as a function of temperature down to 25 °K are currently in progress[5] in order to resolve some of these questions. Thus, the contribution of disorder in stabilizing the metallic state or in providing a mode for electron delocalization is unclear. In summary, $(\mathrm{TTT})_2\mathrm{I}_3$ is an interesting one-dimensional electronic system in which it may be possible to study the combined effects of disorder and interchain coupling on charge transport processes.

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