

**Electrical transport in the organic superconductor β -(BEDT-TTF) $_2$ AuI $_2$
[with BEDT-TTF \equiv bis(ethylenedithio)tetrathiafulvalene]: Influence of x-ray-induced defects
on the normal phase and superconducting ground state**

S. Dolanski Babić, N. Biškup, and S. Tomić

Institute of Physics of the University, P.O. Box 304, 41000 Zagreb, Croatia

D. Schweitzer

3. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 7000 Stuttgart 80, Germany

(Received 10 February 1992)

We have measured the in-plane and out-of-plane resistivities of the organic superconductor β -(BEDT-TTF) $_2$ AuI $_2$ with disorder introduced by x-ray irradiation. [Here BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene]. For defect concentrations less than 0.15% per mole, Matthiessen's rule is obeyed at all temperatures and a T^2 term in the resistivity is found in the temperature region between 5 and 20 K. The long-range superconducting order is destroyed by 0.1% per mole and the transition is completely smeared out above 0.2% per mole of defects. The overall temperature behavior of the resistivity is discussed and the possible importance of the electron-electron interaction and two-dimensional localization for the stabilization of the superconducting ground state is emphasized.

I. INTRODUCTION

The (BEDT-TTF) $_2X$ family of organic compounds [where BEDT-TTF is an abbreviation of bis(ethylenedithio)tetrathiafulvalene] differ from the Bechgaard salts (TMTSF) $_2X$ (where TMTSF is an abbreviation of tetramethyltetraselenabulvalene) principally in the following features: First, by their higher critical temperatures (T_{SC}) of the superconducting (SC) ground state (maximum T_{SC} is 13 K) and second, by the fact that a metallic phase exists at ambient pressure in the whole temperature (T) range from room temperature (RT) down to the SC phase transition (or the lowest temperature attainable experimentally if there is no superconductivity).^{1,2} The clearest examples showing this behavior are members of the isostructural class of β -phase compounds with symmetrical trihalide and dihalide anions, with $X = -I_3$ (β_{high} phase) AuI $_2$, and IBr $_2$, which are ordered at RT. Moreover, the terminal ethylene groups of the BEDT-TTF molecules which are in close contact with trihalide and dihalide anions are also uniformly ordered at RT. Therefore, these systems are free from inherent disorder found, for example, in $X = -I_3$ (β_{low} phase) and BrI $_2$ (β phase). In the former, the terminal ethylene groups are randomly positioned in two distinct orientations with respect to molecular planes at high temperatures and subsequently order to give an incommensurate modulation at low temperatures. In the latter, disorder is due to the asymmetric, dipolar anions. The β structure has the most uniform two-dimensional (2D) BEDT-TTF network and has important overlaps along the molecular stacking direction in addition to the significant interstack sulfur-sulfur contacts characteristic of other BEDT-TTF salts' phases. The organic cation conducting network is confined to the (a, b) planes and alternate with noncon-

ducting anion sheets along the c^* direction. Since the charge transfer is defined by the 2:1 stoichiometry, the conduction band is nominally three-quarters filled. Because of the nearly isotropic molecular overlap in the (a, b) plane, one would expect the Fermi surface to be closed in that plane and open along the c^* direction: From magnetoresistance measurements, the ratios of transfer integrals along the a , b , and c^* directions, $t_a:t_b:t_{c^*}$, are about 56:19:1.³ Optical measurements give even smaller anisotropy in the (a, b) plane ($t_a/t_b \approx 1.5$).⁴ A closed Fermi surface has been obtained in band-structure calculations based on the extended Hückel molecular-orbital method by Mori *et al.*⁵ and experimentally verified by the observation of Shubnikov-de Haas magnetoresistance oscillations.⁶

In spite of extensive studies in the last few years, our present understanding of some basic properties, such as electrical transport in the metallic phase and the origin and nature of the superconducting ground state, are still incomplete for both the (TMTSF) $_2X$ and the (BEDT-TTF) $_2X$ families. For the former, the appearance of microcracks and concomitant jumps in the resistivity along the chain direction presents impediments to any serious, systematic approach to the study of high-temperature electrical transport. Only recently has a development of sample mounting with strain-free contacts opened the research in this area.⁷ For the latter, Bulaevskii has analyzed in-plane conductivity and argued that at all temperatures it is dominated by electron-electron Umklapp scattering over a closed Fermi surface.⁸ This rather interesting result certainly deserves more attention. Furthermore, the RT value of the conductivity ($\sigma_{RT} \approx 30 \Omega^{-1} \text{cm}^{-1}$) is one order of magnitude smaller than in the Bechgaard salts ($\sigma_{RT} \approx 500 \Omega^{-1} \text{cm}^{-1}$), which gives an extremely short mean free path of about 0.1 intermolecu-

lar distance.⁹ It is perhaps surprising and worth noting that this feature does not prevent metallic behavior of the temperature-dependent resistivity.

Existing data for the quasi-one-dimensional Bechgaard conductors have been used to show that long-range uniformity of the potential along the conducting chains is necessary to stabilize superconductivity.¹⁰ However, at ambient pressure the fact that the introduction of disorder by alloying not only suppresses superconductivity but also stabilizes the antiferromagnetic phase, has complicated the analysis for compounds with noncentrosymmetric anions. Nevertheless, investigation of the influence of the irradiation-induced disorder, especially in the system without inherent disorder due to noncentrosymmetric anions, such as $(\text{TMTSF})_2\text{PF}_6$, has shown that SC under a pressure of 11 kbar is suppressed below at least 20 mK by only 0.01% per mole of defects.¹¹

As far as BEDT-TTF compounds are concerned, Tokumoto *et al.* have studied the influence of anion alloying and irradiation on the SC in the β -(BEDT-TTF)₂I₃ system.^{12,13} In the alloy study with $X = (\text{I}_3)_{1-x}(\text{IBr}_2)_x$, $(\text{IBr}_2)_{1-x}(\text{I}_2\text{Br})_x$, and $(\text{IBr}_2)_{1-x}(\text{I}_3)_x$, the authors found a strong correlation between the superconductivity and the residual resistivity (ρ_{res}) in that SC disappears when ρ_{res} exceeds the value of 150 $\mu\Omega$ cm. When disorder is introduced by irradiation, SC is suppressed for the concentrations larger than 0.08% per mole. However, in the β_{high} -(BEDT-TTF)₂I₃, SC remained unaffected even at the level of 0.48% per mole. This rather surprising result has not been verified for any other analogous system.

The aim of the present work is to obtain a better understanding of interactions governing the electrical transport in the normal phase and the SC ground state. We have chosen a system free from inherent disorder [and therefore analogous to the $X = -\text{I}_3$ (β_{high} phase) system] $X = \text{AuI}_2$ (β phase) to avoid complications which might arise due to disordered CH₂ groups or anions. This material is a conductor in the entire T range and has a SC ground state with T_{SC} varying from 2 to 5 K according to different authors.^{14,15} We have studied the temperature-dependent electrical resistivity in the highly-conducting (a, b) plane and along the least-conducting c^* direction down to 1.3 K. In addition, we have investigated the role of disorder induced by x-ray irradiation on the normal phase conductivity and SC ground state.

II. EXPERIMENT

Measurements have been performed on single crystals of β -(BEDT-TTF)₂AuI₂ grown electrochemically.¹⁶ Seven samples from two different preparation batches have been studied. The obtained results were qualitatively the same for all samples studied. The typical dimensions a , b , and c^* of the samples used for ρ_{ab} and ρ_{c^*} measurements were $1.2 \times 0.7 \times 0.25$ and $0.5 \times 0.3 \times 0.3$ mm³, respectively. Gold pads were evaporated onto the samples and 17 μm annealed gold wires were attached to the pads with silver paint in the four-probe configuration. In particular, samples were prepared as described previ-

ously for the c^* -direction resistivity.¹⁷ Furthermore, samples were cooled slowly from RT at 10 K per h in order to avoid resistance jumps. Defects in samples were created at RT by unfiltered radiation from a Cu x-ray tube (35 kV, 24 mA). The concentration of irradiation-induced defects was determined using a tetramethylethylselenafulvalene-dimethyltetracyanoquinodimethane (TMTSF-DMTCNQ) crystal as a reference sample. The correspondence between damage rates and resulting resistance changes of TMTSF-DMTCNQ was established in previous studies.^{18,19} Here we point out that the irradiation dose necessary to produce 1% per mole of defects in BEDT-TTF compounds amounts to 4000 Mrad, i.e., 3.7 times larger than for the TMTSF-DMTCNQ material.²⁰

The RT values of the in-plane (ρ_{ab}) and out-of-plane (ρ_{c^*}) resistivity were 0.032 ± 0.005 Ω cm and 11 ± 1.5 Ω cm, respectively. The temperature dependence of ρ_{ab} and ρ_{c^*} normalized to RT is shown in Fig. 1. For the latter there is a clear change in the slope at about 160 K, followed by a more rapid decrease at lower temperatures. Note also that ρ_{c^*} starts to drop toward zero below 3.5 K, whereas ρ_{ab} at this temperature displays only a small anomaly and only falls definitely below about 2.5 K. Figure 2 shows temperature-dependent in-plane resistivity for a sample irradiated up to the highest level of 0.2% per mole of defects. Deviations from the behavior of the pure sample start below about 100 K. The slope becomes less pronounced and resistivity starts to saturate at higher temperatures. It is useful to define resistivity ratio as $\kappa = \rho_{4.2\text{K}} / \rho_{\text{RT}}$, where $\rho_{4.2\text{K}}$ and ρ_{RT} are the resistivities measured at 4.2 K and RT, respectively. Plotting the resistivity ratio (in-plane κ_{ab} and out-of-plane κ_{c^*}) in the inset in Fig. 2, we can see that it varies linearly with the defect concentration until about 0.15% per mole. Note that, in the same range of defect concentrations, the absolute value and temperature behavior of the out-of-plane resistivity change very little, and the resistivity ratio (κ_{c^*}) increased only by 20%. From Fig. 1 we can deduce the overall temperature dependence of the anisotropy defined as ρ_{c^*} / ρ_{ab} (Fig. 3). On cooling, the anisotropy smoothly

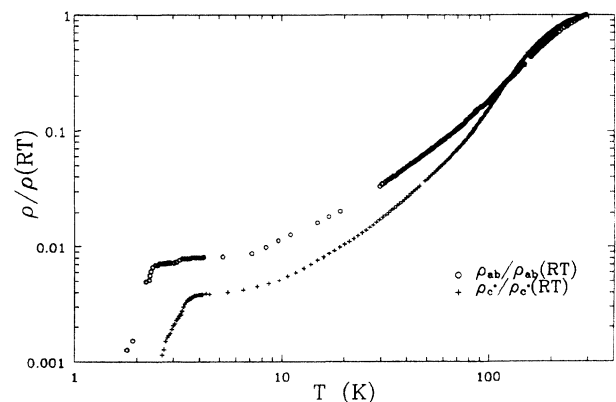


FIG. 1. Logarithm of normalized resistivity [$\rho/\rho(\text{RT})$] vs logarithm of temperature (T) for nominally pure sample. Points and crosses for in-plane and out-of-plane resistivity, respectively.

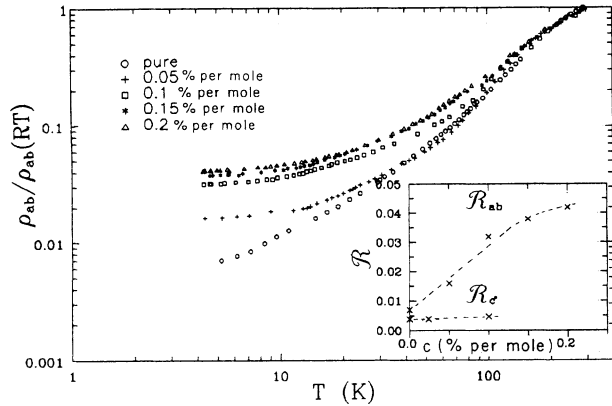


FIG. 2. Logarithm of normalized in-plane resistivity $[\rho_{ab}/\rho_{ab}(RT)]$ vs logarithm of temperature (T) for several defect concentrations. Points, crosses, squares, stars, and triangles for pure sample and 0.05, 0.1, 0.15, and 0.2% per mole, respectively. The inset shows the defect concentration (c) dependence of the in-plane (R_{ab}) and out-of-plane (R_{c^*}) resistivity ratio which is defined as $\rho_{4.2 K RT}$.

increases, reaching a maximum at about 160 K. Then it starts to decrease rather rapidly and levels off below 50 K. At about 3.5 K there is a sharp change in slope indicating that the SC three-dimensional (3D) order sets in. For an irradiated sample with 0.1% per mole of defects the temperature dependence of the anisotropy continues to diminish smoothly below 50 and levels off below 4 K at value of about $\rho_{c^*}/\rho_{ab} \approx 30$ (see inset in Fig. 3). Note that as the temperature decreases below 50 K the magnitude of anisotropy becomes much smaller than in the pure sample. At 4 K the anisotropy ratio between pure and 0.1% per mole irradiated sample is equal to 3. Such behavior indicates that in the presence of irradiation-induced defects the effective dimensionality develops gradually as the system is cooled down below 50 K. Note

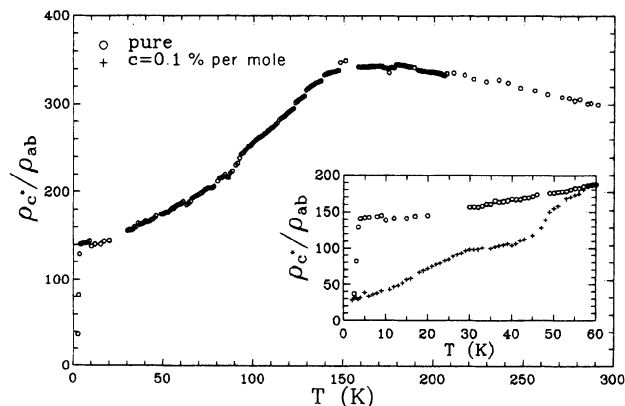


FIG. 3. Overall temperature (T) dependence of the resistivity anisotropy (ρ_{c^*}/ρ_{ab}) for nominally pure systems. The inset shows low-temperature behavior of the resistivity anisotropy (ρ_{c^*}/ρ_{ab}) for pure (points) and irradiated sample (crosses).

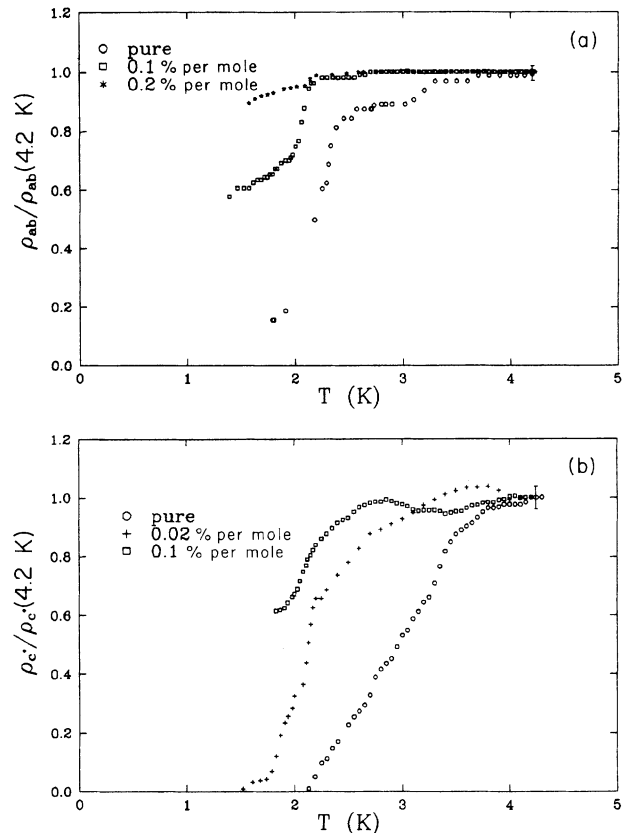


FIG. 4. (a) Normalized in-plane and (b) out-of-plane resistivity $[\rho/\rho(4.2 K)]$ vs temperature (T) for several defect concentrations.

that the nominally pure system attains the same effective dimensionality as the 0.1% per mole irradiated system at about 3 K, i.e., at the SC transition.

The superconductivity, which occurs in pure samples as demonstrated by the rapid drop of ρ_{ab} and ρ_{c^*} , is almost completely suppressed by irradiation to a defect concentration of 0.2% per mole (Fig. 4). Low defect concentrations shift SC transition to lower temperatures and for 0.2% per mole only a barely visible anomaly remains at about 2 K. Note also that already for 0.1% per mole of defects the long-range SC order is destroyed. Namely, both ρ_{ab} and ρ_{c^*} stay finite at the lowest temperature reached. Their values amount to 60% of the normal resistivity measured just above the SC transition. In addition, the observed difference in superconducting transition temperatures for the ρ_{ab} and ρ_{c^*} curves might imply that samples were inhomogeneous.

III. DISCUSSION

In the following discussion we address the origin of interactions defining the electrical transport in the normal phase (Sec. III A) and superconducting phase (Sec. III B). In particular, in III A 1, we discuss the influence of the irradiation-induced defects on the in-plane and out-of-plane electrical conductivity, and in III A 2 we address

the question of the possible importance of electron-electron scattering in the normal phase transport.

A. Electrical transport in the normal phase

In plane and out-of-plane conductivity

In conventional three-dimensional (3D) metals Matthiessen's rule is obeyed, which implies that different scattering mechanisms are additive. In particular, to a first approximation the total resistivity equals the sum of the residual resistivity ρ_0 and the intrinsic temperature-dependent part $\rho(T)$. If Matthiessen's rule is also valid for a 2D metal such as β -(BEDT-TTF)₂AuI₂, then the addition of x-ray-induced defects should increase the resistivity by an amount that does not depend on the temperature. We can write $\rho(c, T) = \rho(0, T) + Ac$, where $\rho(0, T) = \rho_0 + BT^n$ is the total resistivity of the pure system. Our experiments verify the validity of Matthiessen's rule for in-plane resistivity in the range of low defect concentrations ($c \leq 0.15\%$ per mole) to the limited extent that the normalized resistivity increases linearly with c at a fixed temperature (see Fig. 5). Note that the influence of these defect concentrations on the in-plane resistivity is within our experimental uncertainty at high temperatures. In contrast, for the out-of-plane resistivity there was no detectable influence of defects over the large temperature region (4–300 K). Therefore, we can write $\rho_{ab}(c, T) = \rho_{ab}(0, T) + cA$ and $\rho_{c^*}(c, T) \approx \rho_{c^*}(0, T)$ at low temperatures. In this region the in-plane conductivity is coherent, whereas the out-of-plane transport is still diffusive. First, we comment on the influence of defects on the in-plane conductivity. We expect that induced defects are able to reduce the in-plane conductivity in the limit when the mean distance between them L is of the order and larger than the intrinsic in-plane mean free path $l \approx \tau_{ab}^0 v_F$. In the pure sample the mean free path is about $15a$ at low temperatures. The distance L is directly related to the distribution of scattering objects in the system and for the two-dimensional case we can write $L = (1/N)^{1/2}$ where $N = c/a^2$ is a density of defects. For

$c \approx 0.05\%$ per mole we get $L \approx 45a$, which is comparable to the intrinsic in-plane mean free path l . As far as the out-of-plane conductivity is concerned we must keep in mind its diffusive character so that we cannot discuss the c^* direction in terms of the mean free path or relaxation times. However, according to Weger's approach the transverse conductivity is proportional to the interchain hopping frequency, which is proportional to the in-plane collision time (τ_{ab}) .²² In the presence of defects we can write $1/\tau_{ab} = 1/\tau_{ab}^0 + 1/\tau_{ab}(c)$. In other words, in this picture both in-plane and out-of-plane transport depend on the same relaxation time τ_{ab} and therefore we would expect the similar effect for both ρ_{ab} and ρ_{c^*} , contrary to our observations.

The observed deviation from linear behavior with c above about 0.2% per mole is in accordance with data reported by M. Sanquer,¹⁸ and Sanquer and Bouffard²³ for the quasi-1D system R -(TMTSF)₂ClO₄. Also, Zuppiroli *et al.* have already pointed out that one can only explore effects of the defects on the intrinsic temperature dependence of the conductivity in organic conductors at the level of less than a few 0.1% per mole.²⁴ On the other hand, Forro *et al.* found for β -(BEDT-TTF)₂I₃ samples that the linearity is obeyed up to about 3% per mole of defects.²⁰ However, they used samples which contained both β_{low} and β_{high} phases and so were not well defined as far as intrinsic disorder is concerned.

2. Electron-electron scattering

Further, we assume that, following Bulaevskii's proposal, electron-electron scattering plays a major role in limiting the current and in leading, therefore, to a resistivity that varies as BT^2 in the whole temperature region.⁸ That implies that Umklapp U scattering processes are important because normal N electron-electron scattering does not contribute to the resistivity, because of momentum conservation. If the probability ratio of U and N processes is denoted by \mathcal{G}^2 , then the coefficient B is proportional to $\mathcal{G}^2(k_{\text{TF}}^2 v_F E_F)^{-1} E_F^{-2}$.²¹ k_{TF} is the Thomas-Fermi screening wave vector, and v_F and E_F are Fermi velocity and energy, respectively. In normal 3D metals, the effectiveness of U processes is very small ($\mathcal{G} \approx 0.01 - 0.001$), screening is important, and Fermi energies are fairly large (1–10 eV). Therefore, electron-electron scattering cannot prevail over the standard electron-phonon mechanism. However, for the organic conductor such as β -(BEDT-TTF)₂AuI₂, Fermi velocities and Fermi energy are rather small ($E_F \approx 0.1$ eV) and the screening is fairly weak due to the low density of conduction electrons (three orders of magnitude smaller than in the normal metals). In addition, the reduced dimensionality of the system could significantly increase the number of U processes with respect to N ones for "good" band fillings ($4k_F \approx g$, where k_F and g are the Fermi wave vector and wave vector of the reciprocal lattice, respectively).

We have fitted our data from 4.2 to 20 K to the low $\rho(c, T) = \rho(0) + BT^2$, where $\rho(0) = \rho_0 + Ac$, to verify the dominance of electron-electron scattering mechanism for the in-plane and out-of-plane resistivity (Table I and Fig.

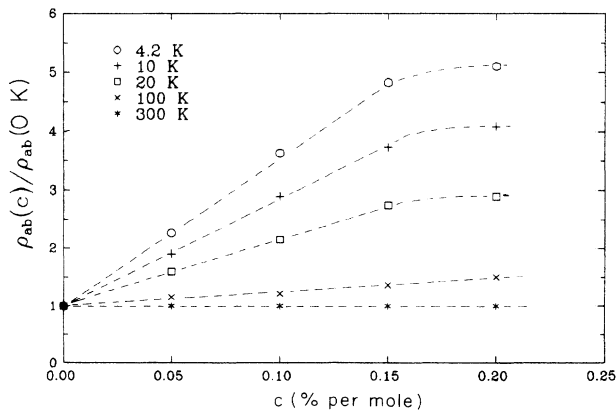


FIG. 5. Normalized in-plane resistivity $[\rho_{ab}(c)/\rho_{ab}(0)]$ vs defect concentration (c) at different temperatures. $\rho_{ab}(c)$ and $\rho_{ab}(0)$ are the resistivity of irradiated and pure sample, respectively.

TABLE I. Parameters for $\rho(c, T) = \rho(0) + BT^2$ plot of the in-plane (ρ_{ab}) and out-of-plane (ρ_{c^*}) resistivity for pure and irradiated samples. $\rho(0)$ and c are the residual resistivity and defect concentration, respectively.

c (% per mole)	ρ_{ab}		ρ_{c^*}	
	$\rho(0)$ ($10^{-4} \Omega \text{ cm}$)	B ($10^{-6} \Omega \text{ cm K}^{-2}$)	$\rho(0)$ ($10^{-2} \Omega \text{ cm}$)	B ($10^{-4} \Omega \text{ cm K}^{-2}$)
0	2 ± 0.3	1.3 ± 0.2	3 ± 0.3	2 ± 0.3
0.05	5	1.3		
0.1	10	1.4	3	2
0.15	12	1.6		
0.2	13	1.6		

6). For the latter, both $\rho(0)$ and B are nearly constant until 0.1% per mole of defects. For the former, $\rho(0)$ increases linearly with defect concentration until about 0.15% per mole, whereas the coefficient B is nearly constant. Above this concentration, Matthiessen's rule fails to be valid (Figs. 5 and 6 and Table I). Namely, the linear rise of $\rho(0)$ saturates and the coefficient B starts to increase. In other words, for higher defect concentrations the additional scattering introduced by irradiation does not enter only in $\rho(0)$, but it also alters the T -dependent term. Note also that the coefficient B of T^2 term for the out-of-plane resistivity is two orders of magnitude larger than for the in-plane. This is associated with lower Fermi velocity in that direction. Further, adequate fits were also obtained from 20 to 80 K, the coefficients $\rho(0)$ and B showing the same behavior with defect concentration as the low-temperature coefficients. However, the values of B are slightly smaller, while those of $\rho(0)$ are all somewhat higher. Note that the similar dependence has been obtained for pure and irradiated quasi-1D system $R\text{-(TMTSF)}_2\text{ClO}_4$.¹⁹

Bulaevskii has argued that in $\beta_{\text{low}}\text{-(BEDT-TTF)}_2\text{I}_3$ the conductivity σ is dominated by electron-electron Umklapp scattering at all temperatures.⁸ Due to (1) the strong pressure dependence of the conductivity and (2) the large compressibility and thermal expansion, the precise magnitude and temperature dependence of the conductivity measured at ambient pressure is quite different

from that at constant volume, as pointed out for organic chain conductors in Ref. 25. As the constant volume conductivity is implicitly considered in the standard transport theory, one should correct experimental ρ versus T curves taken at constant pressure to one at constant volume. However, there is considerable uncertainty in these corrections because the system is anisotropic. Several experimental studies on $\beta\text{-(BEDT-TTF)}_2\text{X}$ and other organic compounds give $d \ln \sigma / dp \approx 30\% \text{ kbar}^{-1}$ for all conducting directions.^{26,27} Further, according to Kobayashi *et al.*, the thermal change of lattice parameters becomes important above 80 K and amounts to

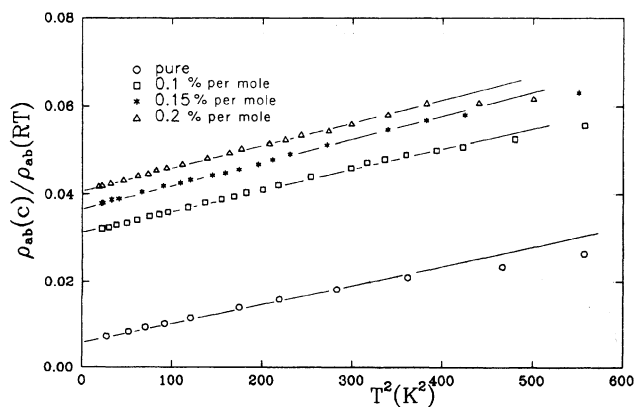


FIG. 6. Normalized in-plane resistivity [$\rho_{ab}(c)/\rho_{ab}(\text{RT})$] vs temperature squared (T^2) for several defect concentrations.

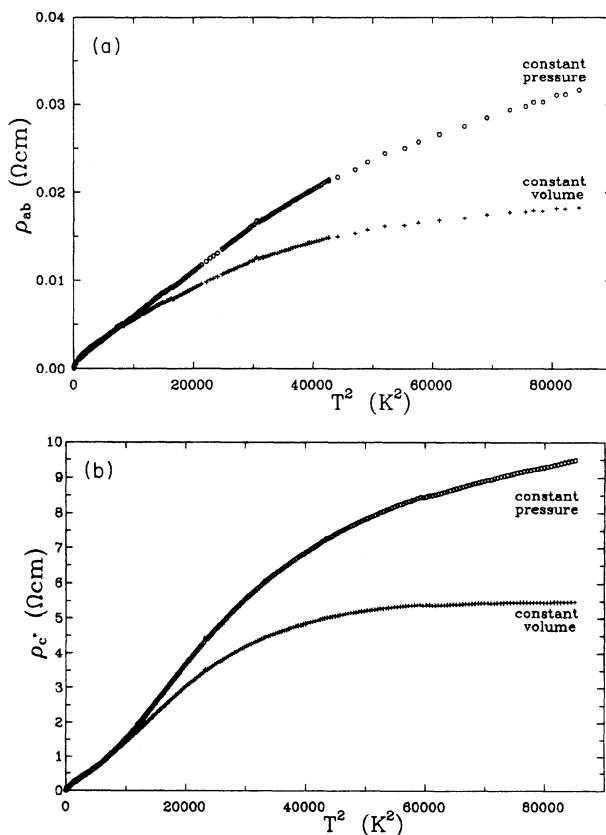


FIG. 7. (a) In-plane (ρ_{ab}) and (b) out-of-plane (ρ_{c^*}) resistivity vs temperature squared (T^2) as measured at constant pressure (points) and corrected to constant volume (crosses).

$\Delta a/a \simeq \Delta b/b \simeq 10^{-2}$ and $\Delta c^*/c^* \simeq 0.5 \times 10^{-2}$ between 80 and 300 K.²⁸ For the volume compressibility we have used $\Delta a/a \simeq \Delta b/b \simeq 3 \times 10^{-3}$ kbar⁻¹ and $\Delta c^*/c^* \simeq 2 \times 10^{-3}$ kbar⁻¹.²⁹ Measured and corrected in-plane and out-of-plane resistivities as a function of temperature squared are displayed in Fig. 7. We have also checked the behavior of $\beta_{\text{low}}\text{-(BEDT-TTF)}_2\text{I}_3$ and obtained similar results. Our study clearly demonstrates that a T^2 law is not obeyed at high temperatures and, therefore, the electron-electron scattering may not be the dominant mechanism in the whole temperature region. However, experimentally observed anomalous frequency-dependent conductivity at RT, as well as the low magnitude of the RT dc conductivity, might be due to important electron-electron correlation effects.⁴ From the RT conductivity value of about $30 (\Omega \text{ cm})^{-1}$ follows an extremely short mean free path of about 0.1 of the intermolecular distance. This result may imply that such a system cannot be interpreted in the framework of standard Landau Fermi-liquid theory. Therefore, the absence of a T^2 law at high temperatures does not necessarily mean that the electron-electron correlations are weak. Here, we would like to point out that even in the low-temperature region (4–80 K) best fits are obtained to the law $\rho_0 + BT^n$ with $n \simeq 1.7$ i.e., with n which is close but not exactly equal to 2.

B. Superconducting phase

We now turn to the question of effects of the irradiation-induced defects on the superconducting transition. We have shown that SC was destroyed by a fairly small amount of defects ($c \geq 0.2\%$ per mole). First, we discuss this result in the framework of the standard theory for singlet superconductivity, which implies the following: (1) The primary effect of irradiation-induced defects was not to increase the potential scattering but to act as pairbreakers for SC; (2) these defects should be magnetic in nature. Indeed, several experimental studies have shown that such defects are often magnetic, whereas alloying introduces nonmagnetic impurities into the system.¹⁸ Therefore, there is no need to introduce the possibility of triplet pairing as was often done previously. However, irradiation defects may not be magnetic in the sense that spins are localized on a molecule; rather, they induce spins localized on segments between defects. Further, the presence of these spins has not been shown experimentally for defect concentrations of less than 0.3% per mole.¹⁸ Therefore, the above picture might not be relevant for the problem. Second, we think that the suppression of superconductivity might be understood in terms of two-dimensional localization: Recently, Mandrus *et al.* have shown that in single crystals of the 2D oxide superconductor $\text{Bi}_2\text{Sr}_2\text{Y}_x\text{Ca}_{1-x}\text{Cu}_2\text{O}_8$ ($0 \leq x \leq 0.8$) superconductivity disappears when the resistivity is 6–8 k Ω per layer.³⁰ This result is in agreement with current theories which predict that the sheet resistance

separating SC and insulating behavior is a universal number $R_{\square} = h/4e^2 = 6.3$ k Ω .³¹ For $(\text{BEDT-TTF})_2\text{AuI}_2$ with conducting (a, b) planes spaced by $c^* = 15$ Å apart, 6.5 k Ω per layer corresponds to a resistivity of 1000 $\mu\Omega$ cm. A 0.2% per mole irradiated sample in which SC was suppressed had a residual resistivity (obtained by extrapolation to zero temperature) of 1120 $\mu\Omega$ cm or $R_{\square} = 7.3$ k Ω per layer in quite good agreement with the theoretical prediction.

In BEDT-TTF compounds superconductivity can also be suppressed by alloying, which was studied by Tokumoto *et al.*¹² They have found in the phase $\beta\text{-(BEDT-TTF)}_2$ trihalide system that SC was suppressed if the magnitude of the residual resistivity was larger than 150 $\mu\Omega$ cm and discussed this result in terms of weak localization. However, 150 $\mu\Omega$ cm corresponds to 1 k Ω per layer and is much less than $h/4e^2$. As far as irradiation-induced defects are concerned, several authors have investigated their influence on SC in $\beta\text{-(BEDT-TTF)}_2\text{I}_3$ material. Tokumoto *et al.* have found that SC is suppressed at the defect concentration of 0.8% per mole in the β_{low} phase and that it remained unaffected in the β_{high} phase at least until 0.48% per mole of defects.¹³ However, these authors assumed the same damage energy as for $(\text{TMTSF})_2\text{PF}_6$ and in that way overestimated the defect concentration by a factor of 4. Therefore, the highest irradiation used in their study of the β_{high} phase was 0.13% per mole, which is still much lower than the one we found to be critical for the suppression of SC in the isostructural $\beta\text{-(BEDT-TTF)}_2\text{AuI}_2$. Further, Forro *et al.* have reported a similar study on $\beta\text{-(BEDT-TTF)}_2\text{I}_3$ samples, but with mixed β_{low} and β_{high} phase structures.²⁰ They found the suppression of SC transition at 8 K at a level of only 0.03% per mole. However, their lowest experimental temperature was 2 K and therefore nothing is known about the resistivity behavior at lower temperatures [note that $T_{\text{SC}} \simeq 1.5$ K in pure β_{low} phase $(\text{BEDT-TTF})_2\text{I}_3$ samples]. Finally, note that the effect of irradiation-induced defects on SC reported in our work is one order of magnitude weaker than that found for the quasi-1D superconductor $(\text{TMTSF})_2\text{PF}_6$.

IV. CONCLUSION

For a level of irradiation damage less than 0.15% per mole we have found that the defects created can be regarded as increasing the temperature-independent scattering rate in the (a, b) conducting planes, while leaving the electrical conductivity perpendicular to the planes unchanged. Electrical transport obeys Matthiessen's rule and can be fitted to a T^2 law up to about 20 K. At higher temperatures the dominant mechanism might still be electron-electron scattering, but a T^2 law is no longer obeyed. Here the short mean free paths obtained may be an indication that Landau Fermi-liquid theory is no longer valid. Higher irradiation doses cause changes in

electronic structure, leading to a different temperature dependence of the conductivity anisotropy below 60 K and deviations from Matthiessen's rule. The superconducting ground state is suppressed, which might be understood with the aid of recent theories of two-dimensional localization, in a similar manner as for high- T_c oxide superconductors.

ACKNOWLEDGMENTS

We would like to thank J. R. Cooper for a critical reading of the manuscript and for bringing our attention to Ref. 30. We also acknowledge useful discussions with I. Batistić, L. Forro, B. Hamzić, K. Maki, E. Tutiš, and L. Zuppiroli.

-
- ¹*The Physics and Chemistry of Organic Superconductors*, edited by G. Saito and S. Kagoshima, Springer Proceedings in Physics Vol. 51 (Springer-Verlag, New York, 1990).
- ²Proceedings of the International Conference on Science and Technology of Synthetic Metals (ICSM'90), Tübingen, 1990 [Synth. Met. **42**, 1991].
- ³B. Korin-Hamzić, Ph.D. thesis, University of Zagreb, 1990 (unpublished).
- ⁴C. S. Jacobsen, J. M. Williams, and H. H. Wang, *Solid State Commun.* **54**, 937 (1985).
- ⁵T. Mori, A. Kobayashi, Y. Sasaki, H. Kabayashi, G. Saito, and H. Inokuchi, *Chem. Lett.* 957 (1984).
- ⁶W. Kang, G. Montambaux, J. R. Cooper, D. Jérôme, P. Batail, and C. Lenoir, *Phys. Rev. Lett.* **62**, 2559 (1989).
- ⁷S. Tomić, J. R. Cooper, W. Kang, D. Jérôme, and K. Maki, *J. Phys. (Paris) I* **1**, 1603 (1991).
- ⁸L. N. Bulaevskii, *Adv. Phys.* **37**, 443 (1988).
- ⁹M. Weger, K. Bender, T. Klutz, D. Schweitzer, F. Gross, C. P. Heidmann, Ch. Probst, and K. Andres, *Synth. Met.* **25**, 49 (1988).
- ¹⁰S. Tomić, D. Jérôme, and K. Bechgaard, in *Low-Dimensional Conductors and Superconductors*, edited by D. Jérôme and L. G. Caron, NATO ASI Series B (Plenum, New York, 1987), p. 335.
- ¹¹M. Y. Choi, P. M. Chaikin, S. Z. Huang, P. Haen, E. M. Engler, and R. L. Green, *Phys. Rev. B* **25**, 6208 (1982).
- ¹²M. Tokumoto, H. Anzai, K. Murata, K. Kajimura, and T. Ishiguro, *Synth. Met.* **27**, A251 (1988).
- ¹³M. Tokumoto, I. Nashiyama, K. Murata, H. Anzai, T. Ishiguro, and G. Saito, *Physica* **143B**, 372 (1986).
- ¹⁴K. D. Carlson, G. W. Crabtree, L. Nuñez, H. H. Wang, M. A. Beno, U. Geiser, M. A. Firestone, K. S. Webb, and J. M. Williams, *Solid State Commun.* **57**, 89 (1986).
- ¹⁵I. D. Parker, D. D. Pigram, R. H. Friend, M. Kurmoo, and P. Day, *Synth. Met.* **27**, A387 (1988).
- ¹⁶K. Bender, I. Hennig, D. Schweitzer, K. Deitz, H. Endres, and H. J. Keller, *Mol. Cryst. Liq. Cryst.* **108**, 359 (1984).
- ¹⁷J. R. Cooper, L. Forro, and B. Korin-Hamzić, *Mol. Cryst. Liq. Cryst.* **119**, 121 (1985).
- ¹⁸M. Sanquer, Ph.D. thesis, Université de Paris-Sud, 1985 (unpublished).
- ¹⁹B. Korin-Hamzić, L. Forro, J. R. Cooper, and K. Bechgaard, *Phys. Rev. B* **38**, 11 177 (1988).
- ²⁰L. Forro, S. Bouffard, and D. Schweitzer, *Solid State Commun.* **65**, 1359 (1988).
- ²¹J. M. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960).
- ²²G. Soda, D. Jérôme, M. Weger, J. Alizon, J. Gallice, H. Robert, J. M. Fabre, and L. Giral, *J. Phys. (Paris)* **38**, 931 (1977).
- ²³M. Sanquer and S. Bouffard, *Mol. Cryst. Liq. Cryst.* **119**, 147 (1985).
- ²⁴L. Zuppiroli, S. Bouffard, K. Bechgaard, B. Hilti, and C. W. Mayer, *Phys. Rev. B* **22**, 6035 (1980).
- ²⁵J. R. Cooper, *Phys. Rev. B* **19**, 2404 (1979).
- ²⁶L. Forro (private communication).
- ²⁷G. Creuzet, J. R. Cooper, F. Creuzet, D. Jérôme, and A. Moradpour, *J. Phys. (Paris) Lett.* **46**, L1133 (1985).
- ²⁸H. Kobayashi, R. Kato, A. Kobayashi, T. Mori, and M. Inokuchi, *Solid State Commun.* **60**, 473 (1986).
- ²⁹V. N. Molchanov, R. P. Shibaeva, V. N. Kachinskii, E. B. Yagubskii, V. I. Simonov, and B. K. Vainstein, *Dok. Acad. Nauk SSSR* **286**, 637 (1986).
- ³⁰D. Mandrus, L. Forro, C. Kendziora, and L. Milhaly, *Phys. Rev. B* **44**, 2418 (1991).
- ³¹M. P. A. Fisher, G. Grinstein, and S. M. Girvin, *Phys. Rev. Lett.* **64**, 587 (1990).