

Article

Spin and Charge Transport in the X-ray Irradiated Quasi-2D Layered Compound: κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl

Ágnes Antal ^{1,2}, Titusz Fehér ^{1,2}, Naoki Yoneyama ³, László Forró ⁴, Takahiko Sasaki ⁵ and András Jánossy ^{1,2,*}

¹ Condensed Matter Research Group of the Hungarian Academy of Sciences, P.O.Box 91, Budapest H-1521, Hungary; E-Mails: agnesantal@gmail.com (Á.A.); tif@phy.bme.hu (T.F.)

² Institute of Physics, Budapest University of Technology and Economics, Budapest H-1111, Hungary

³ Interdisciplinary Graduate School of Medicine and Engineering, University of Yamanashi, Takeda 4-4-37, Kofu, Yamanashi 400-8511, Japan; E-Mail: nyoneyama@yamanashi.ac.jp

⁴ Laboratory of Physics of Complex Matter, École Polytechnique Fédérale de Lausanne, Lausanne CH-1015, Switzerland; E-Mail: laszlo.forro@epfl.ch

⁵ Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan; E-Mail: takahiko@imr.tohoku.ac.jp

* Author to whom correspondence should be addressed; E-Mail: atj@szfki.hu.

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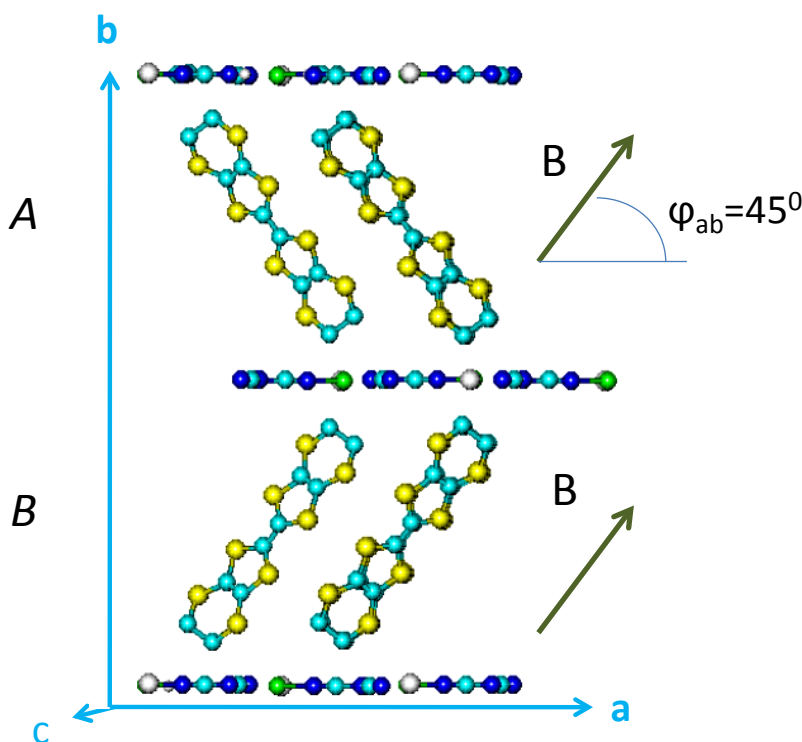
Abstract: The interplane spin cross relaxation time T_x measured by high frequency ESR in X-ray irradiated κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl is compared to the interplane resistivity $\rho_{\perp}$ and the in-plane resistivity $\rho_{\parallel}$ between 50 K and 250 K. The irradiation transforms the semiconductor behavior of the non-irradiated crystal into metallic. Irradiation decreases T_x , $\rho_{\perp}$ and $\rho_{\parallel}$ but the ratio $T_x/\rho_{\perp}$ and $\rho_{\perp}/\rho_{\parallel}$ remain unchanged between 50 and 250 K. Models describing the unusual defect concentration dependence in κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl are discussed.

Keywords: organic; two dimensional; X-ray irradiation

1. Introduction

κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl (κ -ET₂-Cl hereafter) is a layered organic conductor on the borderline of a Mott metal–insulator transition [1,2]. The electronic band is effectively half-filled with one hole per two ET molecules. The electronic structure is two dimensional, layers of ET molecules are separated by polymeric anion layers (Figure 1). Judged from the electrical and magnetic properties of non-irradiated κ -ET₂-Cl, the following temperature ranges are distinguished: (i) Below $T_N = 23$ K: weakly ferromagnetic insulator [3]; (ii) Between 23 and about 50 K: a smooth insulator to a semiconductor transition with an anomalous magnetic field dependent magnetism; (iii) Above 50 K: semiconductor with a very small gap and a temperature-independent paramagnetic susceptibility [4]. In all three temperature ranges the coupling between layers is extremely weak: in ranges (i) and (ii) magnetic oscillations of adjacent layers are independent in general direction magnetic fields; In range (iii) electron hopping between layers is extremely rare. At ambient temperatures electrons diffuse several tenths of a micrometer confined to a single molecular layer [5].

Figure 1. Structure of κ -ET₂-Cl projected onto the (a, b) plane. Only one type of ET molecule per plane is shown for clarity. In the experiments the external field **B** is along $\varphi_{ab} = 45^\circ$, where φ_{ab} denotes the angle from a in the (a, b) plane.



X-ray irradiation at relatively small doses changes drastically the physical properties in all three temperature ranges. Sasaki *et al.* [6] proposed that the deviation from half filling is the underlying reason for the sensitivity to irradiation. The results and discussion of irradiation induced changes of magnetism in range (i) and (ii) will be presented in a forthcoming paper. This paper is restricted to the discussion of the conducting high temperature range. Here the change by irradiation of the resistivity from a semiconductor to a metallic temperature dependence is the most remarkable phenomenon. Section 2

is a brief description of experimental details and the high frequency ESR method to determine the interlayer hopping frequency $\nu_{\perp}$. The relation between interlayer hopping and resistivity (Section 3) is most important for the interpretation. Details of the method and the high frequency ESR results in non-irradiated κ -ET₂-Cl and κ -ET₂-Br were published in References [7,8]. Section 4 presents the experimental results. Irradiation has little effect on both interlayer coupling and intralayer spin relaxation at 250 K and we argue in Section 4.1 that the concentration of irradiation-induced defects is low. In Section 5 we discuss possible mechanisms for the qualitative changes in the electronic transport.

2. Experimental

This study follows previous optical, resistivity, static magnetization and low frequency (9 GHz) ESR measurements on irradiated κ -ET₂-Cl [6,9] and κ -ET₂-Br [10] crystals at Tohoku University (Sendai, Japan). The method of irradiation has been described earlier [6]. Irradiation intensities were about 1/2 MGy per hour. ESR was measured on the same crystal with increasing doses. The total doses of 90 h, 180 h, 360 h, 720 h resulted from incremental irradiation after each ESR measurement. The ESR data on the non-irradiated crystal are taken from measurements on a different crystal. We found that the ESR of non-irradiated crystals are well reproducible. Although the non-irradiated crystal in this study is different from the two crystals in the previous ESR study [7], the results agree well. ESR spectra between 3 K and 250 K were recorded for all irradiation doses in magnetic fields along the crystallographic b and c axes and along $\varphi_{ab} = 45^\circ$ from the a axis in the (a, b) plane. In this paper we present only the $\varphi_{ab} = 45^\circ$ data from which the interlayer hopping rate is determined. The ESR spectra were taken at 111.2 GHz and 222.4 GHz in the BUTE ESR Laboratory [11]. $\rho_{\parallel}$ in-plane and $\rho_{\perp}$ interplane resistivity data were measured on different crystals and with different irradiation doses but under the same conditions as used for ESR. The difficulty of contacting make absolute values of $\rho_{\perp}$ and in particular of $\rho_{\parallel}$ uncertain.

2.1. Method of Measuring $\nu_{\perp}$

The measurement of $\nu_{\perp}$ by ESR is based on the assumption that spin and charge hopping rates are equal. We use the convention $1/T_x = 2\nu_{\perp}$, where $1/T_x$ is the spin cross relaxation rate due to hopping from one layer to the two adjacent layers. In κ -ET₂-Cl T_{2A}^{-1} and T_{2B}^{-1} , the spin relaxation rates of conduction electrons due to processes intrinsic to layers A and B are long (*i.e.*, in the ns range); consequently the ESR spectrum depends sensitively on interlayer hopping.

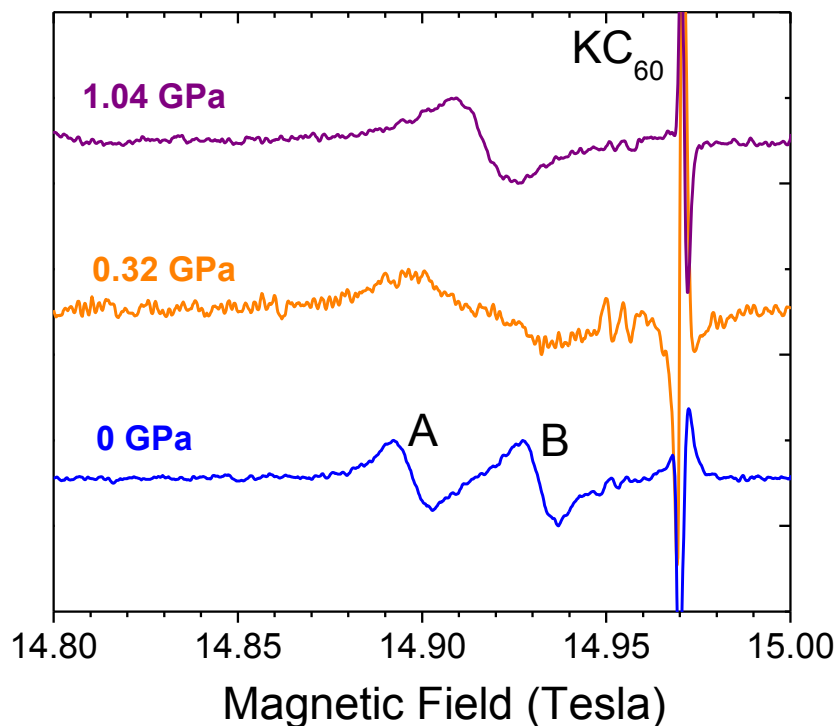
$\nu_{\perp}$ can be measured by electron spin resonance (ESR) in materials like κ -ET₂-Cl where chemically equivalent but structurally different layers (denoted by A and B in Figure 1) alternate [8]. If the A and B layers do not interact, the ESR spectrum is a superposition of two lines at the Larmor frequencies ν_A and ν_B . Since the *g*-factor tensors of A and B layers are differently oriented, the A and B lines are resolved in sufficiently high magnetic fields. In κ -ET₂-Cl the splitting is largest in fields oriented along $\varphi_{ab} = 45^\circ$ in the (a,b) plane while it is zero in the (a,c) plane and in the principal crystallographic directions. For a finite interaction between adjacent layers the ESR spectrum depends on the strength of the interlayer interaction, *i.e.*, on the magnitude of $\nu_{\perp}$:

- (a) $\nu_{\perp} \ll |\nu_A - \nu_B|$. The two lines are resolved; the lineshapes differ only slightly from the non interacting case.

- (b) $\nu_{\perp} \approx |\nu_A - \nu_B|$. The spectrum has a complicated lineshape.
- (c) $\nu_{\perp} \gg |\nu_A - \nu_B|$. A “motionally narrowed” line appears at the frequency $(\nu_A + \nu_B)/2$. The linewidth depends on $\nu_{\perp}$.

The three cases are demonstrated in Figure 2 for κ -ET₂-Cl. Here $\nu_{\perp}$ was increased by applying pressure [7].

Figure 2. Motional narrowing of the ESR lines of adjacent *A* and *B* ET layers in κ -ET₂-Cl under pressure (after [7]). $\mathbf{B} \parallel \varphi_{ab} = 45^\circ$. The spectra were measured at 420 GHz, $T = 250$ K and pressures of 0, 0.32 and 1.04 GPa. KC₆₀ is an ESR reference.



To extract $\nu_{\perp}$ and T_2^{-1} , the lineshapes are fitted to spectra calculated from two coupled Bloch equations. The electronic exchange between layers represented by an effective magnetic field is also included in the fit. In this paper all data were obtained by fitting ESR spectra recorded at two frequencies, 222.4 GHz and 111.2 GHz. g_A and g_B were also free parameters, they change little with temperature.

3. Relation between Interlayer Hopping $\nu_{\perp}$ and Resistivity $\rho_{\perp}$

$\nu_{\perp}$ and $\rho_{\perp}$ are closely related [12]. The resistivity is proportional to the hopping time and inversely proportional to the density of available states, D . The electrical current j is given by

$$j = e\nu_{\perp}\Delta\mu D/F \quad (1)$$

where $\Delta\mu = e\mathcal{E}d$ is the potential difference between adjacent layers due to the electric field $\mathcal{E}$. The relation is particularly simple if the electrons of molecular layers form a Fermi liquid. Then $D = D(E_F)$ is the density of states (DOS) per ET dimer for both spin directions of the metallic layers at the Fermi energy, E_F . The perpendicular conductivity calculated from the hopping rate is

$$\rho_{\perp}^{-1} = e^2 D(E_F) \nu_{\perp} d / F \quad (2)$$

where $1/F$ is the two dimensional charge carrier density. Assuming one hole/dimer in κ -ET₂-Cl, $F = (ac)/2$, where a and c are the in-plane, $b = 2d$ the out-of-plane lattice constants. The significance of Equation (2) lies in the possibility to determine the DOS from measurements of the perpendicular resistivity and spin cross relaxation time without knowing details of the barrier between layers:

$$T_x/\rho_{\perp} = \frac{1}{2}e^2(d/F)D(E_F) \quad (3)$$

In a simple semiconductor with a gap U and phonon assisted hopping with attempt frequency ν_0 , the hopping rate is given by $1/(2T_x) = \nu_0 e^{-U/k_B T}$ and $D(E_F)$ is replaced by $(k_B T)^{-1}$ in Equation (3).

Interlayer hopping measured by ESR in non-irradiated κ -ET₂-Cl and κ -ET₂-Br crystals has been discussed in detail in Reference [7]. At 250 K the interlayer hopping time is $T_x = 2.6 \pm 0.5$ ns for the Cl compound and the same for the Br compound within experimental accuracy. The conduction is quasi-two-dimensional: at 250 K electrons are confined to single layers for times several orders of magnitude longer than the momentum relaxation time. The density of states estimated from Equation (2) is about a factor of 5 larger than calculated from the band structure. Between 250 K and 50 K T_x increases rapidly with decreasing temperature in the Cl compound while it is about constant in the Br compound.

4. Interlayer Hopping Rate and Resistivity above the Metal Insulator Transition

The experimental data of the cross relaxation time $T_x = 1/(2\nu_{\perp})$ and the perpendicular resistivity $\rho_{\perp}$ above the metal-insulator transition at various irradiation doses are summarized in Figure 3. The irradiation doses are in the same range in the two experiments. The scales of T_x and $\rho_{\perp}$ are chosen in the figure so that the experimental points of T_x and $\rho_{\perp}$ coincide at 250 K for the non-irradiated sample. Figure 4 displays the in-plane and interplane resistivities $\rho_{\parallel}$ and $\rho_{\perp}$ respectively of non-irradiated and 300 h irradiated samples. The resistivities in Figures 3 and 4 are similar to the ones measured on a different crystal in Reference [6]. The resistivity $\rho_{\perp} = 60 \Omega \text{ cm}$ in this work agrees reasonably well with the measurements of Reference [13].

We note from Figure 3 that the interlayer hopping time and the perpendicular resistivity are to a good approximation proportional for all irradiation doses and in the full temperature range above 50 K. The non-irradiated sample has a semiconducting behavior with T_x and $\rho_{\perp}$ smoothly increasing as the temperature is decreased. On the contrary, the 180 h and more irradiated samples have a metallic like behavior: T_x and $\rho_{\perp}$ decrease with decreasing temperature. In the irradiated samples the abrupt metal-insulator transition is qualitatively different from the smooth transition in the non-irradiated sample. Furthermore, irradiation shifts the transition to lower temperatures.

Figure 4 shows that $\rho_{\perp}$ and $\rho_{\parallel}$ have similar temperature and irradiation dependence. The ratio $\rho_{\perp}/\rho_{\parallel}$ is about 600 independent of temperature for both the irradiated and non-irradiated samples. This ratio, even if uncertain, is typical for measurements in the literature.

Figure 3. Comparison of the temperature dependence of the spin hopping time (T_x) and interlayer resistivity ($\rho_{\perp}$) in κ -ET₂-Cl. Left scale: T_x after 0, 90, 180, 360 and 720 h of irradiation. Right scale: $\rho_{\perp}$ after 0, 100, 150 and 300 h of irradiation.

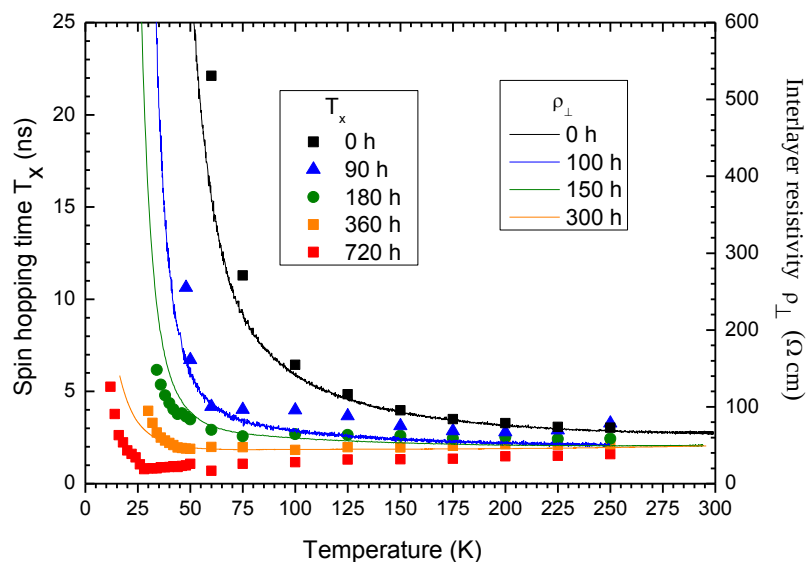
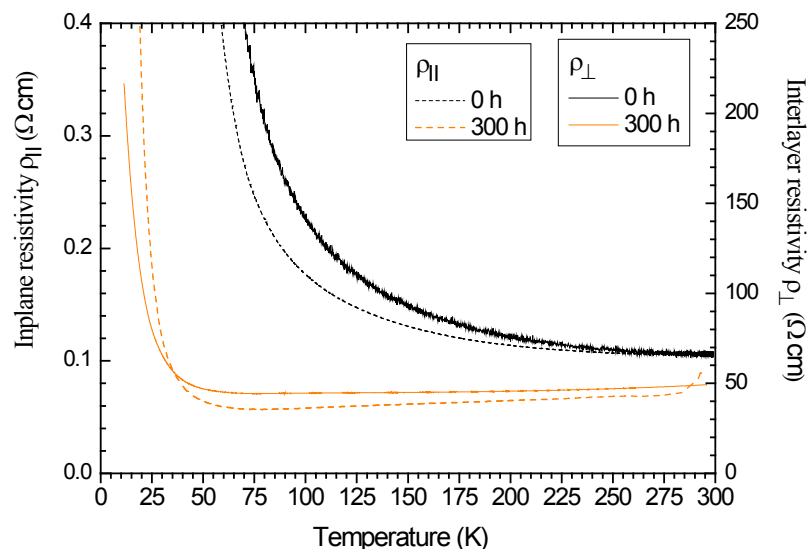


Figure 4. Comparison of in-plane resistivity $\rho_{\parallel}$ and interlayer resistivity $\rho_{\perp}$ in non-irradiated and 300 hours irradiated κ -ET₂-Cl. $\rho_{\parallel}$ and $\rho_{\perp}$ were measured on different samples.



4.1. Irradiation Dependence of $\nu_{\perp}$ and T_2^{-1} at 250 K

The 250 K ESR spectra change little under irradiation (Figure 5a). Even at the highest irradiation dose the interlayer hopping rate remains low since the *A* and *B* layer lines are well resolved. Figure 5b shows the cross relaxation T_x and intrinsic relaxations T_{2A} and T_{2B} calculated from the spectra. T_x decreases by less than a factor of 2 for the largest dose while T_{2A} and T_{2B} are not changed significantly by the irradiation.

The mean free path within ET layers is shorter than the molecular separation, implying a very short in-plane electronic momentum lifetime. In contrast, out of plane hopping events are rare, $\nu_{\perp}$ in κ -ET₂-Cl

is about 10^9 s^{-1} and electrons diffuse to long distances without hopping to adjacent layers. Although the irradiation increases somewhat $\nu_{\perp}$, electrons remain confined to single molecular layers for long times. In addition, T_2 , the electronic spin relaxation time within the conducting ET layers, is independent of irradiation. Usually charged defects effectively increase the spin relaxation rate in conductors. The insensitivity of T_2 to irradiation and the relatively small decrease in T_x are in agreement with earlier findings [9] that the concentration of induced defects is small even under the largest dose.

Figure 5. (a) ESR spectra of κ -ET₂-Cl at 222.4 GHz and 250 K after different doses of X-ray irradiation. There is a small impurity line at 7.935 T in the spectrum of the sample irradiated for 180 h, not present in the other spectra. The signal at 7.943 T is the KC₆₀ reference; (b) Irradiation dose dependence of T_x and T_2 at 250 K.

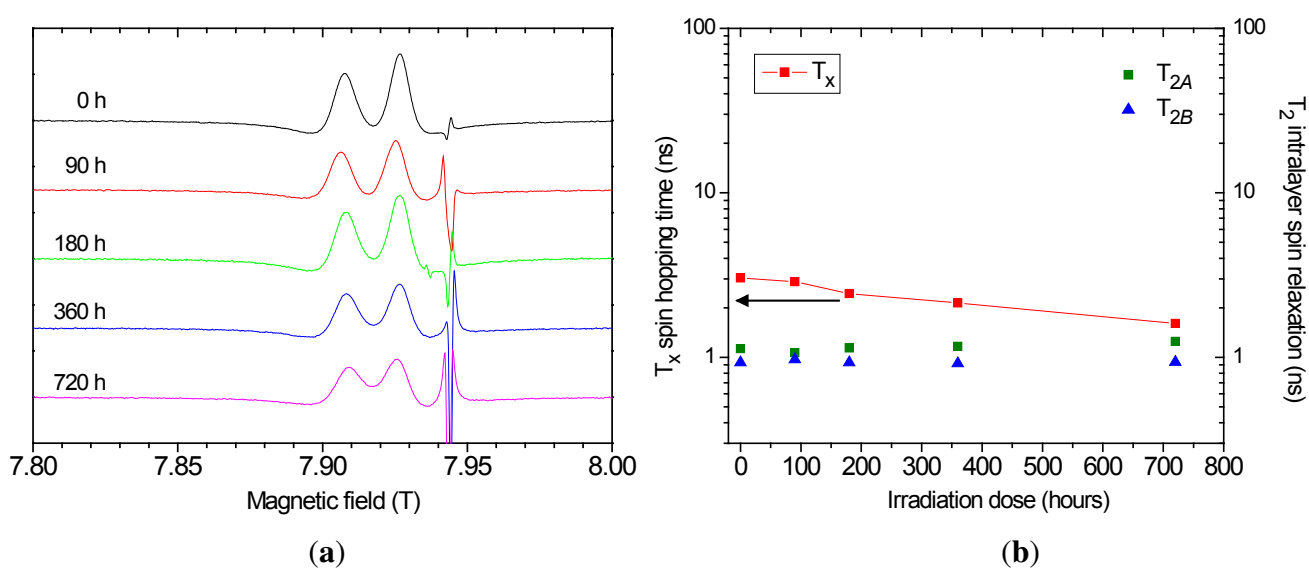
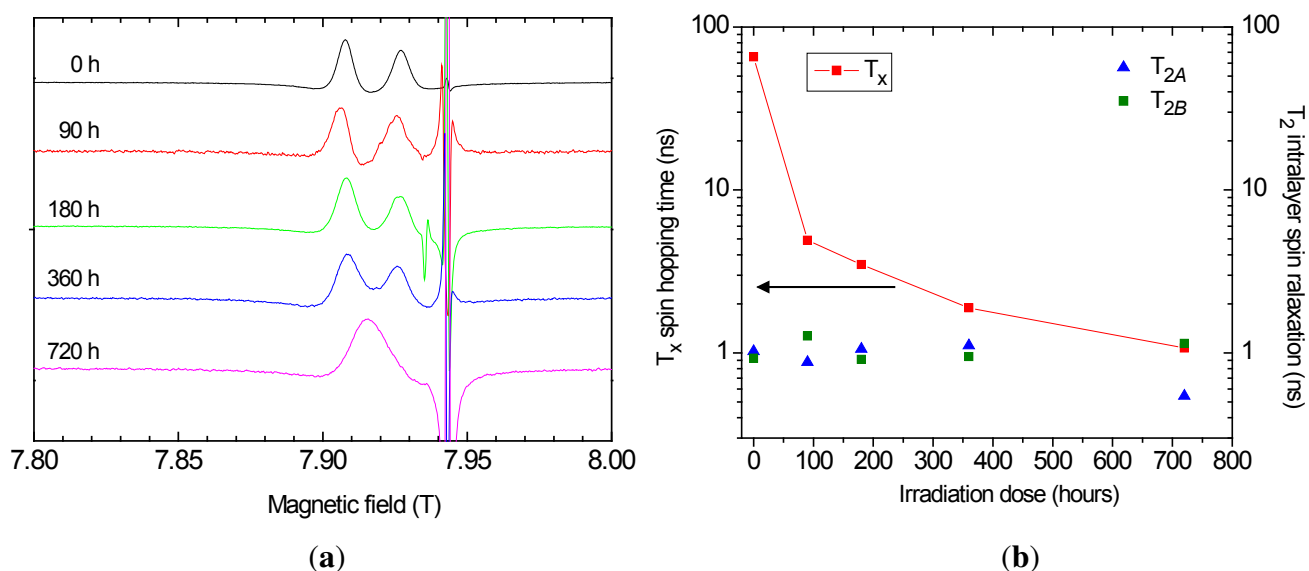


Figure 6. (a) ESR spectra of κ -ET₂-Cl at 222.4 GHz and 50 K as a function of X-ray irradiation dose. The line at 7.935 T for 180 h irradiation, not present in the other spectra, is an impurity line. The signal at 7.943 T is a KC₆₀ reference; (b) Irradiation dose dependence of T_x and T_2 at 50 K.



4.2. Irradiation Dependence of $\nu_{\perp}$ and T_2^{-1} at 50 K

Figure 6 displays the irradiation dependence of the ESR spectra at 222.4 GHz and the spin relaxation times. The interlayer hopping time T_x decreases rapidly with irradiation, especially at low doses. On the other hand, the intrinsic spin relaxation is not sensitive to the irradiation. The single ESR line at 720 h irradiation has an ESR-frequency-dependent width and illustrates the “motional narrowing” case described in Section 2.1.

5. Discussion

We first list the main findings of the present and earlier works on the conducting and magnetic properties in the temperature range between 50 K and 250 K. (Below this temperature the properties change).

- (1) The perpendicular resistivity $\rho_{\perp}$ is semiconducting-like in non-irradiated κ -ET₂-Cl. Irradiation decreases $\rho_{\perp}$ in the full temperature range, the decrease is non-linear with dose. At higher doses the resistivity is metallic, *i.e.*, it increases linearly with temperature.
- (2) The interlayer spin hopping time T_x and $\rho_{\perp}$ have the same temperature and irradiation dose dependence. The ratio $T_x/\rho_{\perp}$ is independent of temperature and irradiation dose.
- (3) The resistivity anisotropy $\rho_{\perp}/\rho_{\parallel}$ is typically between 100 and 1000. It is independent of temperature and irradiation dose.
- (4) The magnetic spin susceptibility is approximately temperature independent and does not change with irradiation [9].
- (5) The intralayer spin relaxation time T_2 is independent of temperature and dose.

The interpretation of the qualitative change in transport properties by irradiation in κ -ET₂-Cl poses a difficult problem. Matthiessen’s rule describes the change of resistivity by defects in usual metals. Impurity scattering in metals increase the resistivity by a temperature independent quantity, the increase is linear with defect concentration and the temperature dependent phonon resistivity is not affected. Clearly, nothing of this applies to κ -ET₂-Cl: irradiation *decreases* the resistivity, the decrease is non-linear with defect concentration and the temperature dependence is drastically changed from semiconducting like to metallic.

At the same time other quantities that depend sensitively on the electronic structure are independent of temperature and defect concentration. In Fermi liquids the magnetic spin susceptibility χ and the perpendicular hopping time to resistivity ratio $T_x/\rho_{\perp}$ are both proportional to the density of states. Although κ -ET₂-Cl is a strongly correlated system and at high temperatures there are no long lifetime quasi-particles, the independence of the susceptibility and the $T_x/\rho_{\perp}$ ratio indicate that the electronic structure is not strongly affected by the irradiation. The defect dose independence of T_2 , the relatively small decrease of T_x at 250 K with dose and earlier magnetic measurements [9] indicate that the defect concentration is small.

Sasaki *et al.* [6] proposed that irradiation of organic layered compounds has two effects: it increases the electron momentum scattering rate and at the same time dopes the material. By creating localized electrons at defects in the anion layer and delocalized holes in the conducting molecular layers a charge

imbalance from half filling takes place. In metallic compounds far from the Mott transition, like κ -(BETS)₂FeCl₄, defects enhance the electron scattering as in other metals. In materials close to the Mott metal-insulator transition, doping of holes changes the electronic properties drastically as the band is no more half filled. At high temperatures the extra holes change the semiconducting behavior to metallic, at low temperatures the metal-insulator and the magnetic ordering transitions are suppressed. To understand the measured electric and magnetic properties one has to assume that the small deviation from half band filling changes the resistivity from semiconducting to metallic-like but has little effect on the band structure in general.

It is not simple to understand within the model the independence of the resistivity anisotropy from irradiation induced defect concentration. Why does adding high mobility carriers increase the in-plane conductivity and the interlayer hopping time in the same way? Kumar and Jayannavar [12] proposed a mechanism for a temperature independent anisotropy $\rho_{\perp}/\rho_{\parallel}$ in materials where the perpendicular conductivity is incoherent. They showed that if the in-plane scattering time τ is much shorter than the interlayer hopping time $1/\nu_{\perp}$, then the ratio $\rho_{\perp}/\rho_{\parallel}$ is independent of τ . To understand the doping independence of $\rho_{\perp}/\rho_{\parallel}$ one has to assume that irradiation introduces carriers with an increased τ but at the same time the parallel and perpendicular electronic overlap integrals remain unchanged.

A different approach has been proposed by Analytis *et al.* [14]. They assumed that irradiation creates two kinds of defects. Like in the model of Sasaki *et al.*, some defects increase the electronic scattering rate. There is a further defect-assisted interlayer channel which decreases the interlayer resistivity. This model gives a good description of the irradiation dose dependence of $\rho_{\perp}$ in κ -(BEDT-TTF)₂Cu(SCN)₂, an organic conductor close to the Mott transition. The in-plane resistivity is assumed to be small and without any effect on $\rho_{\perp}$. A defect-assisted interplane conductivity has also been proposed [15] to explain the metallic zero frequency interplane conductivity and the absence of a Drude peak in the optical conductivity in κ -ET₂-Br. However, it is difficult to understand within this model the irradiation and temperature independence of the anisotropy between 50 and 250 K in κ -ET₂-Cl. Due to difficulties in contacting and/or macroscopic sample imperfections, the current is usually inhomogeneous in strongly anisotropic conductors and in general the interlayer resistivity affects in-plane measurements. If $\rho_{\parallel}$ is very small, the contribution of $\rho_{\perp}$ might dominate the apparent in-plane resistivity. This would explain that $\rho_{\perp}$ and $\rho_{\parallel}$ have (apparently) the same temperature dependence both for the non-irradiated and irradiated crystal. However, this model does not explain why the anisotropy is independent of irradiation dose.

6. Conclusions

In κ -ET₂-Cl, the interplane spin cross relaxation time measured by high frequency ESR is proportional to the interplane resistivity in the temperature range between 50 and 250 K. The ratio $T_x/\rho_{\perp}$ is unchanged in the range of irradiation doses where the resistivity changes from a semiconductor-like behavior to metallic. Since κ -ET₂-Cl is a strongly correlated conductor close to the metal insulator and magnetic ordering transitions, the spin and charge hopping rates could have different temperature or doping dependence. The insensitivity of $T_x/\rho_{\perp}$ to doping and temperature makes this unlikely. The qualitative change of the temperature dependence of the resistivity with irradiation and the insensitivity

of other quantities depending on the density of states poses a difficult problem, which is not yet satisfactorily resolved.

Acknowledgements

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