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Giant Magnetoresistance in Organic Superconductors κ−(BEDT-TTF)$_2$Cu(NCS)$_2$

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Abstract

In this letter, we report transport measurements with field and current parallel to the *a* axis (perpendicular to the conducting plane) in the organic superconductor κ−(BEDT-TTF)$_2$Cu(NCS)$_2$.The magnetoresistance displays a peak effect as a function of field and temperature with the peak fields increasing linearly with decreasing temperatures. The peak resistance is found to be greater than the normal state value extrapolated from both high and low field measurements. This is a first report of above normal resistance in a superconducting state. The results are in sharp contrast to the conventional dissipation mechanisms in the mixed state for anisotropic superconductors, as in the case of $Bi_2Sr_2CaCu_2O_8$. We propose a phenomenological model that the peak in the magnetoresistance is caused by a new scattering mechanism due to a strong coupling to the underlying crystal lattice of fluctuating vortices (vortex polarons). The model can semiquantitatively fit the data.

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Organic charge transfer salts, especially the layered BEDT-TTF [bis(ethylenedithio)-tetrathiafulvalene, abbreviated as ET] family have been of recent interest [1]. $\kappa-(ET)_2X$ with X being $Cu(NCS)_2$ undergoes a superconducting transition with the T_c near 10K at ambient pressure. These materials have an intrinsic layered structure consisting of alternating sheets of metallic (dimerized ET molecules) and insulating (anion, X) planes. Transport measurements show very large anisotropy for conduction parallel and perpendicular to the conducting planes (bc plane) [2]. Unlike most of the high temperature oxide cuprates where charge transport is also 2D-like, the organic $\kappa-(ET)_2X$ family is extremely sensitive to the applied pressure [3,4]. The fact that superconductivity in $(TMTSF)_2PF_6$ was first discovered by the application of pressure shows that the pressure is one of the key parameters in better understanding organic superconductors. Among all organic superconductors, $\kappa-(ET)_2Cu(NCS)_2$ has the largest pressure dependence for T_c ever reported, with dT_c/dP = -3K/kbar initially. The large dT_c/dP value suggests that lattice distortions and thermal fluctuations will play an important role in the charge transport of this system.

In this letter, we report transport measurements with field and current parallel to the *a* axis (perpendicular to the conducting plane). Measurements have been performed at fixed temperatures (field sweep) and fixed fields (temperature sweep). Our results show an anomalously large magnetoresistance below the superconducting transition temperature. The peak resistance is greater than the normal state value extrapolated from both high and low fields measurements. The peak field increase linearly with decreasing temperature with the maximum resistance occurring around 5K. This result is in sharp contrast with the conventional Bardeen-Stephen model, where flux-flow resistivity is always less than the normal state value for field $H \leq H_{c2}$. It is also inconsistent with a simple resistively shunted Josephson junction model where thermal fluctuations give rise to a junction resistance, as suggested in the case of $Bi_2Sr_2CaCu_2O_8$ superconductors. We propose that the peak in the magnetoresistance is caused by an additional scattering mechanism due to a strong coupling to the underlying crystal lattice of fluctuating vortices.

Single crystals of the κ-$(ET)_2Cu(NCS)_2$ superconductor were synthesized by the electrocrystallization technique described elsewhere [5]. Several crystals were used in these measurements with average dimensions of $1\times0.86\times0.33$mm. Extensive measurements were made on one crystal with T_c= 10.2K. Measurements were performed at the National High Magnet Field Laboratory with field up to 18T. The interlayer resistance was measured using the four probe technique with the current leads covering most of the faces. Typical contact resistances between the silver paint and the sample were about 2-5 Ω. A current of $1\mu A$ was used to ensure the linear I-V characteristic. The voltage was detected with a lock-in amplifier at low frequencies of about 312 Hz. We have also checked the two probe configurations with both faces covered completely to ensure current uniformity. Similar results with a slightly temperature dependent contact resistance were obtained. Results presented in this letter are from the four probe measurements. The samples were cooled slowly to below the superconducting transition temperature with the field parallel to the crystallographic a-axis.

Shown in figure 1 is an overlay of the isothermal magnetoresistance versus applied magnetic field at low temperatures of T=2K, 2.5K, 3K, 3.5K, 4K, 4.5K, and 5K. Clearly, a pronounced peak in the interlayer resistance is observed at a peak field H_{peak} for each fixed temperature. For example, at T=2K, the resistance R reaches a maximum of about 4 Ω at $\simeq$ 3T. For low fields $H < H_{peak}$, R decreases rapidly to zero with an onset field of 1.8T. For intermediate fields, R dips into a local minimum at H_{min}. For high fields $H > H_{min}$, R rises again with increasing fields. An oscillatory magnetoresistance commencing at about 12T is observed at this temperature, as shown in the inset. The observation of the Shubnikov-de Haas effect at T=2K demonstrates the high crystal quality of this material. At higher temperatures, similar features are observed. The characteristic field H_{peak} shifts toward lower field value and the peak resistance R_{peak} increases in magnitude. With increasing temperature, the local minimum at H_{min} becomes broader with a slightly decreasing but comparable dR/dH coefficient at higher fields.

For temperature above 5K, qualitatively similar results are observed as shown in figure 2 at temperatures T= 6K, 7K, 8K, 9K, 10K and 11K. In contrast with the low temperature counterpart, the peak resistance R_{peak} decreases with increasing T for T> 5K and the local minimum becomes barely visible in the scale shown. H_{peak} decreases monotonically with increasing T. At T=11K, the system is completely in the normal state. The inset plots an expanded view of the high temperature magnetoresistance. The three parallel lines are guides for the eye that dR/dH is positive and has the same magnitude of $0.1\Omega/T$ at these temperatures and fields.

Figure 3 is a plot of the peak field as a function of temperature. A linear temperature dependence of H_{peak} with $dH_{peak}/dT = 0.37\pm0.05$ TK^{-1} is observed. If we assume a simple form for the peak field $H_{peak}=H_o(1-T/T_c)$, we find H_o to be 3.8 ± 0.5T and the T_c extrapolated this way is 10.2K. This is similar to the magnetically determined T_c, but somewhat less than the transport onset temperature due to fluctuations [6].

The peak resistance as a function of temperature is shown in figure 4. Unlike the monotonic temperature dependence seen in H_{peak}, R_{peak} reveals a maximum value at about 5K. The decrease in R_{peak} is much weaker for T> 5K than that for T< 5K. Overlaid on figure 4 is also the magnetoresistance as a function of temperature at fixed field of H=10T, which is above H_{min} and H_{c2} for all temperatures studied here. The dashed line is an extrapolation from the high temperature normal state R_{normal} resistance to low temperatures. Clearly the R_{peak} is much larger than R(H=10T) and R_{normal}.

The anomalously large magnetoresistance observed in this study has not been recognized in this context in the previous studies of organic superconductors. Rather it has been frequently used to define the upper critical field $H_{c2}(T)$ [7,8]. In the case here, the absence of the peak and local minimum in the R(H) for $T > T_c$ and the normal behavior (positive dR/dH) at high H for $T < T_c$ suggest strongly that it is a property of the mixed state. Comparisons with $H_{c2}(T)$ determined magnetically in similar materials provide another support that the peak field is less than $H_{c2}(T)$. We analyze the data below based on this assumption.

Dissipation mechanisms in the mixed state have been studied extensively for the high T_c cuprates [9,10]. However, to our knowledge, there is no theoretical model predicting above normal state resistance in the superconducting state. In fact, our results are in sharp contrast with the conventional dissipation mechanisms. In the case of flux flow, the dissipation is caused by the motion of vortices which induce an electric field inside and outside the vortex core. The effective resistance is given by the Bardeen-Stephen model [11], $\rho_f = \rho_n \frac{H}{H_{c2}}$ where ρ_f is the flux-flow resistivity and ρ_n is the normal state resistance in the core region. Experimentally, the normal state resistivity ρ_n is obtained by extrapolating the data above the superconducting transition temperature. In the case here the current is parallel to the applied field, and there is no Lorentz force acting on the vortices. The flux flow model thus can not be applied. However, ρ_f typically sets the scale for resistivity in the mixed state.

Transport for the current parallel to the field geometry has been discussed in several recent papers [12–15]. In the highly anisotropic cuprate superconductor $Bi_2Sr_2CaCu_2O_8$, it has been reported that resistance in the direction perpendicular to the superconducting layers peaks at a temperature below T_c. In a model proposed by Briceño *et al* [12], current moving parallel to the c axis is taken to pass through a narrow superconducting channel of area $A = a^2 \approx \frac{\Phi_o}{H}$ between the densely packed vortices. Here a is the intervortex separation and Φ_o is the flux quantum. Dissipation occurs through thermodynamic fluctuations which cause the phase of the superconducting order parameter in the c direction to jump by 2π. Assuming fluctuations in each channel are independent, the dissipation in the c direction can be modeled by a long, narrow Josephson junction with thermal fluctuations [16]. The resistance of the weak link at finite temperature is given approximately by $R = R_n[I_o(\hbar I_c/2ekT)]^{-2}$,where R_n is the normal state resistance, $\hbar$ is the Planck's constant, I_c is the critical current, e is the charge of an electron, and I_o is the modified Bessel function. Using an empirical expression for the normal state resistance $R_n \sim T^7 \exp(\Delta/kT)$ and $I_c \sim (1 - T/T_c)^{3/2}$, the temperature dependence of the peak was able to be fit. In a similar spirit, Gray *et al* [13] proposed that the data can be explained by modeling the c axis conduction as a series of stack of Josephson tunnel junctions. For an intermediate Josephson coupling, the junction conductance is the sum of the quasiparticle conductance Y_{ss} and pair conductance Y_p, i.e. $Y = Y_{ss} + Y_p$. Since the quasiparticle conductance Y_{ss} is thermally activated $Y_{ss} \sim exp[-\Delta(T)/kT]$, and the pair conductance $Y_p \sim [I_o(\hbar I_c/2ekT)]^{-2} - 1$, a distinct peak is also predicted. The model has been used successfully to explain the data on a thin film junction and $Bi_2Sr_2CaCu_2O_8$ crystals. Furthermore, the model predicts the normal state resistance to increase with decreasing temperature above T_c as well, which is not discussed by Briceño *et al.*

Both models successfully explain the peak observed in the c axis resistance below the transition temperature. However, the maximum resistance is always limited by the normal state resistance at the corresponding temperature. Experimentally, the c axis resistance at higher fields is always greater than that at lower fields. In the case of $\kappa-(ET)_2Cu(NCS)_2$, the normal state resistance R_{normal} is quasi-metallic and the out of plane resistivity decreases with decreasing temperatures. For temperature up to 20K, R_{normal} in zero field can be fitted with $R_{normal}=2.55+0.018\ T^{2.26}$. The dashed line in figure 4 are values extrapolated from this fit to lower temperatures. To provide another check, we estimate the normal state value from the magnetoresistance at H=10T. If we neglect the magnetoresistance at low fields and approximate the high fields magnetoresistance coefficient $dR/dH \sim 0.1\Omega/T$ to be a constant, the "normal state" resistance for $T< T_c$ should be shifted by 1Ω from the H=10T data. A down shift of 1Ω will correspond roughly to the R_{normal} as shown in figure 4. A more careful analysis of the R(H) data supports this simple estimation. The peak resistance at T=5K is almost twice as large as its normal state value. This anomalously large magnetoresistance is thus inconceivable in terms of a Josephson junction model with a normal state junction. An additional scattering mechanism is needed for this extra interlayer resistance!

We propose a phenomenological model that the anomalously large magnetoresistance is due to a new lattice distortion and thermal fluctuations of vortices. If the lattice is soft, such is the case for organic superconductors, the coupling of the vortex core to the crystal lattice can lead to a significant local distortion in the lattice structure. The distortions can be characterized by an effective strength θ over a length scale $\ell \sim 2\zeta(T)$ (This lattice distortion associated with a vortex defines the vortex polaron). Such vortex polarons can give rise to a new scattering mechanism for the charge transport.

Similar to the models proposed for the layered oxide superconductors [12,14], we consider transport along an effective Josephson junction of area $a^2 \approx \frac{\Phi_o}{H+H_o}$ between the densely packed vortices, H_o being a fitting parameter. The junction resistance due to thermal fluctuations of the phases is given by $R(H) = R_{n'}[I_o(\frac{E_J}{2kT})]^{-2}$, where $E_J = \frac{\hbar I_c}{e} = \frac{\pi\hbar\Delta(T)}{2e^2 R_{n'}}\tanh[\frac{\Delta(T)}{2kT}]$ is the Josephson coupling energy, $R_{n'}$ is the new junction resistance in the presence of lattice distortions. As I_c depends on the junction geometry, E_J is thus an extrinsic quantity. However, the high field data can be used to determine the intrinsic Josephson coupling energy $e_J = \frac{E_J}{a^2}$, such that $R(H) = R_{n'}[I_o(\frac{e_J\Phi_o}{(H+H_o)2kT})]^{-2}$. If $E_J >> kT$, the junction resistance can be reduced to [14]

$$R(H) = R_{n'}\exp[-\frac{e_J\Phi_o}{(H+H_o)kT}]. \tag{1}$$

To test the model, we have fit the low temperature data to the above expression, as shown by the solid lines in figure 5. Clearly, the model fits quite well the R(H) data over a broad field range. Deviations near the H_{peak} are expected, as discussed below. The inset is a plot of the intrinsic Josephson coupling energy as a function of T. The dashed line is a linear fit to $e_J = a + bT$ with $a = 1.4\times10^{-7}\mathrm{Jm^{-2}}$ and $b = -2\times10^{-8}\mathrm{Jm^{-2}K^{-1}}$. H_o changes from about -0.1T at 6K to about -0.5T at 2K. The junction resistance increases rapidly with decreasing T, with $R_{n'} =$ 10, 18, 60, 174 Ω for T =6.5, 4.5, 3, 2K, respectively.

The peak in the isothermal field dependence of the resistance can be easily understood in this picture. At small vortex densities, the junction resistance increases exponentially. At higher vortex density, when the fields due to neighboring vortices have substantial overlaps, the effective distortion ℓ will be reduced, resulting in a decrease in the junction resistance $R_{n'}$. For $H \geq H_{c2}$, the field is uniform within the sample, the additional scattering due to the distortions disappears, resulting in a reentrance of the normal state property. The temperature dependence of the peak field can be qualitatively understood if we assume the distortion length $\ell(T)$ to be comparable to the intervortex separation, $a = 1.075\sqrt{\frac{\Phi_o}{H}}$ for a triangular lattice. Since $\ell(T) \sim 2\zeta(T) \sim \sqrt{\frac{1}{1-\frac{T}{T_c}}} \sim \sqrt{\frac{\Phi_o}{H}}$, this model gives a linear temperature dependence of the peak field $H_{peak} \sim (1-\frac{T}{T_c})$, as shown in figure 3. The competing effect of decreasing $R_{n'}$ and increasing exp(-E_J/kT) with increasing T naturally explains the peak in $R_{peak}(T)$.

For an order of magnitude estimate, we check the length scales involved in this simple model. For example, at T=4.5K, the peak field is about 2T, corresponding to an intervortex separation a = 345Å. Using results [17–19] from magnetic measurements ζ_o = 70Å, the coherence length at this temperature can be estimated to be $\frac{\zeta_o}{\sqrt{1-T/T_c}} \sim 100$Å. The distortion length $\ell(T) \sim 200$Å is thus comparable to the intervortex separation, in an agreement with our model. We point out that the peak field is still much less than upper critical field at this temperature of 3.3T. H_{c2} in this case is closely related to H_{min}, above which the large positive dR/dH dominates the normal state transport. The increase of $R_{n'}$ at low T is consistent with pressure dependent studies of these materials. For example, an increase of more than 10 fold in resistance was reported when the sample was under a tensile stress along the b axis [4]. However, a lack of systematic studies at different temperatures makes it difficult to estimate the effective distortion due to vortex lattice interactions.

In summary, we have reported a giant magnetoresistance effect observed on an organic superconductor $\kappa-(ET)_2Cu(NCS)_2$. The magnetoresistance displays a peak as a function of the field and temperature in the superconducting state. The peak field increases linearly with decreasing temperatures with the peak resistance being much larger than the normal state resistance. We propose that the giant magnetoresistance is due to a lattice distortion via coupling with the quantized vortices. The distortions give rise to a new scattering mechanism in the model of Josephson junction resistance at finite temperatures. The phenomenological model can semiquantitatively explain the peak effect as well as its field and temperature dependence.

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Figure Captions.

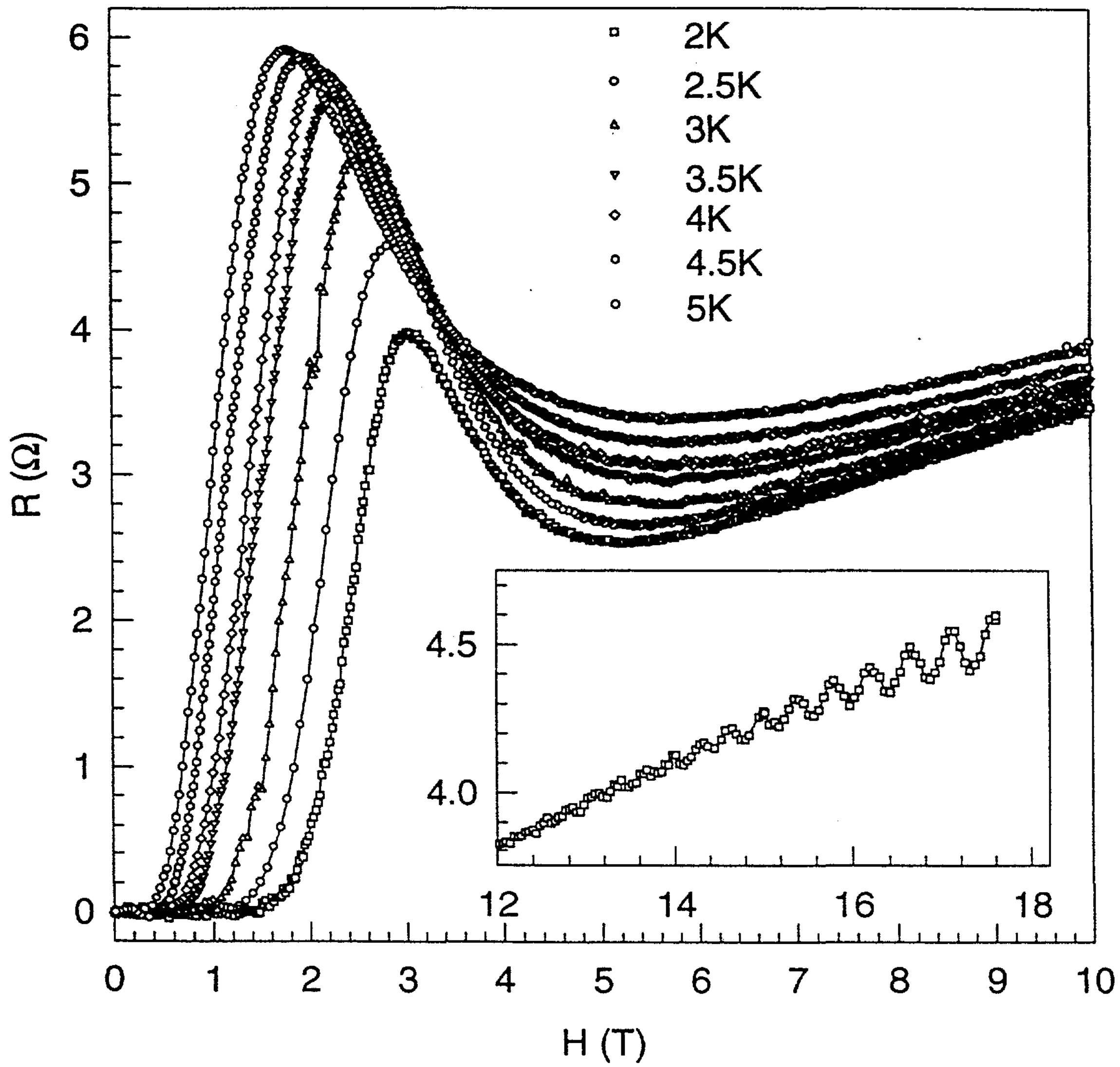
Fig. 1 Magnetoresistance as a function of field at low temperatures. The inset is an expanded view of R(H) at T=2K.

Fig. 2 Magnetoresistance as a function of field at high temperatures. The inset includes normal state R(H) at T=11K.

Fig. 3 Peak field as a function of temperature. The line is a linear fit to the data.

Fig. 4 Overlay of the peak resistance, the magnetoresistance at 10T, and the extrapolated normal state resistance versus temperature.

Fig. 5 Magnetoresistance as a function of field at low temperatures. The lines are fits to the model. The inset plots the temperature dependence of the intrinsic Josephson coupling energy.



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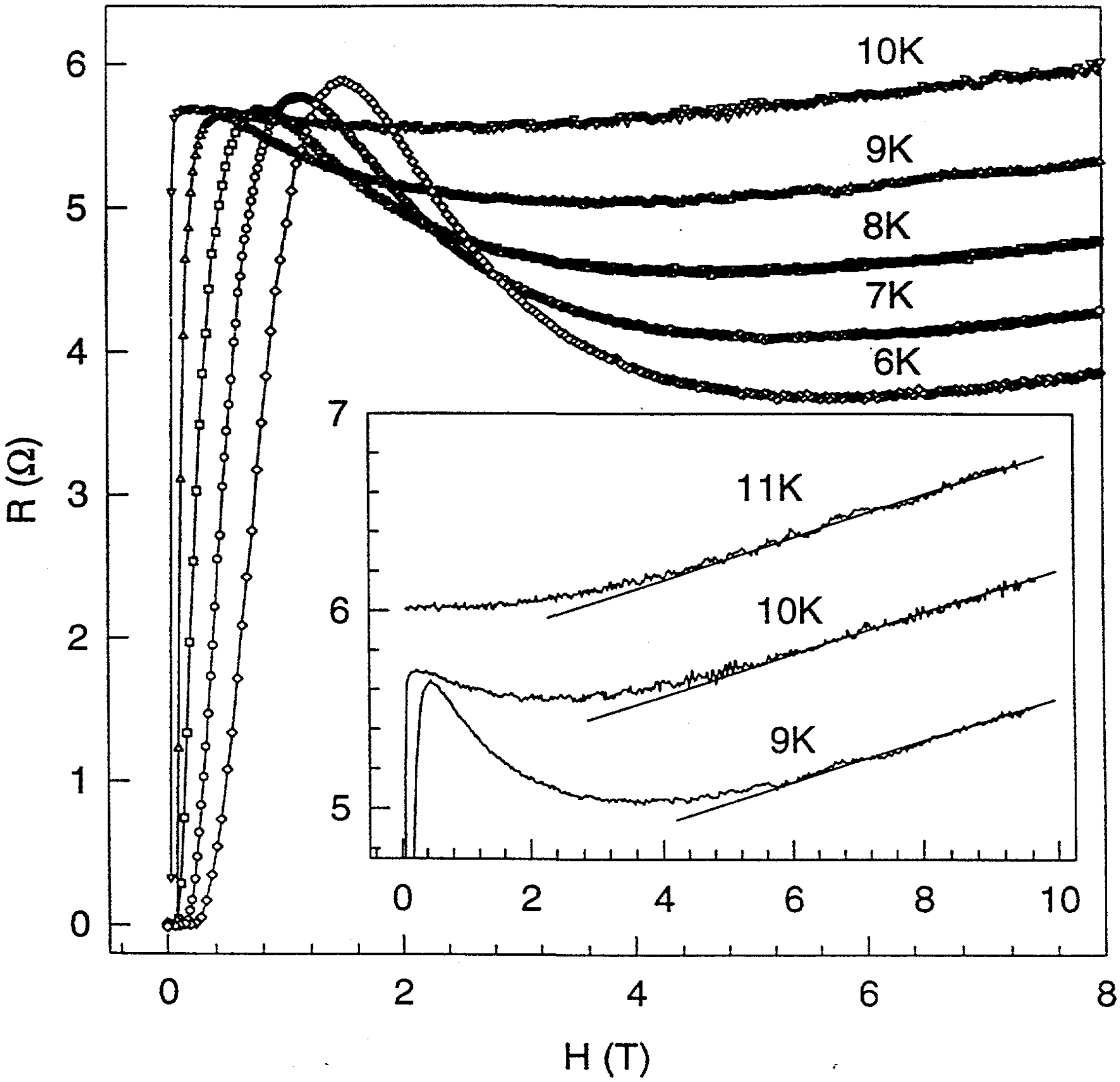

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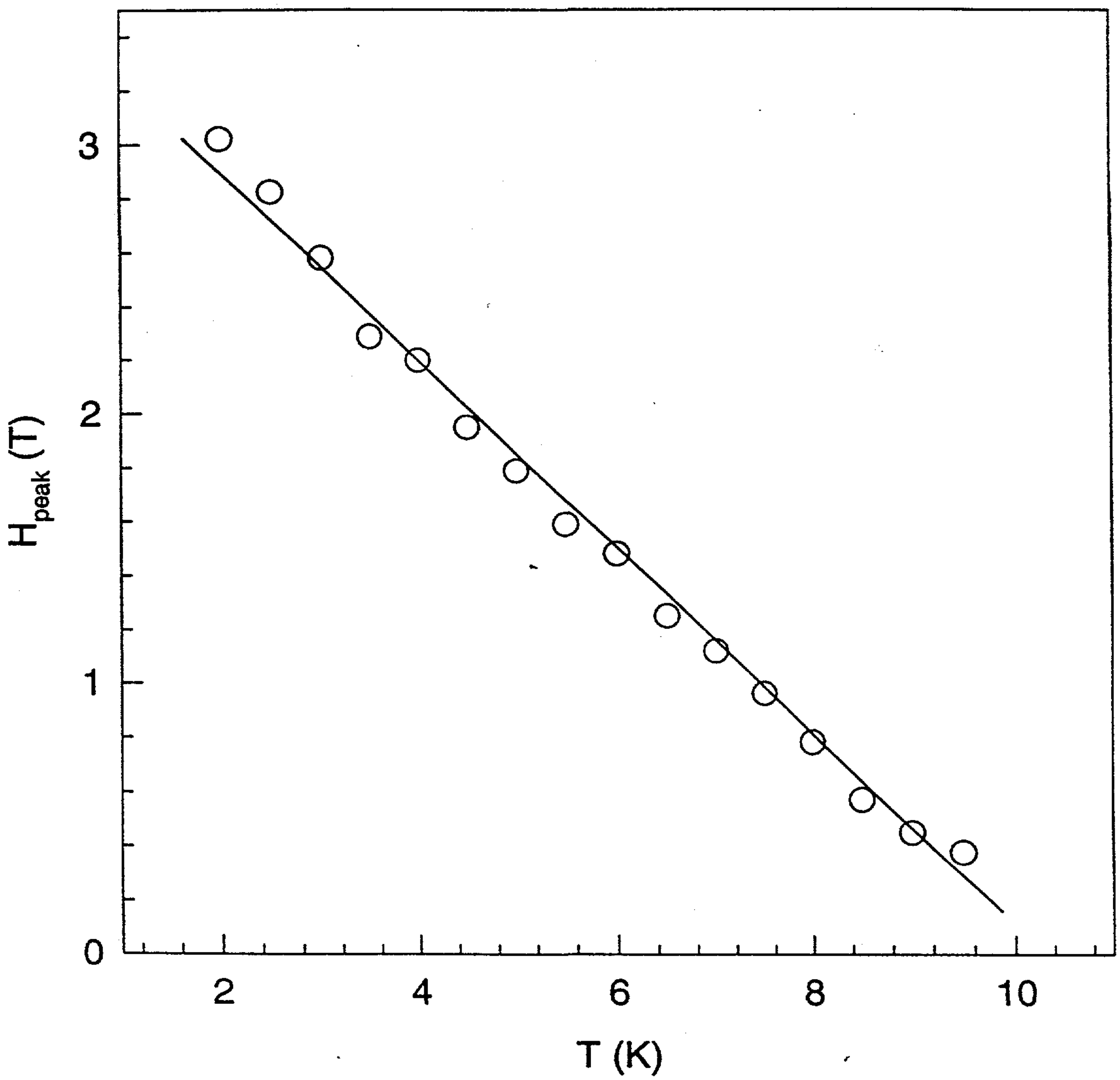

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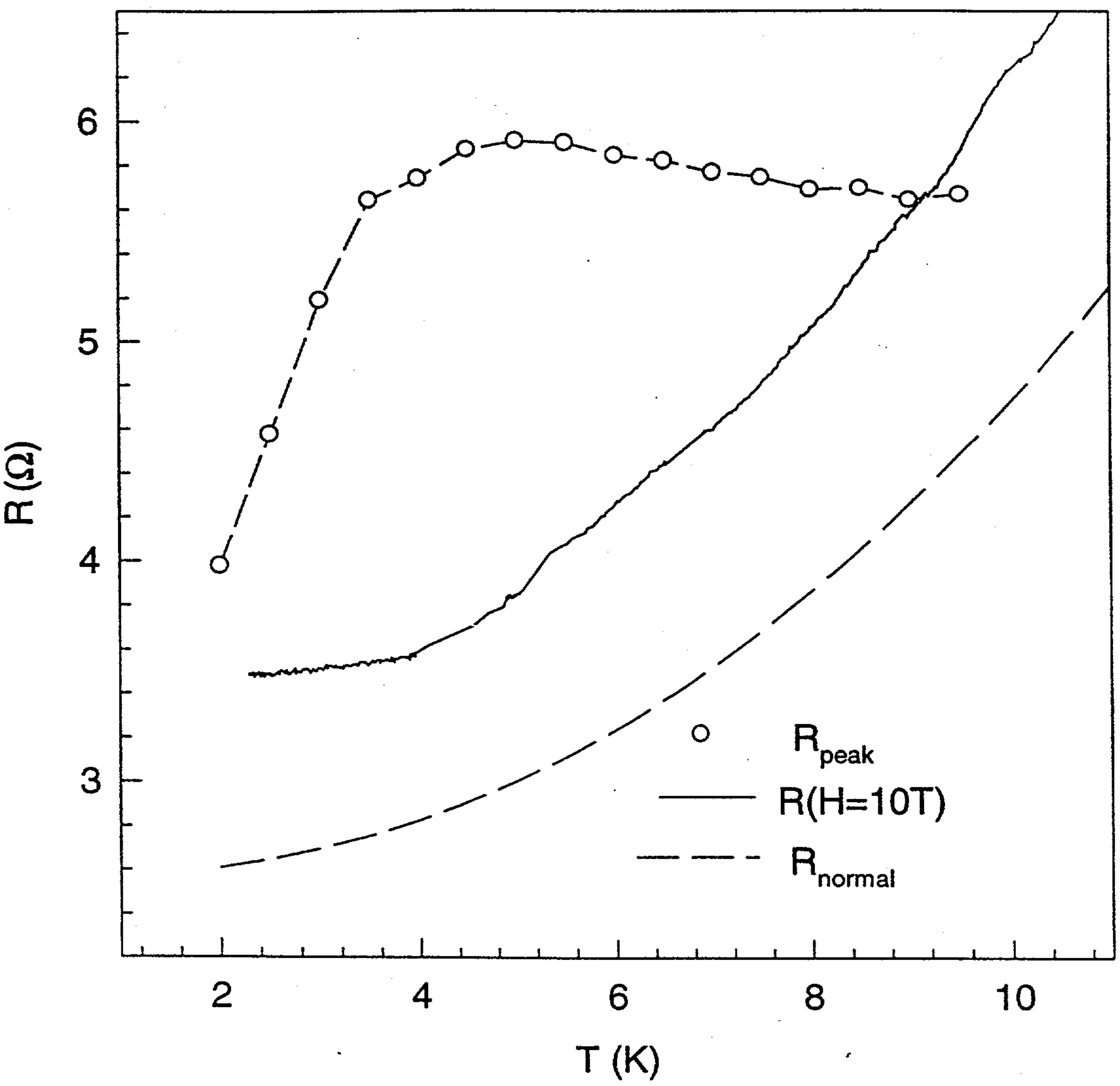

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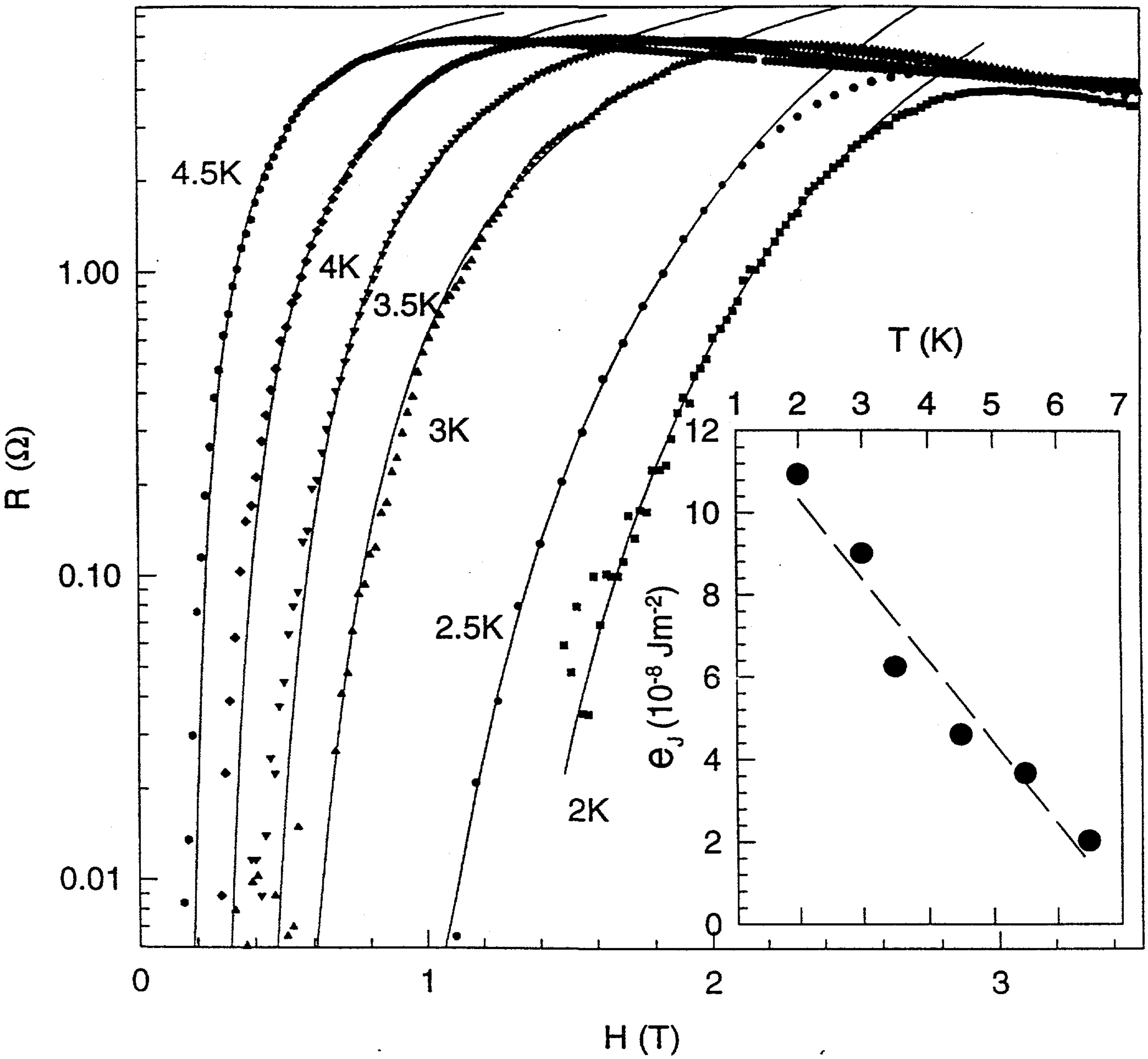

R (Ω)
H (T)
1.00
0.10
0.01
0
1
2
3
4.5K
4K
3.5K
3K
2.5K
2K
T (K)
1
2
3
4
5
6
7
12
10
8
6
4
2
0
e_J (10^-8 Jm^-2)

Zuo_fig5