PHYSICAL REVIEW B VOLUME 57, NUMBER 10 1 MARCH 1998-II

Effects of inhomogeneity on the interlayer magnetoresistance in the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

F. Zuo, X. Su, and P. Zhang
Department of Physics, University of Miami, Coral Gables, Florida 33124

J. A. Schlueter, M. E. Kelly, and Jack M. Williams
Chemistry and Materials Science Divisions, Argonne National Laboratory, Argonne, Illinois 60439

(Received 12 September 1997; revised manuscript received 24 November 1997)

In this paper, we compare the interlayer transport in the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br on samples of different inhomogeneities in terms of resistive transition width ΔT. For field parallel to the current, $H \| I$, in the direction perpendicular to the layers, the isothermal magnetoresistance $R(H)$ displays a peak effect as a function of field for high quality samples. The peak effect decreases gradually with increasing ΔT and it disappears completely for the sample with $\Delta T > 2$ K. The results demonstrate clearly that the peak in $R(H)$ in the interlayer direction is intrinsic to the layered structure in this compound. [S0163-1829(98)51010-7]

Transport in the direction perpendicular to the superconducting layers has been of recent interest. In the case of high temperature cuprates, such as $Bi_2Sr_2CaCu_2O_8$ and oxygen deficient $YBa_2Cu_3O_{7-\delta}$, charge transport in the c-axis direction is thermally activated.[1–4] Magnetoresistance in this direction shows a pronounced peak as a function of temperature. The results can be qualitatively interpreted in the framework of stacked Josephson junctions between the superconducting layers. In the case of layered organic superconductors, i.e., (BEDT-TTF)$_2X$ [bis(ethylenedithio)tetrathiafulvalene, abbreviated as ET], with X being Cu[N(CN)$_2$]Br$^-$. κ-(ET)$_2$Cu[N(CN)$_2$]Br, charge transport normal to the layers is metallic at temperature close to superconducting transition. Magnetoresistance with field parallel to the current displays a peak as a function of field and temperature in the mixed state.[5] To understand the origin of the peak it is critical to establish the experimental fact about the peak effect. Here we present transport studies in the direction normal to the superconducting layers as a function of resistive transition width.

Single crystals of the κ-(ET)$_2$Cu[N(CN)$_2$]Br superconductor were synthesized at the Argonne National Laboratory described elsewhere.[6] Several crystals were used in this study. Samples with a sharp transition show the same field and temperature dependence. In this work we compare the measurements performed on four different samples of various qualities. Results on two needle shaped samples, sample 1 with a sharp transition and sample 4 with a broad transition, are reported here. Samples 2 and 3 are actually different parts of one thick plate sample, whose transition is in between the two needle samples, five leads were mounted to measure simultaneously two segments of the same sample. The interlayer resistance was measured with use of the four probe technique. Contact of the gold wires to the sample was made with a Dupont conducting paste. Typical contact resistance between the gold wire and the sample was about 1–10 Ω. A current of 1–10 μA was used to ensure linear I-V characteristics. The room temperature interlayer conductivity is $(1.5 \pm 0.2) \times 10^{-2}$ s cm^{-1}, independent of samples within the experimental errors. Conductivity near T_c depends on the sample cooling rate, with a typical value around 1.0 ± 0.1 s cm^{-1}. The data reported here are for samples cooled slowly (over a period of 5 hours) to below T_c and the magnetic field and the current were applied parallel to the crystallographic b axis (perpendicular to the layers).

Figure 1 is an overlay of four normalized resistance curves (normalized at 12 K) as a function of temperature near the transition for four samples. Sample 1 has the sharpest transition with ΔT of about 0.5 K and an onset temperature of 11.5 K. Here the transition width ΔT is defined between the onset temperature and the zero resistance temperature. ΔT for samples 2 and 3 is larger than that of

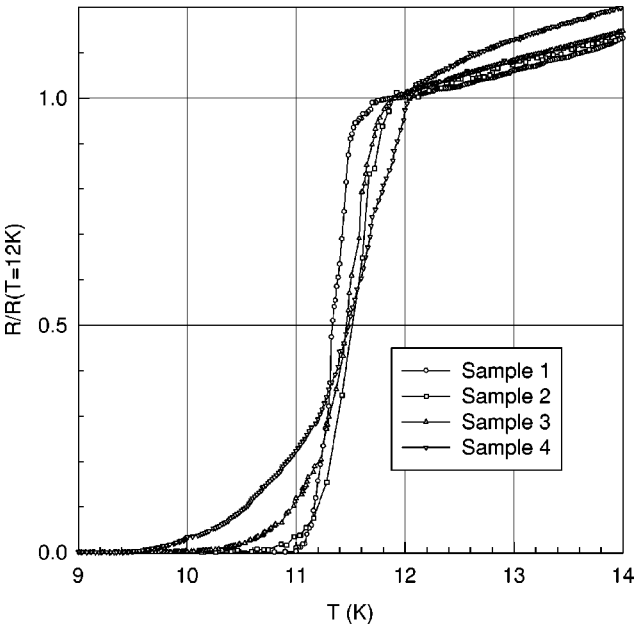

FIG. 1. An overlay of resistive transition as a function of temperature for four single crystal samples of κ-(ET)$_2$Cu[N(CN)$_2$]Br normalized at 12 K.

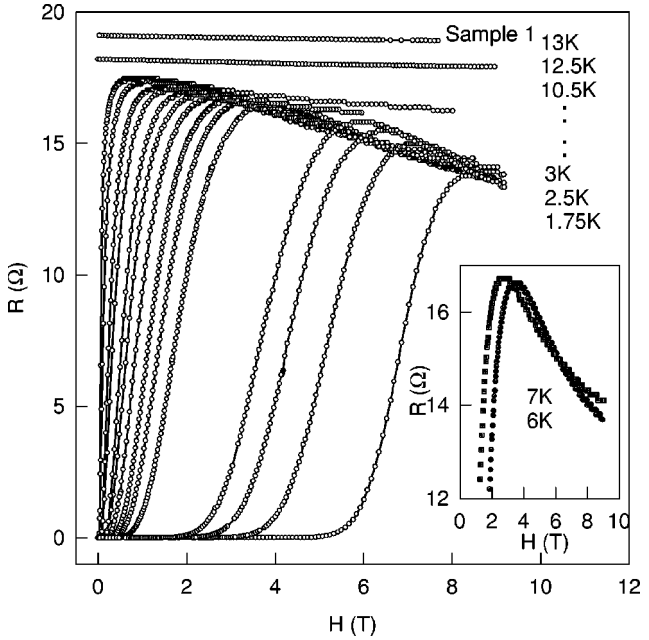

FIG. 2. Magnetoresistance as a function of field for sample 1 at different temperatures. The inset is an expanded view of $R(H)$ around the peak at 6 and 7 K.

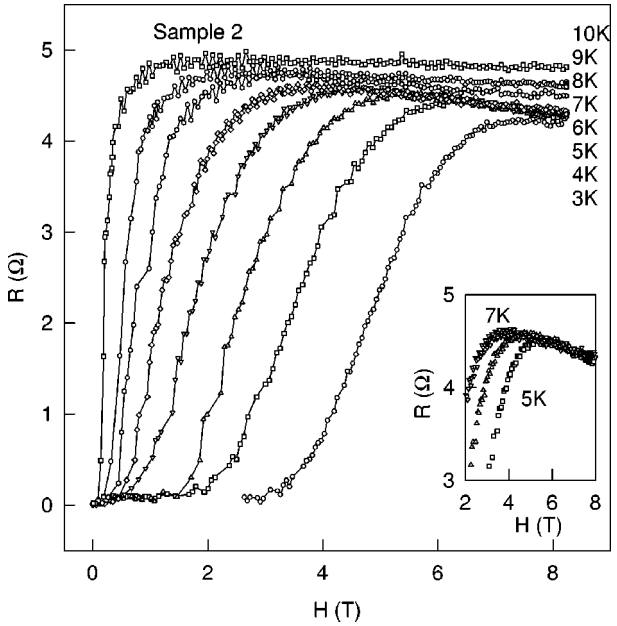

FIG. 3. Magnetoresistance as a function of field for sample 2 at different temperatures. The inset is an expanded view of $R(H)$ around the peak at 5, 6, and 7 K.

sample 1, with sample 3 displaying a broader transition than sample 2. Sample 4 has the largest ΔT of about 2.5 K and a higher onset T_c of 12 K. At temperatures above T_c, it is noted that the normalized resistance shifts upward sequentially for samples with increasing ΔT.

Figure 2 plots the isothermomagnetoresistance $R(H)$ as a function of applied magnetic field H at different temperatures for sample 1. The different curves correspond to temperatures at 13, 12.5, 10.5, 10, 9.5, 8.5, 7.5, 6.5, 6, 5.5, 3.5, 3, 2.5, and 1.75 K. At high temperatures above T_c (13 K, 12.5 K), a small but negative magnetoresistance, with $R(H)$ decreasing slightly with increasing H, is observed. At low temperatures (below T_c), the magnetoresistance displays a peak effect as a function of H. $R(H)$ is zero for field less than an onset field, above which $R(H)$ increases rapidly and reaches a maximum R_{peak} at a peak field H_{peak}. At higher field, $R(H)$ decreases with increasing H. The peak resistance decreases with lowering temperature and the peak field increases monotonically with decreasing temperature. For temperatures not far apart, the magnetoresistance $R(H,T)$ exhibits crossover at two fields (above H_{peak}).

The inset shows an expanded view of the peak effect at two intermediate temperatures $T=6$ K, and 7 K with peak field at 3.5 T and 2.7 T, respectively. If we define the relative change $\Delta R/R=[R_{\text{peak}}-R(8T)]/R_{\text{peak}}$ as a measure of the peak, $\Delta R/R$ is about 16% at $T=7$ K.

Figure 3 is an analogous plot as above for sample 2 with temperature ranging from 3 to 10 K at an increment of 1 K. The overall features are similar, but the peak effect is suppressed drastically. At temperatures near T_c, no peak is observed within the experimental accuracy. At intermediate temperatures where the peak effect is most pronounced in sample 1, a small but finite peak can be observed. At lower temperatures, the peak is not measured within the field range. The inset shows the enlarged view of the peak at $T=5$, 6, and 7 K, with the peak field shifted to larger values at 5.6, 4.8, and 4 T, respectively, and the maximum $\Delta R/R$ at 7 K is decreased to about 6%.

For sample 3, the peak is barely observed, as shown in Fig. 4. At 5 and 6 K (see the inset), two small peaks can be identified. The peak field moved up to 7.2 T and 6.4 T for 5 K and 6 K, respectively, and the relative change $\Delta R/R$ is about 1% at 5 K and zero at 7 K. The magnetoresistance near and above T_c increases with field.

For sample 4, as shown in Fig. 5, $R(H)$ displays a monotonic increase with rising field for all temperatures measured. At temperatures above T_c, $R(H)$ increases almost linearly

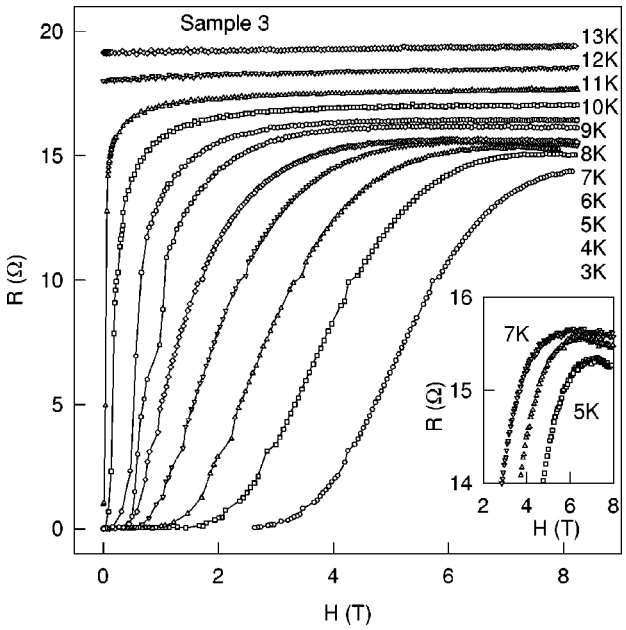

FIG. 4. Magnetoresistance as a function of field for sample 3 at different temperatures. The inset is an expanded view of $R(H)$ around the peak at 5, 6, and 7 K.

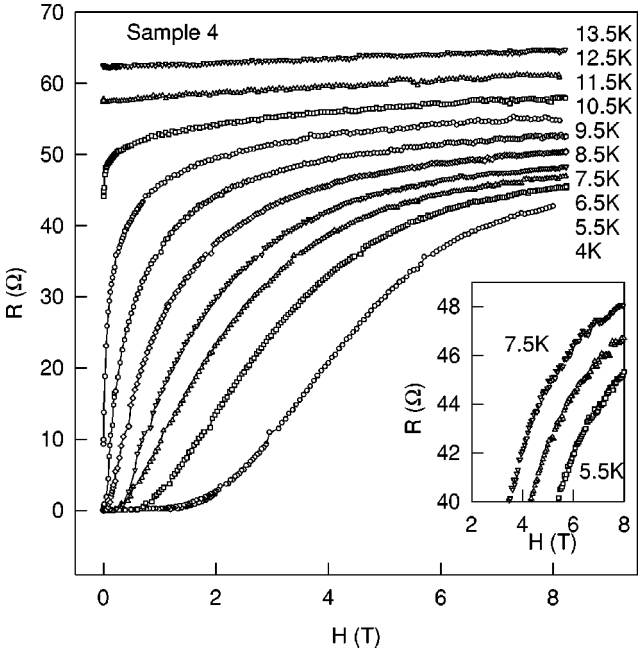

FIG. 5. Magnetoresistance as a function of field for sample 4 at different temperatures. The inset is an expanded view of $R(H)$ at 5.5, 6.5, and 7.5 K.

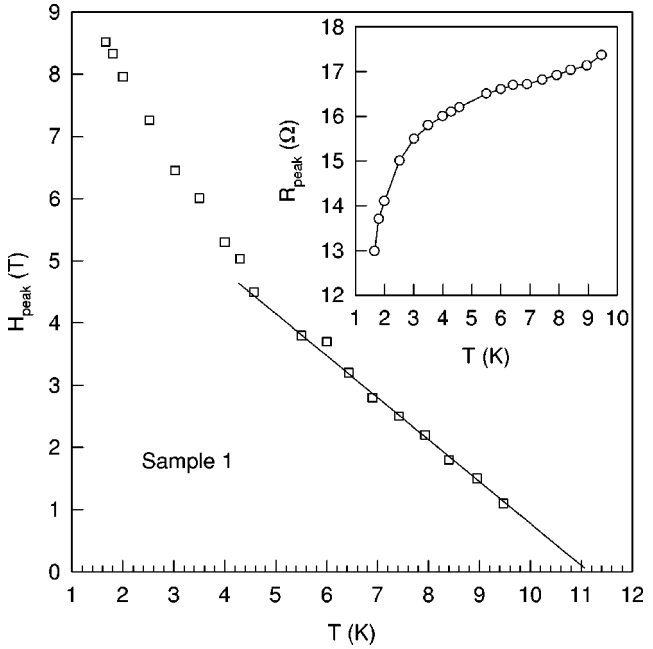

FIG. 6. The peak field as a function of temperature for sample 1. The inset plots the temperature dependence of peak resistance.

with H. At temperature near and far below T_c, $R(H)$ increases rapidly at small field and slowly at high fields. No peak is observed at any temperature or field measured. The inset shows an enlarged view of the data at $T=5.5$, 6.5, and 7.5 K. The increasing R at high H is in sharp contrast to data for the previous samples.

The origin of the finite resistive transition width has been discussed in detail in the case of cuprate superconductors. Generally the transition width is believed to be associated with the distribution of samples with different T_c's. In the case of cuprates, it was easy to realize because of the oxygen inhomogeneities involved. In the case of organic superconductors, it is not clear what is the origin for the broadening of the transition width. Unlike the cuprates, the room temperature conductivity does not show any correlation with the ΔT. However, we have noticed that the samples with broader transition typically have a lower metal-insulator transition temperature, from 60 K to 100 K. One speculation is that the broadening is caused by the disorder of the ethylene groups due to different cooling rates and stresses. Nevertheless, the systematic dependence of the peak effect of interlayer magnetoresistance demonstrates unequivocally that the peak is intrinsic only to the high quality samples. Samples with broad resistive transition has little or no peak at all in $R(H)$. The results are consistent with an earlier report that the interlayer resistance peak remained despite the fact the intralayer resistance peak disappeared with improved sample quality.[7]

The peak field as a function of temperature is plotted in Fig. 6. At high temperatures, H_{peak} is linear with T with a slope of $dH_{peak}/dT=0.7$ T K^{-1} and an intercept at $T=11.2$ K, coinciding with the zero resistance temperature. At lower temperatures ($T<5$ K), H_{peak} curves upward in a quailinear T dependence. The inset is a plot of the peak resistance as a function of temperature. The peak resistance decreases with T, with an inflection point at around 5 K.

Comparisons of $H_{peak}(T)$ with $H_{c2}(T)$ determined by magnetic measurements[8–11] suggest that the peak is in the mixed state. In a mean field approximation, an average slope of $dH_{c2}^{\perp}/dT=-2.2$ T K^{-1} was reported.[8] Scaling analysis including thermal fluctuations gives a temperature dependent $dH_{c2}^{\perp}/dT$ from -1 to -2 T K^{-1}.[10,11] The difference between H_{peak}(T) and $H_{c2}(T)$ increases at lower temperatures. For example, at $T=2$ K, $H_{c2}\simeq 14$ T was obtained.[11] This is considerably larger than $H_{peak}(2\text{ K})\simeq 8$ T. A more careful analysis is required to correlate the $R(H)$ data to $H_{c2}^{\perp}$. Nevertheless, it is clear that the peak effect is in the mixed state, as suggested in several recent studies on organic superconductors.[5,9]

Dissipation in the mixed state has been studied extensively for the high T_c cuprates. Two approaches are known to give rise to a peak in the interlayer resistivity as a function of temperatures. One of them models the resistivity peak as a result of fluctuations above the mean field transition temperature.[12–14] Of the four possible fluctuation contributions to the interlayer resistivity, fluctuations in the density of state (DOS) and the regular Maki-Thompson term contribute to an increasing resistivity with decreasing temperatures. By choosing suitable parameters, the model can fit reasonably the temperature dependence of the resistivity before the peak for the cuprates. However, the model does not include critical fluctuations nor contributions from the vortex state (for $T<T_{peak}$) and thus the field dependence of the peak temperature $T_{peak}(H)$.

Another widely adopted approach emphasizes the nature of Josephson coupling between the superconducting layers. In this model, current moving parallel to the c axis is taken to pass through a narrow superconducting channel of area $A\approx\Phi_0/H$ between the densely packed vortices.[1] Here Φ_0 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 at finite T.[15] The resistance of the weak link 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_0 is the modified Bessel function. Since the normal state resistance is activated in this direction as in the case of $Bi_2Sr_2CaCu_2O_8$, a peak is expected in the junction resistance at $T<T_c$.

A closely related approach models the c axis conduction as a stack of Josephson tunnel junctions.[2] 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 $Y_{ss}\sim\exp[-\Delta(H,T)/kT]$, and the pair conductances given by $Y_p\sim[I_0(\hbar I_c/2ekT)]^{-2}-1$, a peak in $R(H,T)$ can be expected. Both models can describe semiquantitatively the field and temperature dependence of the interlayer transport for the cuprates.

In the case of organic superconductors, the lack of a thermally activated transport above T_c suggests against the dominant roles by the models proposed above. Rather the extraordinary pressure dependence of the superconducting transition temperature and the structure softness of the organic superconductors κ-$(ET)_2X$ studied may be consistent with a recently proposed model, in which an assumed strong vortex interaction with the underlying crystal lattice would give rise to a peak in the magnetoresistance.[5] The vortex-lattice coupling introduces lattice distortions and thus, additional scattering mechanism for the charge carriers. The peak arises when vortices start to overlap with each other, and consequently reducing the effective distortion and resistance. A direct measurement of the lattice structure in the presence of field will be necessary to test this possibility.

In summary, we have reported a systematic study of the interlayer magnetoresistance as a function of the resistive transition width. The peak effect diminishes gradually for samples with increasing ΔT and vanishes completely when $\Delta T>2$ K. The results demonstrate that the peak effect is intrinsic only to the high quality samples in the direction perpendicular to the superconducting layers.

We acknowledge support in part by NSF Grant No. DMR-9623306. Work performed at Argonne National Laboratory was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences, under Contract No. W-31-109-ENG-38.

[1] G. Briceño, M. F. Crommie, and A. Zettl, Phys. Rev. Lett. **66**, 2164 (1991); Physica C **204**, 389 (1993).
[2] K. E. Gray and D. H. Kim, Phys. Rev. Lett. **70**, 1693 (1993).
[3] J. D. Hettinger *et al.*, Phys. Rev. Lett. **74**, 4726 (1994).
[4] A. S. Alexandrov *et al.*, Phys. Rev. Lett. **76**, 983 (1996).
[5] F. Zuo *et al.*, Phys. Rev. B **54**, 11 973 (1996).
[6] A. M. Kini *et al.*, Inorg. Chem. **29**, 2555 (1988).
[7] T. Ishigoro *et al.*, Physica B **201**, 470 (1994).
[8] W. K. Kwok *et al.*, Phys. Rev. B **42**, 8686 (1990).
[9] H. Ito *et al.*, Physica C **185-189**, 2659 (1991).
[10] M. Lang *et al.*, Phys. Rev. B **49**, 15 227 (1994).
[11] V. Vulcanescu *et al.*, Phys. Rev. B **53**, 2590 (1996); **52**, 471 (1995).
[12] L. B. Ioffe *et al.*, Phys. Rev. B **47**, 8936 (1993).
[13] V. V. Dorin *et al.*, Phys. Rev. B **48**, 12 951 (1993).
[14] A. S. Nygmatulin *et al.*, Phys. Rev. B **53**, 3557 (1996).
[15] V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. **22**, 1364 (1969).