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Magnetoresistance Peak in the Mixed State of the Organic Superconductor $\kappa-(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$

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Abstract

In this letter, we report transport measurements with field and current parallel to the *b* axis (perpendicular to the conducting plane) in the organic superconductor $\kappa-(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$. The isothermal magnetoresistance R(H) displays a peak effect as a function of field. The peak resistance is substantially larger than that in large fields. The results are in sharp contrast to the conventional dissipation mechanisms in the mixed state of anisotropic superconductors, as in the case of $Bi_2Sr_2CaCu_2O_8$. Comparison with $H_{c2}(T)$ obtained from magnetic measurements shows that the peak effect in R(H) occurs in the mixed state. Analysis of the data suggests a much larger Josephson junction resistance in the mixed state than that in the normal state, indicative of a new charge trasnport scattering mechanism in the presence of vortices.

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Charge transport in the direction perpendicular to the superconducting layers of the cuprates has been of recent interest [1,2]. Magnetoresistance in the c axis direction shows a pronounced peak as a function of temperature before it drops to zero [3–7]. This was first observed in the very anisotropic $Bi_2Sr_2CaCu_2O_8$ superconductors [3]. Similar results were reported in the oxygen deficient $YBa_2Cu_3O_{7-\delta}$ materials [6]. The results can be qualitatively interpreted in the framework of stacked Josephson junctions between the superconducting layers. To test this model, we have performed magnetoresistance measurements in the organic charge transfer salts, $(BEDT\text{-}TTF)_2X$ [bis(ethylenedithio)tetrathiafulvalene, abbreviated as ET] superconductors, with X being $Cu[N(CN)_2]Br^-$. $\kappa-(ET)_2Cu[N(CN)_2]Br$ undergoes a superconducting transition with the T_c near 12K at ambient pressure [8]. The material has 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 (ac plane) [9,10]. Unlike most of the high temperature oxide cuprates, the organic $\kappa-(ET)_2X$ family is extremely sensitive to the applied pressure [11]. For $\kappa-(ET)_2Cu[N(CN)_2]Br$, it has been reported that T_c decreases strongly with the applied pressure, with $dT_c/dP = -2.8$K/kbar initially. It is second to the largest reported for $\kappa-(ET)_2Cu(SCN)_2$ ($dT_c/dP = -3$K/kbar). The large dT_c/dP value suggests that lattice distortions will play an important role in the charge transport of these systems.

In this letter, we report magnetoresistance measurements of $\kappa-(ET)_2Cu[N(CN)_2]Br$ with field and current parallel to the b axis (perpendicular to the conducting ac plane). Measurements have been performed at fixed temperatures (field sweep) and fixed fields (temperature sweep). Our results show a pronounced peak in the magnetoresistance as function of field at fixed temperatures, with the peak field increasing with decreasing temperatures. The peak resistance is substantially larger than the magnetoresistance at saturation fields. A comparison with the $H_{c2}(T)$ data obtained magnetically suggests that the peaks are clearly in the mixed state. Analysis of the data in terms of Josephson junction model gives an anomalously large junction resistances. This result is in sharp contrast to that of anisotropic cuprate superconductors. We propose that the peak in the magnetoresistance is caused by an additional scattering mechanism in the mixed state, possibly due to a strong coupling to the underlying crystal lattice of fluctuating vortices.

Single crystals of the κ-$(ET)_2Cu[N(CN)_2]Br$ superconductor were synthesized by the electrocrystallization technique described elsewhere [8]. Several crystals were used in these measurements with average dimensions of $1\times0.78\times0.33$mm. Extensive measurements were made on one crystal with T_c= 12K. Measurements were performed at the National High Magnet Field Laboratory with field up to 18T. The interlayer resistance was measured with use of 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 linear I-V characteristics. 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 b axis.

Shown in figure 1 is an overlay of the isothermal magnetoresistance versus applied magnetic field H at low temperatures of T=2K, 2.5K, 3K, 3.5K, 4K, 4.5K, 5K, 5.5K, 6K, and 6.5K. For each fixed temperature, the magnetoresistance R(H) increases rapidly for H greater than an onset field. R(H) reaches a peak at H_{peak}. For H> H_{peak}, R(H) decreases with increasing field and saturates at very high fields. For example, at T=2K, R reaches a maximum of about 2.2 Ω at $\simeq$ 9T and it is saturated for H> 14T. At higher temperatures, H_{peak} decreases monotonically with increasing T and R_{peak} increases slowly to about 2.6 Ω at 6.5K. The peak becomes broader at higher temperatures with R(H) at different T crossing each other. The inset shows an expanded view of the same data around the peak. Clearly, the change of resistance dR/dH is much larger for H< H_{peak} than the rate of decrease for H> H_{peak}.

For temperature above 7K, qualitatively similar results are observed as shown in figure 2 at temperatures T= 6K, 6.5K, 7K, 7.5K, 8K, 8.5K, 9K, 9.5K, 10K, and 11K. The peak resistance R_{peak} increases again with increasing T with a corresponding decrease in H_{peak}. At T=11K, R(H) is saturated at H=4T, as shown in the inset.

Figure 3 is a plot of the peak resistance, magnetoresistance at H=10T, and the resistance at H=0 as a function of temperature. Clearly, R(H=10T) overlaps well with the R(H=0) data at high temperatures (T> 13K), consistent with the low magnetoresistance in this material. R_{peak} is substantially larger than R(H=10T) with a maximum separation occurring around 4K. The closing of the two resistances at low T is due to the fact that H=10T is close to the peak field at these temperatures. At higher T, the gap closes again due to the weak field dependence of R(H) at high fields. The inset is a plot of H_{peak} as a function of temperature. The line is a linear fit with $dH_{peak}/dT = 0.75 \pm 0.05\ TK^{-1}$. Deviation from the line is clear at lower temperatures. If we assume a simple form for the peak field $H_{peak}=H_o(1-T/T_c)$, we find H_o to be 9.0±0.5T and the T_c extrapolated is about 12K. This coincides with the temperature when R reaches zero at H=0.

Figure 4 displays the low temperature magnetoresistance in the semilog scale. The solid lines, to be discussed, are fits to the data for $H< H_{peak}$. It is clear that the lines fit the data over three decades in R(H).

To understand the field and temperature dependence of the magnetoresistance in the κ-$(ET)_2Cu[N(CN)_2]Br$ system, we compare our results to those of highly anisotropic cuprate superconductors, namely $Bi_2Sr_2CaCu_2O_8$ and the oxygen deficient $YBa_2Cu_3O_{7-\delta}$. In the case of $Bi_2Sr_2CaCu_2O_8$, published work shows mostly magnetoresistance as a function of temperature at fixed fields [3–5]. The peak temperature decreases with increasing field. The magnetoresistance on the higher temperature side of the peak approaches a normal state value, without crossing each other. On the lower temperature side of the peak, R(H,T) increases with increasing H and T, again without crossing. The $Bi_2Sr_2CaCu_2O_8$ data is very similar to the results obtained from oxygen deficient $YBa_2Cu_3O_{7-\delta}$, where explicit field dependence at fixed T has been reported [6]. Another important feature of the cuprates is that the peak resistance as well as the normal state resistance (in the overlapping region), increases monotonically with decreasing temperature. In sharp contrast to these results, the organic superconductor studied here exhibits a crossing in R(H,T) and the normal state resistance is metallic rather than semiconducting. R_{peak} is larger than the magnetoresistance at H=10T. In fact, a pronounced peak in $R_{peak}(T)$ has been recently observed [12] in another organic superconductor κ-$(ET)_2Cu(SCN)_2$.

Comparisons of $H_{peak}(T)$ with $H_{c2}(T)$ determined by magnetic measurements [13–16] suggest that the peak is indeed in the mixed state. In a mean field approximation, an average slope of $dH_{c2}^{\perp}/dT= -2.2TK^{-1}$ was reported [13]. Scaling analysis including thermal fluctuations gives a temperature dependent $dH_{c2}^{\perp}/dT$ from -1 to $2TK^{-1}$ [15,16]. The difference between $H_{peak}(T)$ and $H_{c2}(T)$ increases at lower temepratures. For exmaple, at T=2K, $H_{c2} \simeq 14$ T was obtained [16]. This is considerablly larger than $H_{peak}(2K) \simeq 9T$. The 14T is rather close a field where a change of slope, dR/dH, becomes visible, prior to the saturation. 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 where quantized vortices exist! The $H_{peak}(T)$ is not directly associated with $H_{c2}^{\perp}(T)$, as initially suggested in κ-$(ET)_2Cu(SCN)_2$ [17,18].

Dissipation mechanisms in the mixed state have been studied extensively for the high T_c cuprates. However, to our knowledge, there is no theoretical model predicting above normal state resistance in the mixed state. In fact, our results are in sharp contrast to the conventional dissipation mechanisms. In the case of flux flow, which is not the case here, the effective resistance is given by the Bardeen-Stephen model [19], $\rho_f = \rho_n \frac{H}{H_{c2}}$ where ρ_f is the flux-flow resistivity and ρ_n is the normal state resistance in the core region. ρ_n is reached only when $H=H_{c2}$. Dissipation in the geometry $H \parallel I \parallel c$ for the cuprates has been discussed in several recent papers [3–7]. In a model proposed by Briceño *et al* [3], current moving parallel to the c axis is taken to pass through a narrow superconducting channel of area $A \approx \frac{\Phi_o}{H}$ between the densely packed vortices. Here 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 at finite T [20]. 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_o is the modified Bessel function. Since the normal state resistance is activated in this direction, a peak is expected in the junction resistance at T< T_c. An alternative approach which models the c axis conduction as a series of stack of Josephson tunnel junctions has been proposed by Gray *et al* [4]. 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 in R(H,T) arises naturally.

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. If we assume the normal R_n(H) at T< T_c is similar to that at T> T_c, where negligible magnetoresistance has been reported [13,14]. The saturated R_n(H) at high field can be used as a guide of the normal state value for T< T_c. The large peak effect thus implies a greater resistance in the mixed state than that in the normal state. This is inconceivable in terms of a Josephson junction model with a "normal" state junction. An additional scattering mechanism is needed for this extra resistance!

A careful comparison bewteen the organic superconductor and the cuprates shows a very similar field dependence of R(H) for H< H_{peak} [6], which points to a common physical origin in this field range. To provide a quantitative picture, we analyze the data in terms of a stacked Josephson junction model with a new junction resistance in mind [3,6]. Charge transport is considered to be 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 mixed state. Because I_c is proportional to the junction area, E_J is also. It is natural to define an 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 [6]

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

Data at several different temperatures are fitted to eq.1, as shown by the solid lines in figure 4. Clearly, the model fits the R(H) data quite well over a broad resistance range. Deviations near the H_{peak} are expected, as discussed below. The junction resistances, thus fitted, are shown in the inset. The intrinsic Josephson coupling energy increases with decreasing T except at T=2K, with e_J =2.0, 2.9, 2.5, 1.8, 0.9 $\times 10^{-7}$JM^{-2} for T = 2, 3.5, 4.5, 5.5, 6.5K, respectively. We noted a large negative H_o needed for the best fit at low temperatures. For example, $H_o \sim -3$T is obtained at T=2K, while it is almost constant ~ -0.1T for T> 4K.

The large R_{peak} and $R_{n'}$ indicate a new scattering mechanism in the charge transport in the mixed state. The peak effect in R(H) suggests its disappearance at large flux densities. The fact that the organic superconductor is extremely sensitive to the applied pressure suggests a possible effect due to a coupling of the vortex to the underlying crystal lattice. This can lead to a significant local distortion in the lattice structure seen by the charge carriers. 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). In this picture, the peak in the isothermal magnetoresistance can be easily understood. At small vortex densities, the distortion due to each vortex is independent, resulting in a junction resistance given by eq.1. At higher vortex density, when ℓ is comparable to intervortex separation, 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 to the normal state property. The temperature dependence of the peak field can be qualitatively understood if we assume $\ell(T) \sim a = 1.075\sqrt{\frac{\Phi_o}{H}}$ 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 at high temperatures $H_{peak} \sim (1-\frac{T}{T_c})$, as shown by the solid line in figure 3.

For an order of magnitude estimate, we check the length scales involved in this simple model. For example, at T=6K, H_{peak} is about 5T, corresponding to an intervortex separation $a = 218Å$. With use of results [13,14] from magnetic measurements $\zeta_o = 37Å$, the coherence length at this temperature can be estimated to be $\frac{\zeta_o}{\sqrt{1-T/T_c}} \sim 52Å$. The distortion length $\ell(T) \sim 100Å$ is thus comparable to the intervortex separation, consistent with our model.

In summary, we have reported a peak effect in the isothermal magnetoresistance as a function of field on an anisotropic organic superconductor $\kappa-(ET)_2Cu[N(CN)_2]Br$. This result is in sharp contrast with that observed on highly anisotropic cuprate superconductors. The peak resistance is substantially larger than that at high field, consistent with the large junction resistance obtained by modeling the data to stacks of Josephson junctions. Comparison with $H_{c2}(T)$ determined magnetically shows unambiguously that the peak effect occurs in the mixed state. We propose that the large magnetoresistance is due to a lattice distortion via coupling with the quantized vortices. The distortions give rise to a new scattering mechanism in the 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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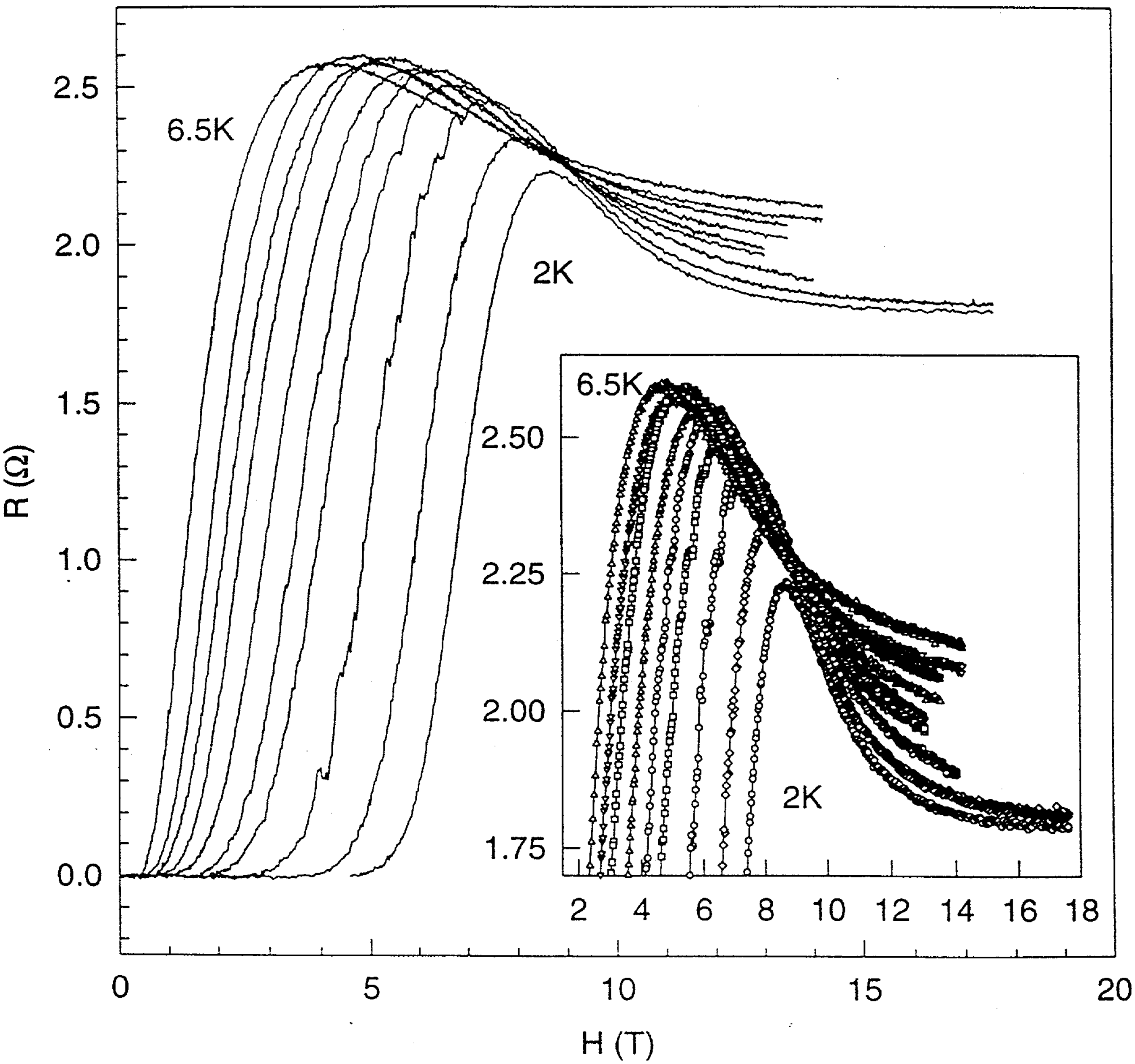
Figure Captions.

Fig. 1 Magnetoresistance as a function of field at low temperatures (T=2−6.5K in 0.5K increments). The inset is an expanded view of R(H) around the peak.

Fig. 2 Magnetoresistance as a function of field at high temperatures(T=7−11K in 0.5K increments). The inset is an expanded view around the peak.

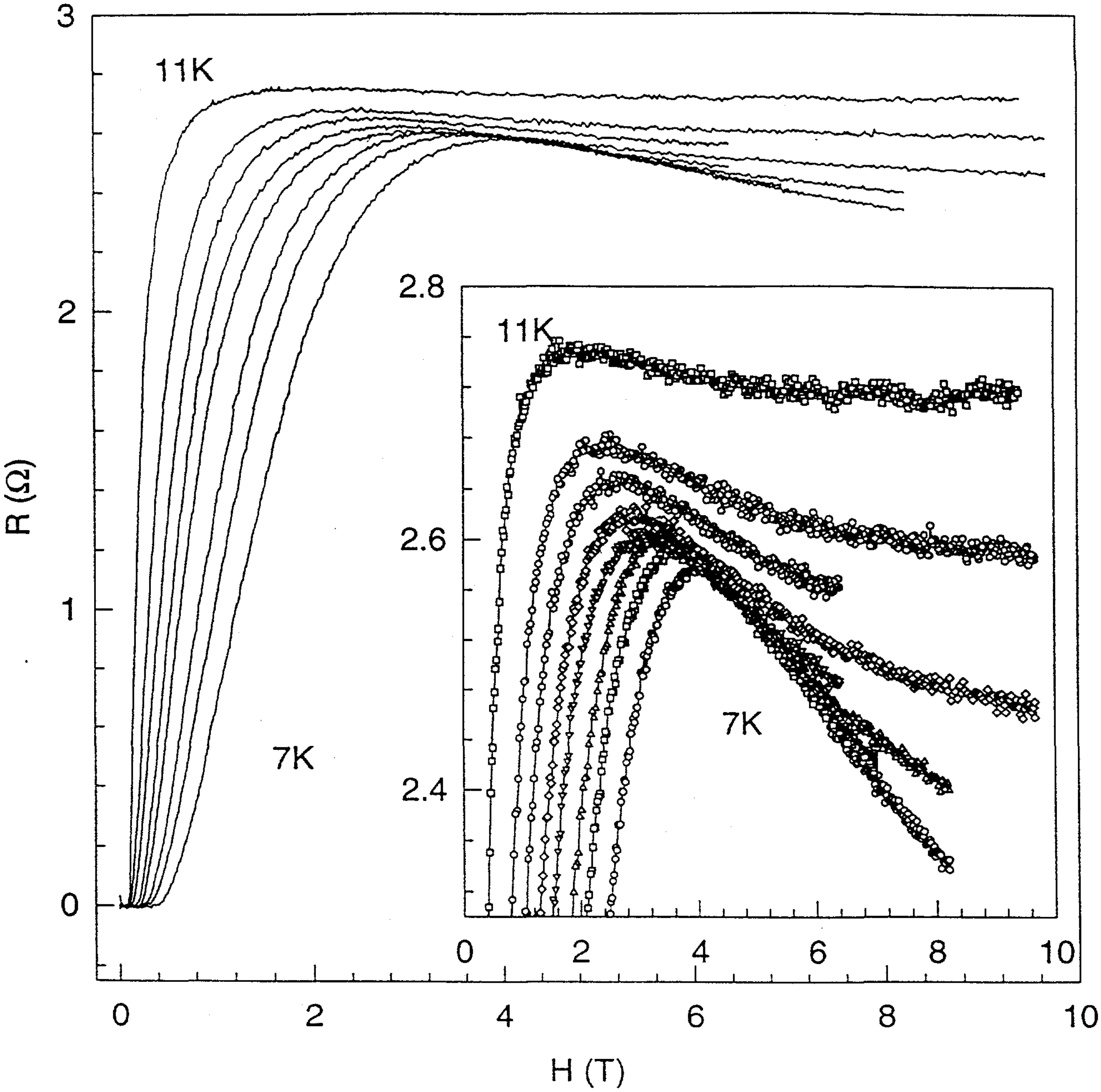
Fig. 3 Overlay of the peak resistance, the magnetoresistance at 10T, and the H=0 resistance versus temperature. The inset plots the peak field versus temperature.

Fig. 4 Magnetoresistance as a function of field at low temperatures. The lines are fits to the model. The inset plots the temperature dependence of $R_{n'}$.



2.5
2.0
1.5
1.0
0.5
0.0
R (Ω)
6.5K
2K
0
5
10
15
20
H (T)
6.5K
2.50
2.25
2.00
1.75
2K
2
4
6
8
10
12
14
16
18

Rh_Br_fig1



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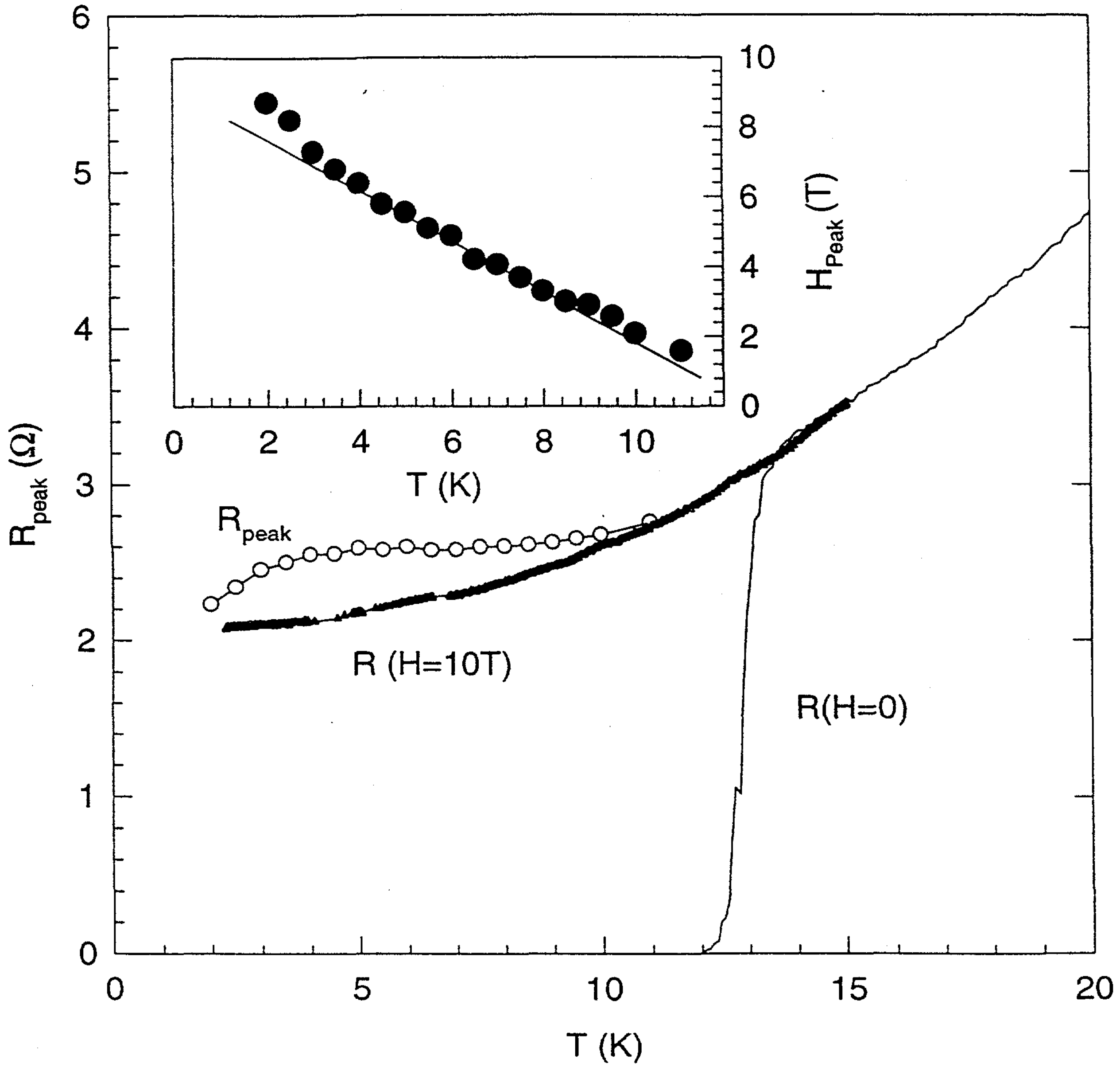
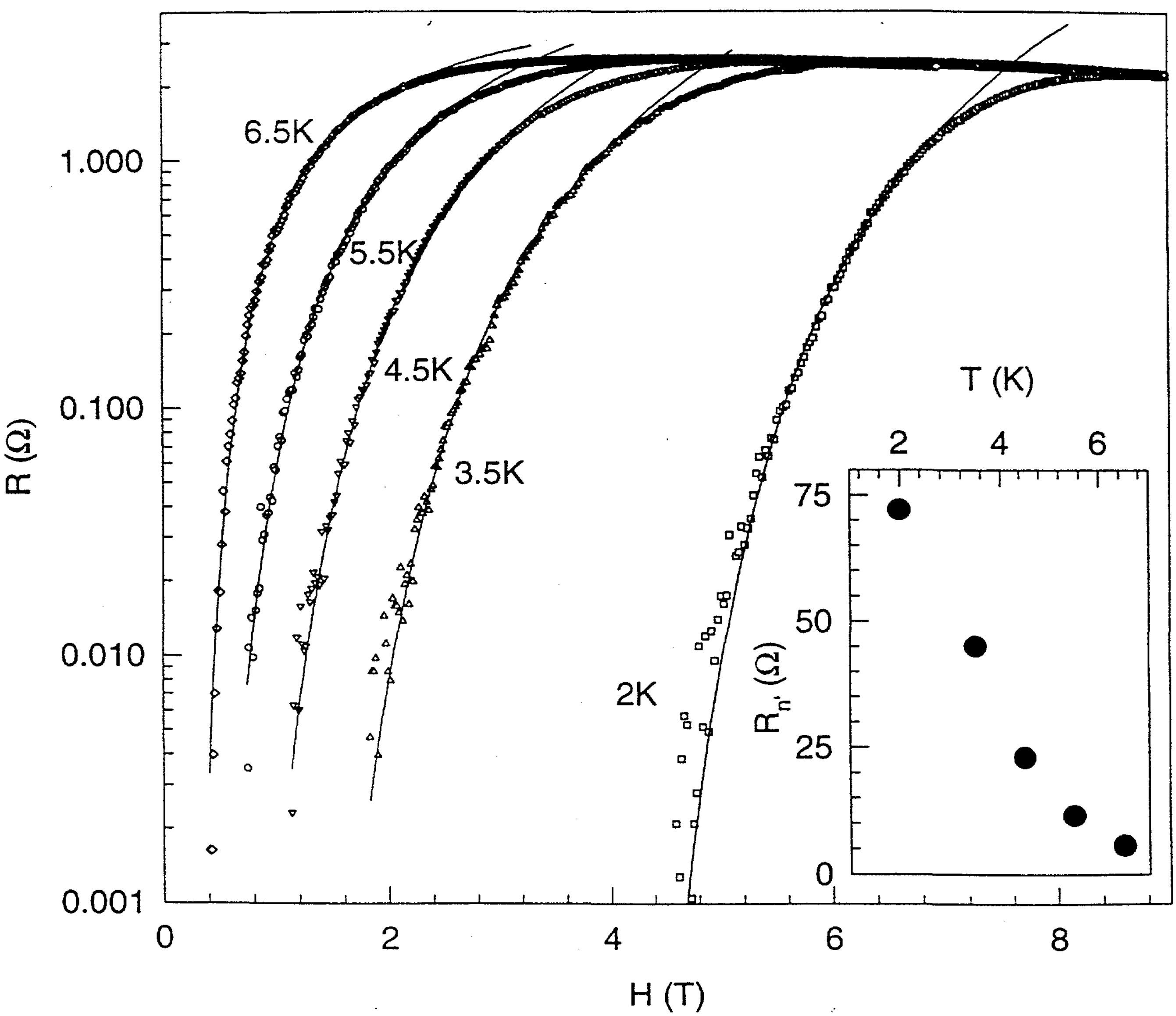


0
1
2
3
4
5
6
R_peak (Ω)
0
5
10
15
20
T (K)
R_peak
R (H=10T)
R(H=0)
0
2
4
6
8
10
T (K)
H_Peak (T)

Rh_Br_fig3



6.5K
5.5K
4.5K
3.5K
2K
1.000
0.100
0.010
0.001
R (Ω)
0
2
4
6
8
H (T)
T (K)
2
4
6
75
50
25
0
R_n' (Ω)

Rh_Br_fig4