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Critical Current Density and Vortex Pinning Strength in the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br Organic Superconductor

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ABSTRACT: *This paper systematically studied the critical current density and pinning strength as functions of the cooling rate through the order-disorder transformation near 80 K for both the hydrogenated (κ-H_8-Br) and deuterated (κ-D_8-Br) κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$] Br organic superconductors. By analysing the magnetic hysteresis loops using Bean's model, we estimated the critical current density (J_C). Our results have shown that cooling the sample at different rates through the order-disorder transformation near 80 K had a dramatic effect on critical current density, the superconducting transition temperature (T_C) and the pinning strength (F_P). The effect was pronounced for the deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br compound where C_2H_4 hydrogen molecules were replaced by the deuterium.*

Keywords: Hysteresis, critical current density, cooling rate, magnetisation, pinning strength

1. INTRODUCTION

An organic superconductor, especially the κ-(BEDT-TTF)$_2$ X (with X = Cu[N(CN)$_2$] Br or Cu[N(CN)$_2$]Cl), is a quasi-two-dimensional organic superconductor called Bechgaard salt, where X is an inorganic monovalent anion and BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene. The κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$] Br has shown physical states similar to high-temperature cuprates, including unconventional metallic properties.[1] The materials on the molecule BEDT-TTF are more two-dimensional than those based on tetramethyltetraselenafulvalene (TMTSF) or tetramethyltetrathiafulvalene (TMTTF). This is especially true for materials that crystallise according to the arrangement of κ. Organic superconductors are characterised by a structure consisting of a pile of planes in the direction c (metallic sheets: dimer molecules of BEDT-TTF) with insulating chains (anion X) alternately inserted between the planes.[2] At a higher temperature, an ethylene molecule such as BEDT-TTF oscillates rapidly between two different conformations. The κ-(ET)$_2$Cu[N(CN)$_2$]Br exhibits other typical features, such as a structural transformation occurring around 80 K, introducing a certain degree of disorder in the conducting planes. This can considerably influence the physics of vortex lattices of these compounds and their associated magnetic behaviour.

We have reported on systematic investigations of the relationship between superconducting properties and the structural transition occurring around 80 K in fully deuterated organic salt κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. The transition temperature is strongly dependent on the cooling rate. Upon cooling, these thermal fluctuations gradually slow; simultaneously, a long-range order builds up among the ethylene groups.[3,4] They undergo a structural transition (also called a “glassy transition”) involving the ethylene groups in the vicinity of 80 K.[5–8] The effect is especially pronounced for deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, which is presumably at the boundary between the magnetic and superconducting phases, with both superconducting and transitions magnetic.[9,10] This type of material exhibits interesting magnetic and superconducting phase transitions.

We systematically studied the DC magnetisation, the critical current density and the pinning strength as functions of the temperature and under the cooling rate through the order–disorder transformation near 80 K for the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$] Br organic superconductor. We determined the critical current density (J_C) and the pinning strength for the hydrogenated (κ-H_8-Br) and deuterated (κ-D_8-Br) compounds using experimental measurements of the hysteresis cycles. However, to the best of our knowledge and despite the number of investigations devoted to organic superconductors, we found no pinning strength studies. That is how we became interested in this study.

2. EXPERIMENTAL

The two samples studied in this work are single crystals of an organic superconductor of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br that were synthesised at the Jean Rouxel Institute of Materials, Nantes, France. Our samples had good magnetic quality and were stable; the results remained unchanged despite repeating the experiment several times. Our study focused on the magnetic characterisation of compounds and not on their preparation. The dimensions are $1 \times 1 \times 0.25$ mm^3 and $0.7 \times 0.7 \times 0.2$ mm^3 for deuterated and hydrogenated samples, respectively, with the superconducting transition temperature near 12 K. The samples were cooled at different rates to below the superconducting transition temperature with the applied magnetic field perpendicular to the superconducting layers. The magnetic measurements were done with a quantum design Superconducting Quantum Interference Device (SQUID). The magnetic hysteresis cycles M(H) ($0 < H < 10000$ Oe) were typically measured between 2 K and 15 K.

Before the start of each measurement, the sample was warmed to a specific temperature far beyond (transition temperature) T_C. Once the residual field was eliminated, the sample was zero-field-cooled (ZFC) or field-cooled (FC) to the desired temperature.

3. RESULTS AND DISCUSSION

We investigated the DC magnetisation of the superconducting phase as a function of the cooling rate in the ZFC conditions. The sample cooling process was as follows: in the slow cooling rate, we cooled the sample from 160 K to 90 K at a rate of about 2 K min^{-1}, and from 90 K to 70 K with a cooling rate of 0.1 K min^{-1}, then kept it at this temperature for 20 h. The sample was directly cooled to 2 K at a cooling rate of 5 K min^{-1}. The average rate was 10 K min^{-1}. In rapid cooling conditions, the sample was immersed directly in the dewar at 2 K. The superconducting transition of the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br was measured using DC magnetisation between 2 K and 13 K in both the slow and rapid cooling conditions. Figure 1 shows the magnetisation curves, or M(T), measured in various applied magnetic field values perpendicular to the superconducting layers. We used the hysteresis loops to study reversible and irreversible magnetisation.

We observed a strong decrease of superconducting T_C with increasing cooling rate (Figure 1). The transition T_C began at 11.88 K and 10.50 K for slow and rapid cooling, respectively. The origin of the bump observed near T_C is not clear but could be due to some interplay between antiferromagnetism and superconductivity. Both curves showed that the cooling rate had a great effect on the superconducting transition.

For rapid cooling, the magnetisation showed a linear behaviour, unlike the slow cooling where the superconducting T_C remained essentially unchanged during the increase of the field. The slope (dM/dT) varied depending on the cooling rate and the values of the applied magnetic field. The rapid cooling increased the magnetic phase and suppressed the superconductivity phase. However, a much higher sensitivity to cooling rate was obtained; that is, with the absence of a diamagnetic saturation in the rapid cooling, the deuterated system was situated in the critical region of the transition between a superconducting phase and an antiferromagnetic phase.[9] A particularly interesting feature is the existence of the crossing point of magnetisation M*(T*) curves at which the magnetisation is independent on the field. As with the critical T_C, the temperature T* depended strongly on the cooling rate of the sample. As seen in Figure 1, the values of T* changed from 11.75 K for the slow cooling to 10 K for the rapid cooling. The magnetisation M*(T*) of the crossing point was $M^* = -3.\ 89 \times 10^{-6}$ emu at T* = 11.75 K for slow cooling and $M^* = -2.92 \times 10^{-6}$ emu at T* = 10 K for rapid cooling. The rapid cooling introduced disorder and reduced the flux pinning, thereby diminishing the volume fraction of the superconducting phase. The reduction in T* may arise from the spin scattering of the canted moment in the antiferromagnetic phase.[11] Bulaevskii et al. considered that changes of the magnetisation are due to the contribution of thermal distortions to the free energy and showed the vortex-lattice state at $H \ll H_{C2}$ get.[12] Koshelev noted that the formula proposed by Bulaevskii was a good approximation of the weak fluctuation contribution.[13] Using the last expression where k_B and Φ_0 are the Boltzmann constant and flux quantum, respectively, and S is the effective interlayer distance, our results, Table 1 shows values of the effective interlayer distance S that are close to values found in others' research:[14]

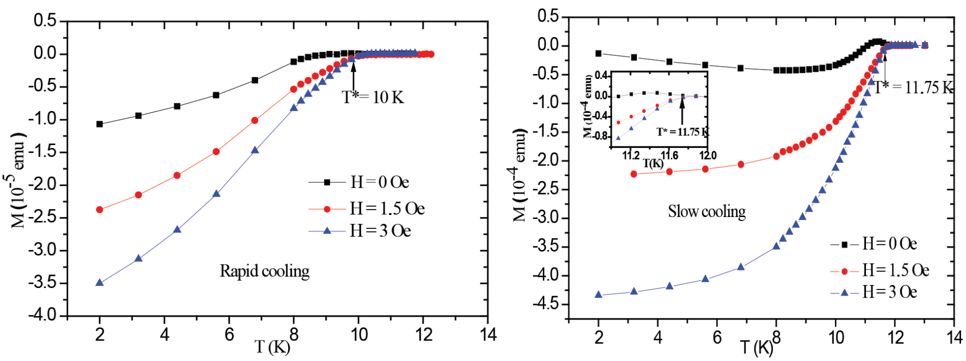

Figure 1: Temperature dependence of the magnetisation of the deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br crystal after slow and rapid cooling for divers applied magnetic fields. The inset shows the crossing point. Solid lines are guides for the eye.

Table 1: Effective interlayer distance "S" of the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br for slow and rapid cooling.

	−M*(emu)	T* (K)	S (10^{-10} m)
Slow cooling	3.89×10^{-6}	11.75	37.74
Rapid cooling	2.92×10^{-6}	10.00	14.44

Figure 2 shows the magnetic hysteresis loops of both κ-H$_8$-Br and κ-D$_8$-Br at two temperatures (T = 2 K and T = 4.2 K), where the magnetic field varies from −10 KOe to 10 KOe. Our results show that the area of the hysteresis loops M(H) depended strongly on the temperature and the applied magnetic field; we observed a decrease in the area of increasing temperature and magnetic field. The effect was more pronounced in the deuterated (κ-D$_8$-Br) sample.

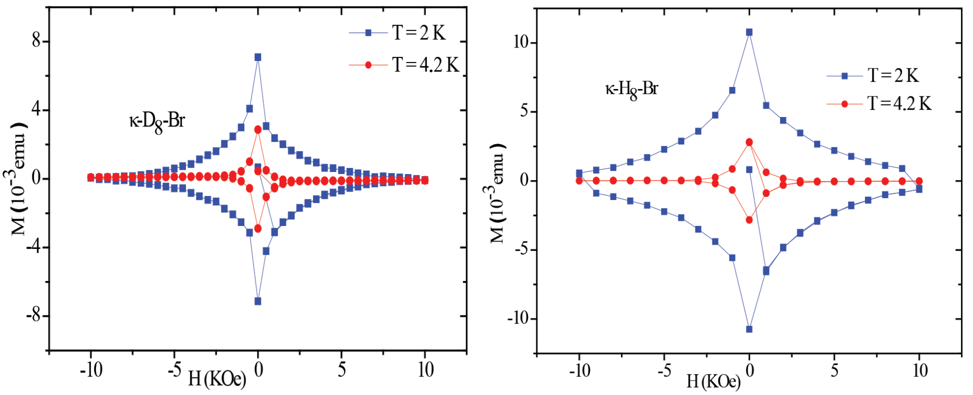

Figure 2: Magnetic hysteresis cycles at 2 K and 4.2 K for κ-H$_8$-Br and κ-D$_8$-Br.

The magnetic hysteresis loops of the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br were measured at different temperatures with the applied magnetic field perpendicular to the ab planes. J_C was calculated from the magnetisation hysteresis loops using Bean's model, $J_C = 30\Delta M/d$. ΔM (emu) and shows the difference between upper and lower branches of magnetic hysteresis loop, with d (m) the diameter of the sample. Figure 3 shows an example of the variations of this density as a function of the magnetic field for different temperature values and different cooling rates.

The J_C decreases with increasing temperature. We obtained high J_C values under slow cooling. As seen in Figure 3, the curve is divided into two regions in the field dependence of J_C. For each temperature, J_C(H) exhibits a slight decrease for magnetic fields below a value H*. H* is the field where the slope of the J_C(H) curve changes. The decrease is even more rapid for H > H*. H* depends strongly on temperature. In the two regions, J_C(H) varies approximately as $H^{-\alpha}$.

The increase in the magnetic field produced depinning of vortices from the pinning centres, followed by a rapid decrease of J_C. The existence of many pinning centres led to a higher current.

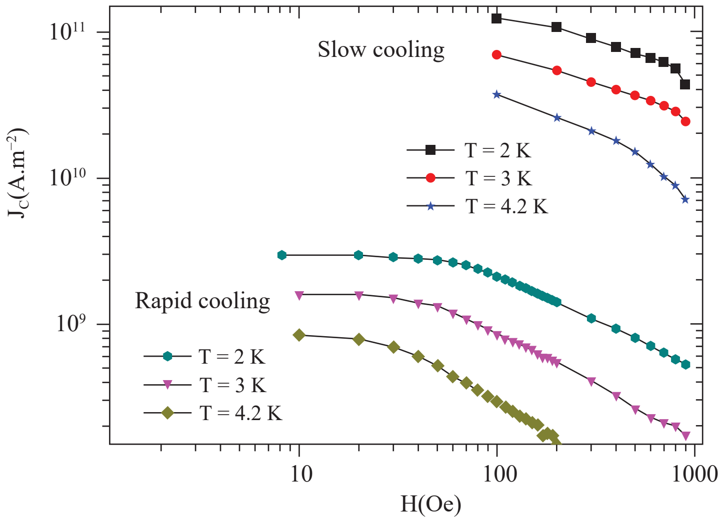

Figure 3: Magnetic field dependence of the J_C for different temperatures in slow and rapid cooling for a deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br sample. Solid lines are guides for the eye.

In the mixed state of a type II superconductor, the Lorentz force is the combination of the flux penetration and the transport current, with a direction perpendicular to both the current density and the magnetic field. Under the action of this force, the vortex begins to move; however, there is a pinning strength or "pinning force" that opposes this motion. There is a competition between the Lorentz and pinning forces: When the Lorentz force is greater than the pinning strength ($F_L > F_P$), the phenomenon is called "flux flow."[15] In the opposite scenario, when the pinning force is greater than the Lorentz force, we find "flux creep." The pinning of the flux lines in a type II superconductor is produced from vortex interaction with material defects that may be intrinsic or be induced through cooling. The borderline between the two forces is called the "irreversibility line."

In our study, the pinning strength of the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br was calculated using where H is the magnetic field and J_C(H) is the critical current density derived from the magnetisation hysteresis loops using Bean's model. Figure 4 shows the pinning strength F_P as a function of the applied magnetic field at 2 K and of the cooling rate for hydrogenated (κ-H_8-Br) and deuterated (κ-D_8-Br) κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br samples.

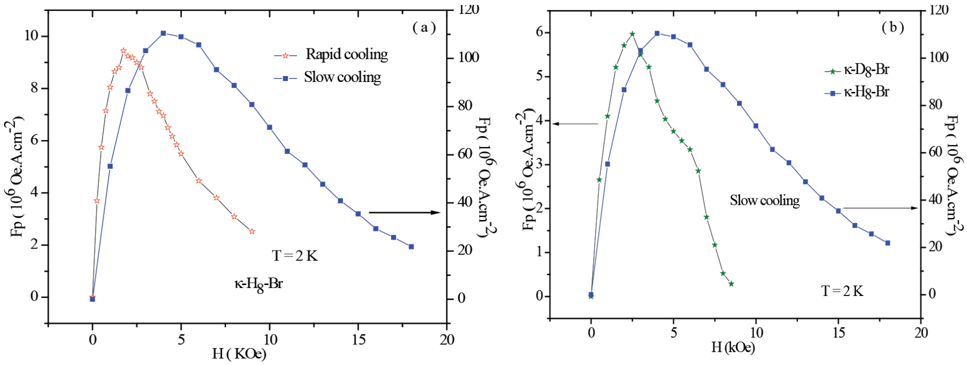

Figure 4: Variation of the pinning strength F_P with (a) magnetic field at T = 2 K for slow and rapid cooling of the κ-H_8-Br and (b) κ-H_8-Br and κ-D_8-Br for slow cooling at T = 2 K. Solid lines are guides for the eye.

Figure 4 shows the magnetic field dependence of the pinning strength $F_P(H)$ of samples after slow cooling and after slow and rapid cooling. Cooling through the structural transformation in the vicinity of 80 K has a dramatic effect on critical current density and pinning strength F_P. The slowly cooled sample in Figure 4(b) shows a high pinning strength $F_P(H)$ compared to the rapidly cooled sample, as in Figure 4(a). This result showed us that the pinning strength in the κ-H_8-Br superconductor was reduced with rapid cooling, and the slow cooling was effective in attaining high pinning strength. The effect is pronounced for the deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br compound where the hydrogen molecules of C_2H_4 are replaced by the deuterium. We postulated that the reduced pinning strength in Figure 4(a) was due to high density of impurity, with the rapid cooling producing disorder in the sample. Figure 5 shows the variation of the pinning strength (F_P) of deuterated (κ-D_8-Br) as a function of the applied magnetic field perpendicular to the ab planes at different temperatures.

This result shows that the pinning strength in the κ-D_8-Br superconductor depends strongly on temperature: a decrease of the pinning strength with increasing temperature. It should be noted that the pinning strength's strong temperature dependence is due to the large thermally activated flux motion in the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br system. For a given magnetic field, the decrease of pinning strength with temperature increase can be explained by the effective driving force (J-J_C) Φ_0 acting on the vortices. J is the current density and Φ_0 is the flux quantum. This driving force allows the vortices to overcome the pinning barriers and move along the superconductor. As J_C decreases with temperature (see Figure 3), an increase in temperature produces large values of the driving force; the small fractions of the pinning vortices accordingly reduce the pinning strength. For T = 10.5 K and

T = 10.8 K, the sample is not yet resistive; the pinning strength is still present but less intense. The two temperatures are situated at the superconducting transition region, and the pinning strength is very sensitive to small temperature variations, with thermal fluctuations contributing significantly to conductivity. Dissipation appears gradually, and we perceive a decrease of the pinning strength. The type II superconductors contain a variety of pinning centres randomly distributed in the sample. At low temperatures, pinning strength is more important than thermic activation. The bands of vortices with the largest dimensions are pinned. When the temperature increases, the depinning is controlled by that thermic activation. The bands of vortices with greater dimensions are not pinned but are in motion, giving rise to strong dissipation. When the temperature increases near the T_C, the dynamic of the vortices is dominated by the Lorentz force, decomposing the bands of vortices into units with small dimensions.

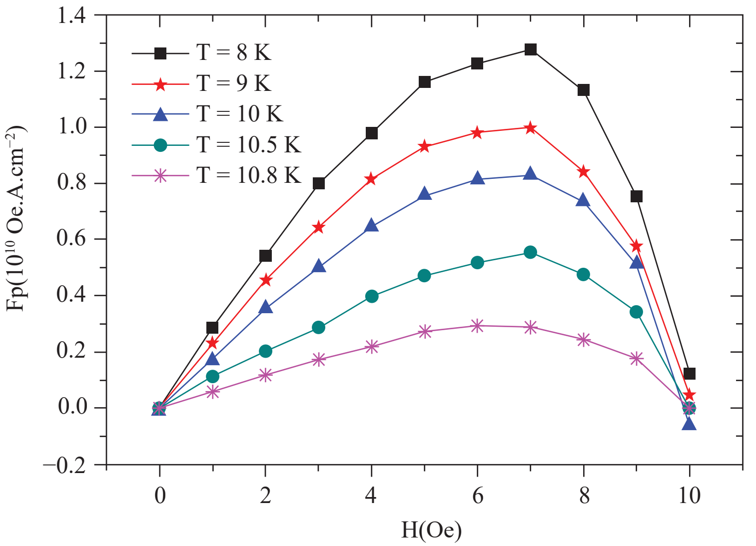

Figure 5: Pinning strength at different temperatures for slow cooling of deuterated κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$. Solid lines are guides for the eye.

4. CONCLUSION

In this paper, we have studied the effect of the cooling rate on magnetisation, the critical current density, and the pinning strength for both the hydrogenated and deuterated κ-$(BEDT\text{-}TTF)_2$ $Cu[N(CN)_2]Br$ samples. When cooled slowly, the pinning strength of κ-H_8-Br is intense compared to that of a rapidly cooled sample. The increase of the temperature decomposes the bands of vortices in small dimensions, with the pinning decreasing. The DC magnetisation strongly depends on the cooling rate. Rapid cooling through 80 K suppresses the superconducting phase and increases the magnetic phase. Our results show that the cooling through the order–disorder transformation at the vicinity of 80 K has a pronounced effect on the superconducting transition temperature, the pinning strength and the critical current density.

5. ACKNOWLEDGEMENTS

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