Temperature dependence of the magnetic moment relaxation rate of the organic superconductor $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$

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We have investigated relaxation of the residual magnetic moment of $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$, an organic superconductor, in the temperature range 2.4 to 9.5 K. In these investigations, we observed that the reduced relaxation rate $S=d\ln P_m/dt \ln t$ remains constant for $T/T_c<0.7$, and then increases rapidly with T. We also determined the temperature dependence of the critical current density in zero magnetic field. In comparing the functions $S(T)$ and $J_c(T)$ for this superconductor with their counterparts for single-crystal high-temperature superconductors, we have found that their behavior is most analogous to that of $s(T)$ and $J_c(T)$ for YBaCuO. Using the relaxation curves, we have calculated the current–voltage characteristics $E(J)$, which are well-described by a power law function of the type $E\propto J^n$.

1. INTRODUCTION

The $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$ superconductor , which was synthesized recently in the laboratory of E. B. Yagubskii,[1] has the highest critical temperature at ordinary pressure ($T_c\approx 11.6$ K) of any member of the family $\varkappa$-(BEDT-TTF)$_2$X. The physical properties of this cation radical salt are being studied intensely at this time.[2–4] As noted by Metlushko *et al.*[2] and Kushch *et al.*,[4] this superconductor is considerably less anisotropic than other compounds in this family. Thus, on the basis of their studies of reflection spectra, R. M. Vlasova *et al.*[3] have asserted that electron conductivity between cation layers is present, which has not been observed in any other member of this family. After determining that the conductivity anisotropy $\sigma_\parallel/\sigma_\perp \approx 10^2$ in the normal state at $T=290$ K and $\approx 10^3$ at $T=12$ K, while the anisotropy of the upper critical fields $H^\perp_{c2}/H^\parallel_{c2}\approx 5$, Kushch *et al.*[4] concluded that $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$ is a more three-dimensional superconductor than its pure bromide analog (for which $\sigma_\parallel/\sigma_\perp \approx 10^3$ at $T=290$ K and $\approx 10^4$ at $T=12$ K, with $H^\perp_{c2}/H^\parallel_{c2}\approx 10$). This conclusion also agrees with data from x-ray structural analysis.

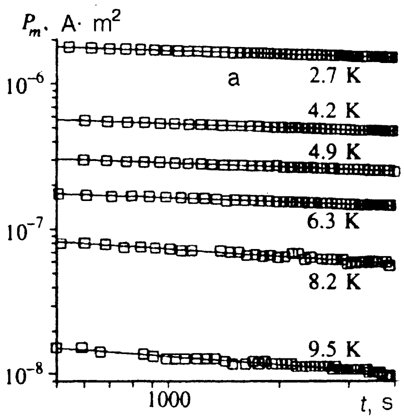
In their paper on high-temperature superconductors,[5] Metlushko *et al.* actively discussed the question of whether there is a connection between the anisotropy $\gamma=\xi_\parallel/\xi_\perp$ and the form of the functions $S(T)$ and $J_c(T)$. In light of this possibility, we decided it would be useful to investigate the relaxation of the magnetic moment and the function $J_c(T)$ over a wide range of temperatures and to compare the results we obtained with analogous data for single-crystal high-temperature superconductors.

2. OBTAINING THE SAMPLES AND EXPERIMENTAL METHOD

Single crystals with characteristic sizes $0.6\times 0.6\times 0.2$ mm^3 were prepared according to the standard electrochemical method.[1] The magnetic moment was measured with a SQUID magnetometer[6] for samples in the orientation **B**$\perp$ *bc*. The temperature was stabilized to an accuracy of ± 0.03 K. The relaxation of the magnetic moment was measured in the following way: the sample was first cooled in zero field from $T\approx 25$ K down to the required temperature. Then a rather large field (≈ 70 mT$>2B^*$, where B^* is the field corresponding to complete penetration of the magnetic field into the sample; for $T=2.1$ K, $B^*\approx 18$ mT) was first switched on and then off. The rates at which the field was switched on and off were kept the same in all the measurements, in order to exclude the possibility that differences in these rates could affect the results of the relaxation rate measurements. Note that our procedure is, in practice, equivalent to cooling in a field with subsequent removal of the field while ensuring that the condition $D_{\max}\approx 2B^*$ is satisfied. The relaxation of the magnetic moment was measured for $t=30$ to 4000 seconds in the range of temperatures 2.4 to 9.5 K. The reduced relaxation rate $S=d\ln P_m/d\ln t$ was determined on the interval 1000 to 4000 s by the least-squares method. Typical relaxation curves are shown in Figs. 1a, 1b. It is clear that the experimental points are close to the curve when plotted in logarithmic coordinates.

The temperature dependence of the critical current density was obtained by a similar experimental method: after the sample was cooled to $T=2.1$ K, a magnetic field was switched on and then off (under the same conditions as the relaxation study). The magnetic moment was then measured over the temperature range 2.1 to 10.5 K, and these measurements were used to determine the value of $J_c(T)$ according to Bean's model[7] (in our case, the value of the magnetic moment corresponded to the half-width of the hysteresis loop of $\Delta P_m(B_e,T)/2$ in zero field $B_e\approx 0$).

The values of $\Delta P_m(B_e\approx 0,T)/2$ obtained by measuring the hysteresis of $P_m(B_e)$ at a fixed temperature T were in



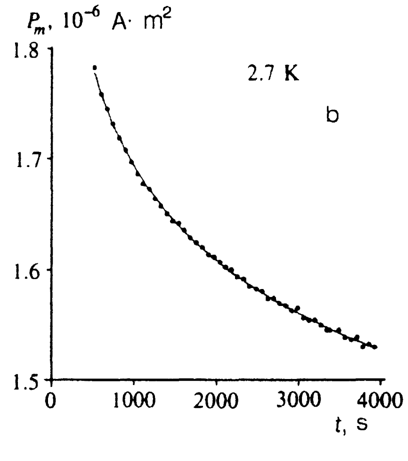


FIG. 1. a) Relaxation curves $P_m(t)$ for the compound $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$ for B⊥ *bc*; b) relaxation curve $P_m(t)$ on a larger scale (the solid curve is a fit using $P_m(t) \propto t^{-S}$).

good agreement with the data obtained by our method.

3. RESULTS AND DISCUSSION

The results of our measurements are shown in Figs. 2 and 3. It is clear that the function $J_c(T)$ has a quasi-exponential character in the low-temperature region, and that for $T/T_c \gtrsim 0.7$ the critical current density decreases more abruptly. Analogous behavior of $J_c(T)$ is often encountered in the high-temperature superconductors.[8] The reduced relaxation rate $S(T)$ is likewise almost temperature independent in the range $T/T_c \lesssim 0.7$, while for $T/T_c \gtrsim 0.7$ it increases abruptly. Figure 4 (taken from Ref. 5) shows the functions $J_c(T)$ and $S(T)$ for a number of high-temperature superconductor single crystals. In comparing these data with our results it is clear that, based on the character of the function $J_c(T)$ (for $0.17 \lesssim T/T_c \leqslant 0.82$) and $S(T)$ (for $T/T_c \lesssim 0.7$), the superconductor $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$ is qualitatively closest to YBaCuO. The rise in $S(T)$ for $T/T_c \gtrsim 0.7$ is somewhat reminiscent of the behavior of BaKCuO in the region $T/T_c \gtrsim 0.8$. The resemblance we have noted could be connected with the fact that YBaCuO and $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$ have approximately the same anisotropy ($\gamma \approx 5$; see Refs. 4, 9). For other members of the family $\varkappa$-(ET)$_2$X the value of γ is of the same order ($\gamma \approx 4$ to 8; see Refs. 10 to 12). The scatter in values of γ is associated with differences in the method for determining $T_c(H)$.

However, it is worth noting that absolute values of the relaxation rate S in the organic superconductor are roughly four times larger, while the absolute values of T_c and J_c are respectively a factor of ten and a factor 10^2 to 10^3 times lower than they are in the cuprate superconductors.

From the temperature dependence of the relaxation rate $S(T)$ we can determine the temperature dependence of the effective pinning energy $U_{\text{eff}} = k_B T/S$ (Fig. 5). The weak dependence of the relaxation rate for $T/T_c \lesssim 0.7$ corresponds to an increase in $U_{\text{eff}}(T)$. A likely explanation for this could be the fact that the pinning energy has a broad distribution.[13] If this is the case, at low temperatures the shallow centers will play the dominant role in pinning, with deeper centers replacing them at higher temperatures.

The values we have obtained for $U_{\text{eff}}(T)$ are in good agreement with the results for $U_{\text{eff}}(T,B)$ obtained by measuring the relaxation while cooling the sample to $T=4.2$ K in zero field, both for the compound under investigation and for the salts $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br and $\varkappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$.

Another explanation for the plateau in the curve $S(T)$

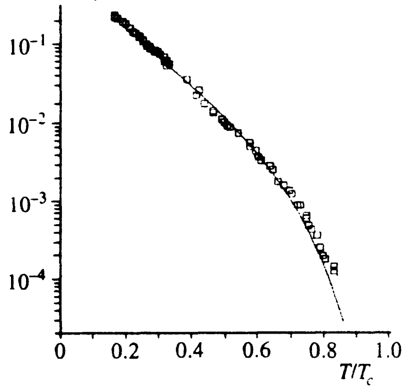


FIG. 2. Temperature dependence of the critical current density for $\varkappa$-(BEDT-TTF) $_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$, B⊥ *bc*, $T_c \approx 11.6$ K.

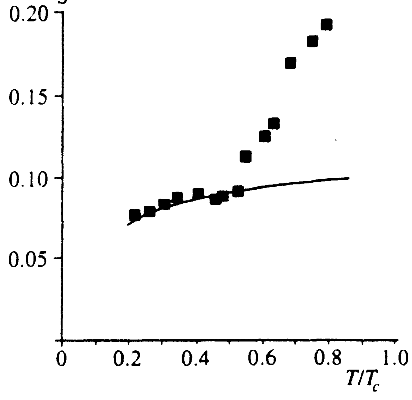


FIG. 3. Dependence of the reduced relaxation rate on the reduced temperature for $\varkappa$-(BEDT-TTF) $_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$, B⊥ *bc*, $T_c \approx 11.6$ K.

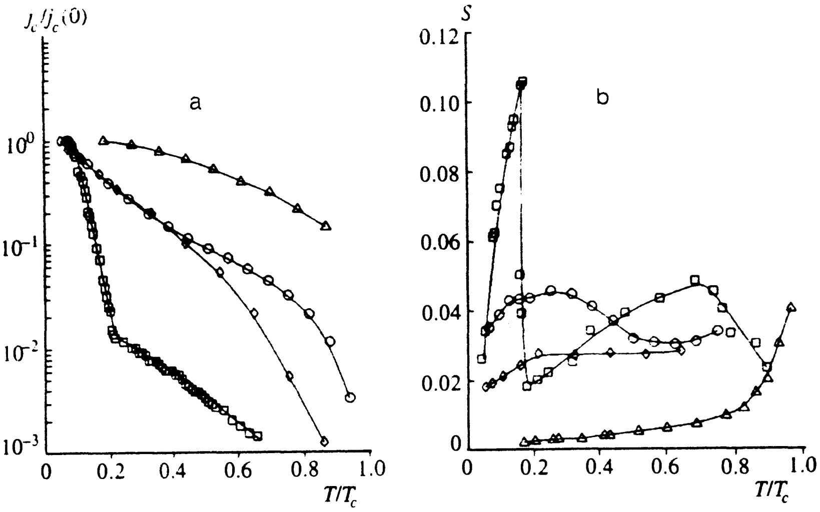


FIG. 4. Dependence of the critical current density J_c (a) and the reduced relaxation rate S (b) on temperature[5] for a number of high-temperature superconducting compounds: □—BSCCO, ◇—YBCO, △—BKBO, ○—PSYCCO.

was proposed by Thompson *et al.*,[14] who argue as follows: relaxation of the magnetic moment is due to thermally activated vortices, whose creation leads to the measured decrease in the critical field. As the temperature approaches T_c the relaxation rate increases so much that the temperature dependence of J_c is primarily determined by flux creep. Starting from the theory of collective pinning, these authors proposed the following relation between the critical current J_{c0}, which has a BCS type of temperature dependence, and the measured critical current J_c:

$$J_c(T)=J_{c0}\left[1+\left(\frac{\mu k_B T}{U_0}\right)\ln\left(\frac{t}{t_{\mathrm{eff}}}\right)\right]^{-1/\mu}, \tag{1}$$

where U_0 depends only on temperature. The reduced relaxation rate in this case is expressed as follows:

$$S(T)=-\frac{k_B T}{U_0+\mu T k_B \ln(t/t_{\mathrm{eff}})}. \tag{2}$$

The temperature dependences of J_c0 and U_0 have the form

$$J_{c0}=J_{c00}[1-(T/T_c)^2]^{3/2}, \tag{3}$$

$$U_0=U_{00}[1-(T/T_c)^2]^{3/2}.$$

Fitting Eqs. (1)–(3) to our experimental data gives the following parameters: $\mu\approx 0.5$, $\mu \ln(t/t_{\mathrm{eff}})\approx 20$, $U_{00}\approx 1$ meV, and $t_{\mathrm{eff}}\approx 10^{-8}$ s. The solid curves for the functions $J_c(T)$, $S(T)$ plotted in Fig. 2 correspond to Eqs. (1)–(3) with these parameters. An admitted deficiency of this procedure is that the fitted expressions are unable to describe the abrupt increase in the relaxation as T_c is approached.

The relaxation curves $P_m(t)$ can be used to obtain the current–voltage characteristics $E(J)$, because $J\propto P_m$ and $E\propto dP/dt$. For this we used the following relation:[2]

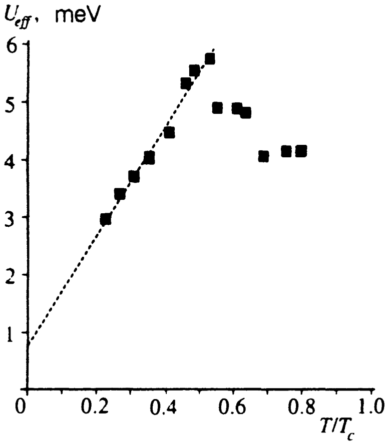


FIG. 5. Temperature dependence of the effective pinning energy U_{eff}.

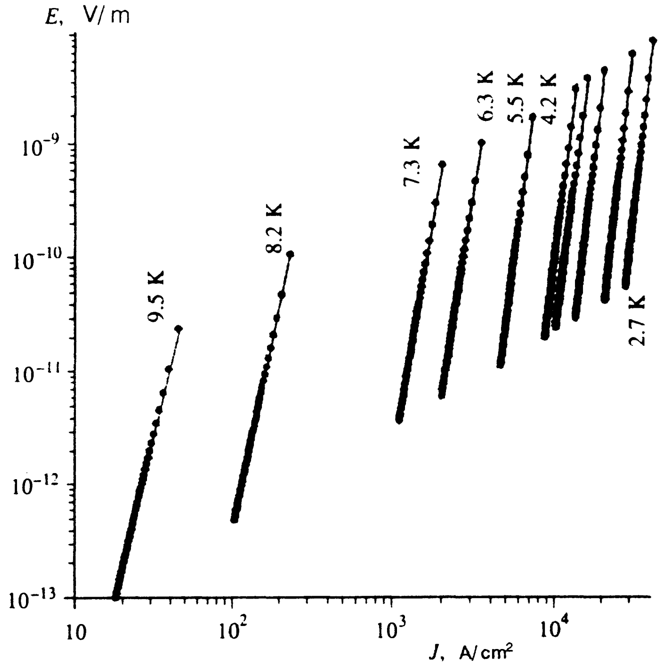


FIG. 6. Current–voltage characteristics of $\varkappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl$_{0.5}$Br$_{0.5}$.

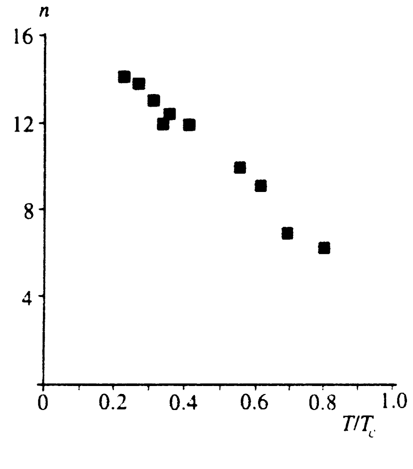


FIG. 7. Temperature dependence of the exponent n.

$$E = \frac{3\mu_0 I}{2\pi^2 R^2} \frac{dP_m}{dt},$$

where R is the characteristic size of the sample and $I=3.328$. The $E(J)$ characteristics we obtain are shown in Fig. 6. It is clear that they are well-described by power law functions of the form $E/E_0=(J/J_0)^n$ over the entire temperature range studied. This result is in good agreement with the data of Metlushko *et al.*,[2] who noted that for small fields the $E(J)$ curves are good straight lines in logarithmic coordinates, although at large fields $E \propto \exp(-J/J_0)$. The dependence of the exponent n on temperature shown in Fig. 7 is analogous to the field dependence $n(B)$ given in Ref. 2. As already noted in this reference, for $t \gg t_0$ the exponent of the power law n equals $1/S$ (see Figs. 3 and 7).

4. BASIC RESULTS AND CONCLUSIONS

We have investigated the relaxation of the residual magnetic moment of the superconductor $\varkappa-(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$ in the temperature range 2.4 to 9.5 K, and have shown that the reduced relaxation rate $S=d \ln P_m/d\ln t$ of this superconductor remains constant for $T/T_c \lesssim 0.7$, and then increases abruptly. We have established that the temperature dependence of the critical current density has a quasi-exponential character for $T/T_c \lesssim 0.7$.

It is noteworthy that the functions $S(T)$ and $J_c(T)$ for this superconductor are qualitatively similar to analogous curves for YBaCuO. This fact may be considered a new argument in favor of the correlation[5] between anisotropy γ and the presence of this or some other feature in the curves $S(T)$ and $J_c(T)$.

From the relaxation curves we calculated the current–voltage characteristics $E(J)$, which are well-described by power law functions of the type $E \propto J^n$ over the entire temperature range under study.

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