Orientational dependence of the magnetic moment relaxation rate of the organic superconductor κ-$(ET)_2Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$

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Hysteresis curves and relaxation curves (in a residual magnetization regime) are obtained for the organic superconductor κ-$(ET)_2X(X=Cu[N(CN)_2]Cl_{0.5}Br_{0.5})$ at various values of the angle θ between the external magnetic field **B** and the basal plane of the crystal. It is established, on the one hand, that the critical current density in the plane $J_c^{\parallel}$ does not depend (to within the accuracy of the measurements) on the orientation of the single crystal and, on the other hand, that the relaxation rate decreases as θ increases. © *1996 American Institute of Physics.* [S1063-7761(96)01910-5]

1. INTRODUCTION

One characteristic feature of both high-T_c superconductors and organic superconductors of the κ-$(ET)_2X$ family is their layered structure: the crystal structure consists of alternating conducting and insulating layers. Such a structure leads to qualitatively different (compared with isotropic superconductors) behavior in a magnetic field. In the two limiting orientations $\mathbf{B}\|\hat{\mathbf{c}}$ and $\mathbf{B}\perp\hat{\mathbf{c}}$ the structure of a vortex is characterized by $\lambda^{\parallel}$ and $\xi^{\parallel}$ and by $\lambda^{\perp}$ and $\xi^{\perp}$, respectively. The structure of the vortices varies as the orientation varies, causing variation of the elastic moduli of the vortex lattice and, therefore, variation of the behavior of the superconductor in the critical state. In the case of the pinning of vortices on randomly distributed point defects, a situation in which the pinning energy does not depend on the orientation is possible under certain conditions. Then the orientational dependence of such characteristics as the relaxation rate S and the critical current density J_c is attributed mainly to the variation of the properties of the vortex lattice.

In layered superconductors two critical current densities must be distinguished: the critical current density along the layers $J_c^{\parallel}(\theta)$ and the critical current density perpendicular to the layers $J_c^{\perp}(\theta)$ (see Fig. 1). The critical current density perpendicular to the layers $J_c^{\perp}(\theta)$ is significantly smaller than the critical current density parallel to the layers $J_c^{\parallel}(\theta)$; therefore, in magnetic measurements the magnetic moment is determined mainly by $J_c^{\parallel}(\theta)$. The form of the dependence of $J_c^{\parallel}$ on θ is determined by the pinning regime, which depends on the mean intervortex distance $\langle a_0\rangle\approx(\Phi_0/\langle B\rangle)^{1/2}$ (in addition to the properties of the pinning centers). For the pinning of individual vortices neither $J^{\parallel}(\theta)$ nor $U(\theta)$ depends on the angle θ. For the pinning of small "bundles" the functions $J(\theta)$ and $U(\theta)$ have[1] the following forms:

$$J_c^{\parallel}(\theta)\simeq J_c^{\parallel}(0)\varepsilon(\theta),\quad U(\theta)\simeq U_c(0)(\varepsilon(\theta))^{1/2},$$

$$\varepsilon(\theta)=(\cos^2\theta+(\gamma\sin\theta)^2)^{1/2}.$$

Here $\gamma=\xi^{\parallel}/\xi^{\perp}$ is the anisotropy parameter. Thus, the orientational dependences of the critical current density and the relaxation rate are intimately related to the flux creep regime. It was established in Refs. 2 and 3 that the dependence of the activation energy on the current density in the organic superconductor κ-$(ET)_2Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$ can be described as $J^{-\mu}$ with $\mu\approx0.5$. According to the collective pinning theory, such a value of μ corresponds to the pinning of small bundles in weak fields, where the mean intervortex distance $a_0\approx(\Phi_0/B)^{1/2}$ is greater than λ/γ (where λ is the coherence length). In addition, it was pointed out in Ref. 4 that the anisotropy of J_c cannot be attributed to the anisotropy of the pinning energy U. Therefore, measurements of $J_c(\theta)$ and $S(\theta)$ might provide interesting information on the vortex pinning mechanism.

2. EXPERIMENTAL METHODS

Single crystals of the organic superconductor κ-$(ET)_2Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$ were obtained by means of the standard electrochemical technology.[5] A single crystal (measuring $0.5\times0.5\times0.2$ mm^3) was attached by a Ramsay cylinder to a holder made from fused quartz (see Fig. 2). This holder makes a negligibly small contribution to the measured signal. The orientation was varied manually under a microscope on a standard rotating stage. The quartz tube was rigidly fixed relative to the rotating stage, and the flat surface of the movable cylinder with the crystal was located at the focus of the microscope. The angle θ was determined from the deviation of the ruled line relative to the axis of the holder (the diameter of the holder was only 0.5 mm less than the diameter of the guides, making it possible to determine the orientation of the holder relative to the magnetic field $\mathbf{B}_a$ to better than 0.2°). The accuracy of the orientation of the crystal was approximately $\pm1°$.

The measurements of the hysteresis curves and the relaxation dependences were performed on a SQUID magnetometer[6] according to the following procedure. The sample was slowly cooled (≈1 h) to 4.2 K in a zero field. Then the field was applied in approximately equal "steps," and the field dependence of the magnetic moment was measured at a given orientation. During this process the magnetic field was first increased to $B_{max}\approx70$ mT and then decreased to zero (the residual field amounted to ≈0.1 mT). When a zero field was achieved, measurements of the temporal dependence of the magnetic moment were performed over the

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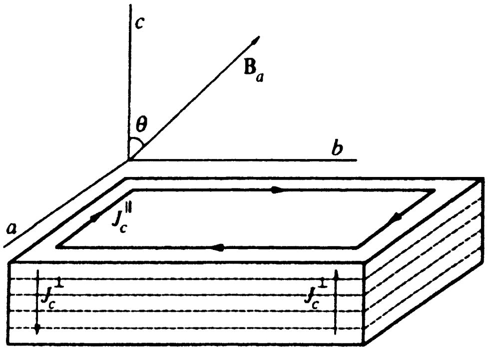

FIG. 1. Orientation of the single crystal relative to the field.

course of 4000 sec. Before removing the holder with the sample to change the orientation of the sample, we slowly heated the sample (≈ 1 h) to room temperature. Slow heating was employed to avoid strong mechanical stresses on the boundary between the crystal and the Ramsay cylinder. Measurements were performed for $\theta = 0°$, $7°$, $15°$, $21°$, $30°$, $37°$, $43°$, $52°$, $61°$, $74°$, $82°$, and $86°$.

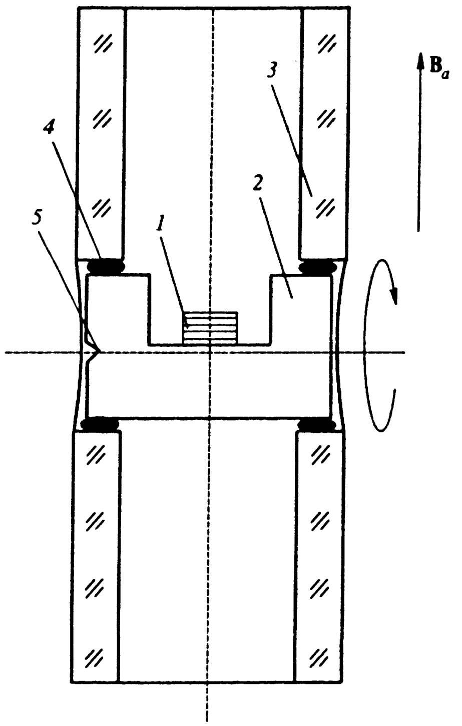

FIG. 2. Quartz holder: *1* — single crystal, *2* — movable quartz cylinder, *3* — quartz tube, *4* — Ramsay cylinder, *5* — ruled line on the end surface of the flat cylinder.

3. RESULTS AND DISCUSSION

The magnetization curves obtained are presented in Fig. 3a. It is seen that the width of the hysteresis loop decreases systematically as the angle θ increases. The relaxation rate of the residual magnetic moment $S = d \ln P_m / d \ln t$ also decreases with increasing θ (see Fig. 4 below).

Note that we intentionally did not perform measurements of the hysteresis curves at angles close to $\theta = 90°$ for the following reasons. For the $\mathbf{B} \| ab$ orientation the magnitude of the magnetic moment P_m is determined only by $J_c^{\perp}$, and since it satisfied $J_c^{\perp} \ll J_c^{\parallel}$, even slight nonuniformity of the magnetic field ($\delta B_a / B_a \approx 0.1\%$ in the scan length of the sample) can lead to complete magnetization reversal of the superconductor and a signal of irregular form[7] and can, therefore, render the measurements meaningless. It is noteworthy that the magnetization reversal is due to the weakness of the complete penetration field B_t ($B_t \simeq \mu_0 w J_c < \delta B_a$, and w is the transverse dimension of the sample) and depends weakly on the shape of the sample.

In discussing the results obtained the following is noteworthy. The dependence of the magnetic moment on the orientation can be attributed to several factors. First of all, even in the case of an isotropic sample of aspherical shape, the residual magnetic moment depends on the orientation (so-called shape anisotropy[8]). This is because the magnetization $m = P_m / V$ depends on the characteristic geometric dimension $W(\theta)$: $m \simeq W(\theta) J_c / 3$. Here

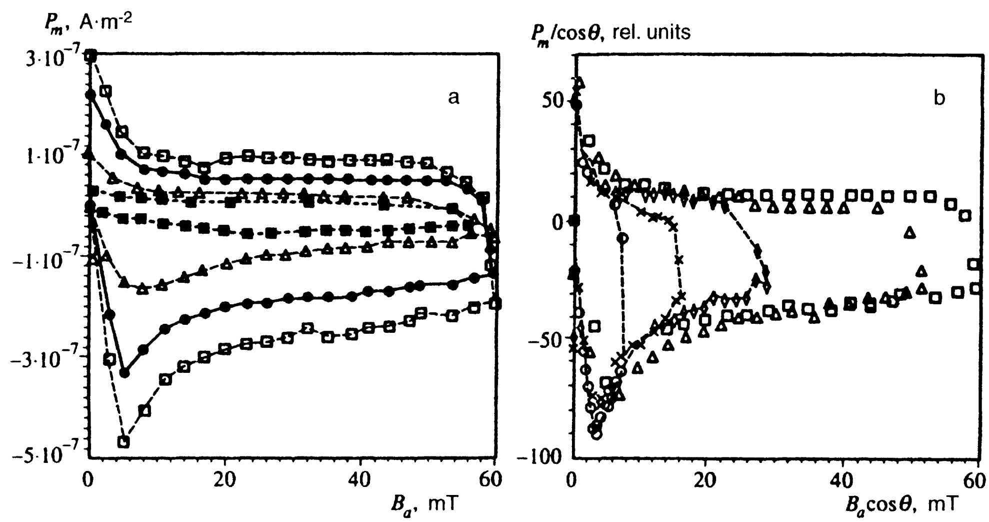

FIG. 3. Hysteresis curves for several values of θ: a—$\theta = 0°$ (□), 45° (●), 61° (△), 82° (■); b — $\theta = 0°$ (□), 30° (△), 61° (◇), 74° (×), 82° (○).

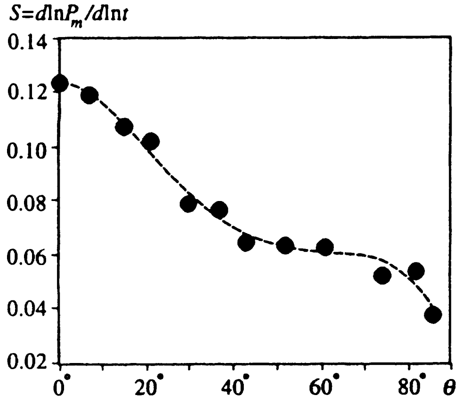

FIG. 4. Orientational dependence of the normalized relaxation rate $S(\theta)$.

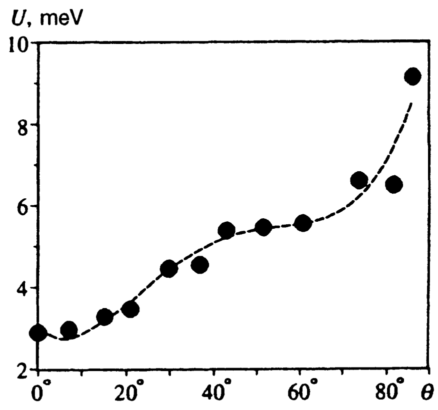

FIG. 5. Orientational dependence of the pinning energy $U(\theta)$.

$$W(\theta)\simeq((w \cos \theta)^2+(t \cos \theta)^2)^{1/2},$$

where t is the thickness of the sample. In our case the influence of the shape anisotropy was insignificant, since the sample had the small ratio $w/t\approx 2.5$. In the case of an anisotropic superconductor, the anisotropy of the critical current is also superimposed on the shape anisotropy. It should be noted that there are two parameters which characterize the degree of two-dimensionality of a superconductor: the first is the anisotropy of the coherence length $\gamma=\xi^{\parallel}/\xi^{\perp}$, which is determined by the anisotropy of the effective mass of the current carriers; the second is the ratio of the coherence length to the distance between layers $\xi^{\perp}/d$, which characterizes the effectiveness of the interaction between the conducting layers. We stress that the latter parameter $\xi^{\perp}/d$ is especially small in organic superconductors [≈ 0.3 (Ref. 9)], although the anisotropy of the coherence length is comparatively small ($\gamma=\xi^{\parallel}/\xi^{\perp}\approx 5-8$, i.e., it is approximately the same as in Y–Ba–Cu–O).

The experimental data obtained for different values of θ satisfactorily fit a single curve (see Fig. 3b), if they are represented in $P_m(B_a,\theta)/\cos\theta$ versus $B_a \cos \theta$ coordinates. Of course, the quantity $B_{\max}\cos\theta$, which corresponds to the turning point of the hysteresis curve is different for different orientations. Such a result can be interpreted in the following manner. The magnetic moment P_m is governed by the current $J_c^{\parallel}$ in the ab basal plane, which remains practically unchanged for all orientations.

Such behavior correlates well with the predictions of the collective creep theory for the case in which pinning on randomly distributed point defects involves individual vortices. Point defects are supported by the strong exponential dependence of the critical current density on the magnetic field $J_c(B) \propto \exp(-B/B_0)$ (Ref. 4). The pinning potential created by such point defects is essentially isotropic, and in the case of the pinning of individual vortices this leads to the lack of a dependence of the current in the plane $J_c^{\parallel}$ on the angle θ.

Unlike $J_c^{\parallel}$ the normalized relaxation rate $S(\theta)=d \ln P_m/d \ln t$ does not remain constant, but decreases monotonically (see Fig. 4). The decrease in $S(\theta)$ corresponds to an increase in the pinning energy $U(\theta)$ as θ increases (for brevity we call the activation energy for the motion of Abrikosov vortices the pinning energy) (see Fig. 5). An increase in $U(\theta)$, unfortunately, is not consistent with the picture considered above of the pinning of individual vortices, since in that case S, like $J_c^{\parallel}$ does not depend on θ. When the results are interpreted within the collective creep theory, the dependence of the relaxation rate on the orientation indicates the pinning of small bundles. This is also consistent with the value of the exponent $\mu\approx 0.5$ in the dependence $U(J) \propto J^{-\mu}$ (Refs. 2 and 3), which corresponds to the pinning of small bundles in weak fields $\langle a_0\rangle\approx(\Phi_0/\langle B\rangle)^{1/2}>\lambda/\gamma$. Since in a residual magnetization regime we have $\langle B\rangle\approx P_m/V\approx 5$ mT, $\lambda\approx 500$ nm, and $\gamma\approx 5-8$, we obtain $\langle a_0\rangle\approx 1000$ nm$>\lambda/\gamma\approx 160$ nm.

We note that the different dependences of $S(\theta)$ and $J_c^{\parallel}(\theta)$ in κ-$(ET)_2Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$ can indicate behavior that is more complicated than the pure pinning of individual vortices or the pure pinning of small bundles.

4. MAIN RESULTS AND CONCLUSIONS

The orientational dependence of the magnetic moment and relaxation rate in the organic superconductor κ-$(ET)_2Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$ has been investigated.

It has been established that to within the experimental error the hysteresis curves are determined by the currents $J_c^{\parallel}$ in the ab plane, which do not depend on the angle θ.

An analysis within the collective creep theory allows us to state that the hysteresis data indicate the pinning of individual vortices, but the relaxation measurements are more consistent with the pinning of small bundles of vortices. The latter assertion is also consistent with the data in Refs. 2 and 3, where it was noted that the pinning energy can be described as $U(J) \propto J^{-0.5}$.

As a whole, the behavior of the superconductor κ-$(ET)_2X$ ($X=Cu[N(CN)_2]Cl_{0.5}Br_{0.5}$) in the critical state is interpreted satisfactorily within the collective pinning model, which gives a consistent description of relaxation measurements in different regimes.[1]

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