

# The current-density dependence $U(J)$ of the activation energy for the motion of Abrikosov vortices in the organic superconductor $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Cl<sub>0.5</sub>Br<sub>0.5</sub>

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We have investigated the thermally activated motion of flux in the organic superconductor  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Cl<sub>0.5</sub>Br<sub>0.5</sub> over a wide range of current densities  $J$ , and have obtained the dependence of the activation energy  $U$  for this motion on current density. © 1995 American Institute of Physics.

## 1. INTRODUCTION

Whereas in classical type-II superconductors flux creep is observed<sup>1</sup> only at temperatures close to the critical temperature  $T \rightarrow T_c$ , relaxation of the critical current takes place over practically the entire temperature range in oxide superconductors<sup>2</sup> and in the cation-radical salts<sup>3,4</sup>  $\kappa$ -(ET)<sub>2</sub>X. Furthermore, this relaxation also occurs over a wide range of current densities, although for the classical superconductors it is observed only for  $J \approx J_c$ . Strong relaxation of  $J_c$  in high-temperature superconductors and cation-radical salts is associated in particular with small coherence lengths  $\xi$ , which give very small pinning energies  $U_p \approx (B_c^2 \xi^3)/\mu_0$  and large penetration depths  $\lambda$  for magnetic fields. This in turn leads to “softening” of the vortex lattice and facilitates depinning of the vortices.<sup>5</sup>

Several models have been proposed to describe the relaxation of screening currents in type-II superconductors.<sup>6–9</sup> All of them are based on an Arrhenius relation between the hopping frequency  $\nu$  of the Abrikosov vortex and the activation energy  $U$ :

$$\nu = \nu_0 \exp[-U(J, B, T)/k_B T],$$

the only difference between these models is the different forms of the functions  $U(J)$ .

The basic relation that describes thermally activated motion of flux is the equation for flux conservation<sup>10</sup>:

$$\frac{\partial \mathbf{B}}{\partial t} = \left[ \nabla \frac{[\mathbf{B}[\mathbf{J}\mathbf{B}]]}{|[\mathbf{J}\mathbf{B}]|} \right] \nu_0 \exp[-U(J, B, T)/k_B T]. \quad (1)$$

There exist several methods based on Eq. (1) for “de-embedding” the function  $U(J)$  from relaxation data. Let us discuss one method, proposed in Refs. 11, 12, for the example of a thin film (with thickness  $h$  and width  $w$ , and a length  $\gg w$ ) oriented perpendicular to the field. Assume that screening currents flow through the entire bulk of the sample, and that the external field satisfies  $B_e \approx 0$  (this corresponds to measuring the relaxation of the residual magnetic moment). Then, by integrating Eq. (1), we obtain (assuming that  $J = \text{const}$  for a given  $T, t$ ):

$$\frac{d\langle B \rangle}{dt} = L \langle B \rangle \nu_0 \exp[-U(J, B, T)/k_B T],$$

where  $L \approx (1/w) \ln(w/h)$ .

Measurement of the relaxation of the residual magnetic moment yields  $\langle B \rangle \propto P_m/V$ . Then for fixed temperatures  $T$  [neglecting the dependence  $J(B)$ ], we find:

$$\frac{U(J)}{k_B T} = \ln \left| \frac{dP_m(t)}{P_m(t) dt} \right| - C, \quad (2)$$

where  $C = \ln(L\nu_0)$ .

In what follows, this relation will be used to analyze relaxation curves taken at various temperatures. The need to use relaxation data at various temperatures is connected with the fact that for ordinary measurement times (i.e.,  $t_{\text{mes}} = 10^3 - 10^4$  s), the relative change in  $J$  is not large ( $\approx 10\%$ ). In this case we must introduce certain scaling functions  $g(T)$  and  $f(B)$  in order to obtain a smooth curve  $U(J)$ .

Another method of obtaining the function  $U(J)$  is the inverse scheme proposed in Refs. 13, 14. However, this method is tedious, and its correct use requires a very large amount of experimental data.

One of the goals of this work was to find an alternate method for obtaining the function  $U(J)$ , one that perhaps would require less time to implement.

## 2. EXPERIMENTAL METHOD

We obtained crystals of  $\kappa$ -(ET)<sub>2</sub>-Cu[N(CN)<sub>2</sub>]Cl<sub>0.5</sub>Br<sub>0.5</sub> with characteristic dimensions  $0.6 \times 0.6 \times 0.2$  mm<sup>3</sup> by standard electrochemical techniques.<sup>15</sup> The relaxation of the magnetic moment was measured using a SQUID magnetometer<sup>16</sup> at liquid helium temperatures. The magnetic field  $B_e$  was applied perpendicular to the conducting layer of the crystal. Temporal stability of the magnetic field over the time of measurement was better than  $10^{-9}$  T/s.

Normally, the relative decrease in the magnetic moment seen in relaxation experiments over these time periods is  $\leq 10\%$ . Therefore, to extend the range over which  $J$  changes we must measure the relaxation over a much longer time<sup>17</sup> ( $t \approx 3 \cdot 10^6$  s). In order to decrease this measurement time we devised the following procedure.

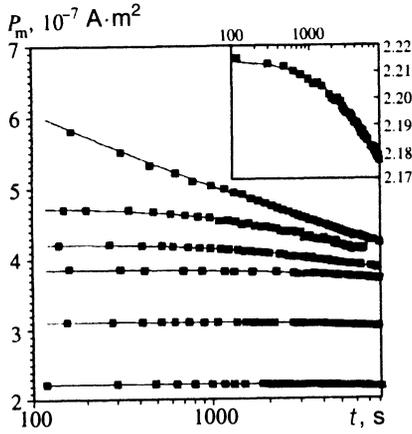


FIG. 1. Typical relaxation curves  $P_m(t)$ . The inset shows the relaxation curve for small values of  $J$  on an expanded scale.

The crystal under study was first cooled in zero magnetic field from  $T \approx 25$  K ( $T_c = 11.6$  K) down to a certain temperature  $T_1 > 4.2$  K. The external field was then increased to  $B_{\max} \approx 70$  mT, after which it was decreased to zero (the residual field  $B_e \approx 0.1$  mT). Because the maximum field satisfies  $B_{\max} \gg 2B^*$  (where  $B^*$  is the field at which the magnetic flux fully penetrates the sample), we obtain a superconductor throughout whose entire volume a current flows that is close to the critical current at temperature  $T_1$ . After this, the sample is rapidly cooled ( $\approx 1-2$  s) down to  $T = 4.2$  K, where it is maintained for five minutes, and the relaxation  $P_m(t)$  is measured over a period of  $\approx 8000$  s. Using this method, we can vary the magnitude of the current flowing through the bulk of the sample by varying the temperature  $T_1$ .

### 3. RESULTS AND DISCUSSION

Typical relaxation curves are shown in Fig. 1. It is clear that as the quantity  $P_m \propto J$  decreases, the relaxation becomes nonlogarithmic. A similar deviation from the logarithmic law is observed for Y-Ba-Cu-O (Ref. 17) when the long-period relaxation ( $t = 10^2 - 3 \cdot 10^5$  s) of the critical current is measured at a temperature  $T = 70$  K over a time  $t > 10^5$  s. It is noteworthy that for  $j \approx 5 \cdot 10^2$  A/cm<sup>2</sup> we observe no decrease in the magnetic moment ( $\Delta P_m \approx 4 \cdot 10^{-11}$  A·m<sup>2</sup> within experimental error) when  $P_m(t)$  is measured over  $\approx 3 \cdot 10^1$  s. Thus, the rate of relaxation for  $J \approx 5 \cdot 10^2$  A/cm<sup>2</sup> is smaller than for  $J \approx 10^{-15}$  A·m<sup>2</sup>·s<sup>-1</sup>.

In order to use Eq. (1) to analyze the relaxation data, we assume that  $C = \text{const}$  for  $T = \text{const}$ . If we measure the activation energy from the level  $U'$  corresponding to the maximum relaxation rate observed in experiment, Eq. (1) becomes

$$\frac{U(J) - U'}{k_B T} = \ln \left| \frac{dP_m(t)}{P_m(t) dt} \right| - \ln \left| \frac{dP'_m(t')}{P'_m(t') dt} \right|, \quad (3)$$

where  $U'$  is the activation energy for the corresponding relaxation rate  $dP'_m(t')/dt$  at time  $t'$ . Note that the choice of  $U'$  does not affect the shape of the curve  $U(J)/k_B T$ .

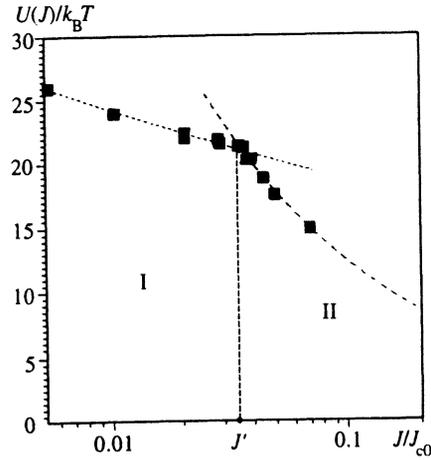


FIG. 2. The function  $U(J)/k_B T$ . For the dashed curves see the text.  $J_{c0} = 5 \cdot 10^4$  A/cm<sup>2</sup>.

In order to obtain the absolute value of  $U(J)$ , it is convenient to take for  $dP'_m(t')/dt$  and  $P'_m(t')$  the residual value of the exact magnetic moment for the corresponding "true" critical current  $J_{c0}(T)$ , because  $U(J_{c0}) \equiv 0$ . The characteristic time for the beginning of flux creep  $t_{FC}$  was estimated in Ref. 18:

$$t_{FC} = \frac{E_c}{\rho_c} \frac{\mu_0 d^2}{dB_e/dT} \frac{k_B T}{U_c}.$$

Substituting  $k_B T/U_c \approx 10^{-2}$ ,  $E_c/\rho_c \approx 10^5$  A/cm<sup>2</sup>,  $d \approx 10^{-4}$  m,  $dB_e/dT \approx 10^{-1}$  T/s, we obtain  $t_{FC} \approx 10^{-5} - 10^{-6}$  s.

In what follows, by approximating the initial segment of the relaxation curve as a power-law function  $P_m \propto t^{-s}$  and extrapolating to  $t' \approx t_{FC}$  we obtain  $C \approx \ln(S/t') \approx 10$  and  $v_0 \approx wS/t' \approx 10^3$  m/s,  $J_{c0} \approx 5 \cdot 10^{-1}$  A/cm<sup>2</sup>. In view of the logarithmic accuracy of Eq. (3), this method is rather accurate.

The function  $U(J)/k_B T$  we obtained is shown in Fig. 2. Clearly, two segments of the function  $U(J)/k_B T$  can be identified: the first in which the activation energy depends strongly on the current density (I), and the second with a rather weak dependence (II). These results can be interpreted within the framework of three-dimensional collective creep.<sup>19</sup> According to this theory we have  $U(J) \propto J^{-\mu}$ , with  $\mu = 1/7, 3/2, 7/9$  for creep of individual vortices, weak coupling between vortices, and strong coupling between vortices, respectively.<sup>19</sup> Regions I and II can be described by algebraic functions of the form  $J^{-\mu}$  with  $\mu \approx 0.11$  and  $0.5$  for the first and second segments, respectively. The value of  $\mu$  for segment I is close to the theoretical  $1/7$  (creep of individual vortices). The value of  $\mu$  for segment II is in good agreement with the value we obtained in Ref. 4, where we investigated the functions  $S(T)$  and  $J_c(T)$ . If we estimate the average distances  $\langle a_0 \rangle$  between vortices (since  $\langle a_0 \rangle \approx (\Phi_0 / \langle B \rangle)^{1/2}$ ,  $\langle B \rangle = P_m^{\text{rem}}/V$ ), we find that the kink point in Fig. 2 corresponds to  $\langle a_0 \rangle \approx 500-600$  nm, which is close to the value of the penetration depth for  $\kappa$ -(ET)<sub>2</sub>Xl  $\approx 500-600$  nm.<sup>20-21</sup> It is probable that the observed be-

havior of  $U(J)$  may be connected with the fact that the interaction between vortices decreases as the quantity  $\langle a_0 \rangle$  increases.

#### 4. CONCLUSION

We have proposed a new method for obtaining the function  $U(J)$ , one that requires only a single fitting parameter and the experimentally appropriate measurement time  $t_m$ .

Using this method, we have obtained the function  $U(J)$  for the organic superconductor  $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Cl}_{0.5}\text{Br}_{0.5}$ .

We have established that the function  $U(J)$  has two segments, which may both be described by functions of the form  $J^{-\mu}$ , with  $\mu \approx 0.11$  and  $0.5$  for  $J < J'$  (region I) and  $J > J'$  (region II), respectively. The value of  $\mu$  for region I is close to  $\mu = 1/7$ , as predicted by the cooperative flux creep model for the case of creep of individual vortices. The value of  $\mu$  for segment II is in good agreement with the value we obtained in Ref. 4, where we investigated the temperature dependence of the relaxation rate.

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