High-field fluctuations of a κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br single crystal

V. Vulcanescu, C. Lenoir, P. Batail, and L. Fruchter
CNRS, Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay Cedex, France
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Torque measurements were used to study the magnetization of a κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$] Br single crystal at fields up to 35 kOe close to T_c. A crossing point is found where the magnetization becomes independent of the applied field. The magnetization at this point agrees well with the expression given by Koshelev. The Ginzburg fluctuation parameter is found of the order of 0.025.

The contribution of fluctuations to the magnetization near $H_{c2}(T)$ has now been extensively investigated in the case of the high-T_c superconductors. The large value of the Ginzburg parameter and of the superconducting transition temperature gives birth to a large temperature and field interval in the superconducting phase diagram where critical fluctuations are manifest. In particular, the existence of a crossing point where the magnetization is found independent of the applied field has been investigated for the great majority of the high-T_c superconductors, both experimentally and theoretically. For the organic compounds, thermal fluctuations should also be enhanced by the large anisotropy. However, due to lower T_c values, the temperature interval for critical fluctuations is expected to be reduced strongly in this case. Thus, it is an experimental challenge to investigate the superconducting phase diagram for such compounds. In this paper, we show that the torque technique allows strong magnetic-field measurements inaccessible to the standard magnetization measurements and permits one to evaluate the superconducting phase diagram near $H_{c2}(T)$ for an organic compound.

Two samples were used in our study. They were both grown by the electrochemical technique (route No. 3 in Ref. 1) and issued from the same batch. The smaller crystal, with dimensions $0.67\times0.41\times(e=0.17)$ mm^3 (where e stands for the direction perpendicular to the plane), was used for torque measurements. The larger one, with dimensions $0.79\times0.84\times(e=0.26)$ mm^3 was used for superconducting quantum interference device (SQUID) measurements. Torque measurements were performed using a capacitive method in which the deflection of the capacitance electrode measures the torque on the sample. For highly anisotropic samples, the monitoring of the exact orientation of the sample with respect to the applied field maybe crucial. In order to check that the deflection of the capacitance did not play any role in our case, the measurements at the lower temperatures—involving the largest torque signal and possible deflection—were also performed with a modified capacitive setup in which the rotation of the sample could be nulled down to about $10^{-2\circ}$, using a feedback on a current loop. The data obtained in this way did not show any significant difference with those obtained with the uncompensated setup, indicating negligible deflection of the capacitance in the latter case. Then, as the first of the two methods was more convenient for small signals close to T_c (due to smaller parasitic signals), it was used for all the measurements reported here. The nonlinearity of the sensor was checked to be always negligible (relative capacitance variations were in the range 10^{-2}). Magnetization and ac susceptibility measurements for fields applied along the normal to the planes were performed using a Quantum Design SQUID magnetometer. A quartz rod was used for mounting the sample, so that the small magnetic moment of the sample was always much larger than any parasitic signal from the sample holder. A temperature-independent diamagnetic contribution of the order of $\chi=M/H\approx-10^{-6}$ emu cm^{-3} Oe^{-1} was detected above T_c. Using for the material density[2] 5.04×10^{-4} mol cm^{-3} and summing up the core susceptibilities[3] for the different elements in the chemical formula yields the estimate $\chi_{\rm core}\approx-2\times10^{-7}$ emu cm^{-3} Oe^{-1}, which reasonably accounts for the observed normal-state susceptibility. This contribution was systematically subtracted from the raw dc magnetization data.

As it is important that all measurements are unaffected by pinning, we first located the irreversibility line for our sample, using the onset of the imaginary part of the ac susceptibility data. Dc fields were applied to the sample perpendicular to the layers and the ac susceptibility for a 1 Oe alternative field at frequency 33 Hz was measured. The onset of irreversibility is shown in Fig. 1. The data

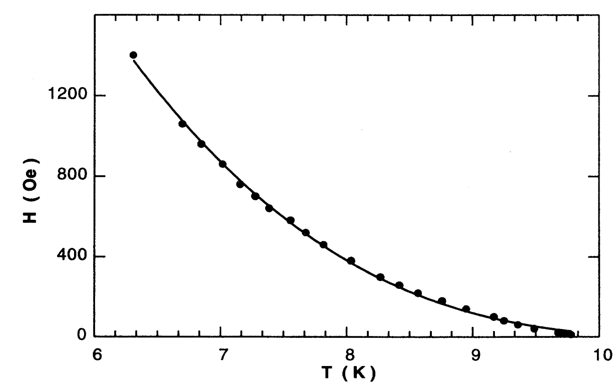

FIG. 1. Irreversibility line as inferred from the onset of the imaginary part of the ac susceptibility. The line is the fit to the thermal melting theory.

were fitted to the thermal melting expression,[4] $H(T) \approx H_m(1-T/T_c)^2$, using $H_m = 9.6$ kOe and $T_c = 10.1$ K. The Lindemann criterion predicts $H_m \approx \beta_m (c_L^4/G_i) H_{c2,\perp}(0)$, where $\beta_m \approx 5$, $c_L \approx 0.3$, $H_{c2,\perp}(0)$ is the transverse upper critical field extrapolated to $T=0$, and G_i is the Ginzburg fluctuation parameter. As underlined by Blatter[5] this parameter does not characterize the width of the fluctuation regime in the case of strongly layered superconductors: it should not be confused with the two-dimensional Ginzburg number used below in the analysis of the critical fluctuations. Using $G_i = [\gamma T_c / H_c^2(0)\xi^3(0)]^2/2$, with $H_c(0) \approx 1.74 H_c(T=0)$ and $\xi(0) = 1/1.36\xi(T=0)$, we find $G_i \approx 0.8$ and $H_{c2,\perp}(T=0) \approx 30$ kOe, in rough agreement with the data in Ref. 17. However, G_i depends upon the penetration depth as λ^4 so that, considering the uncertainty on this parameter, a factor of 2 is not unlikely on both the Ginzburg number and the upper critical field derived in this way. The data shown in Fig. 1 allows for the determination of the reversible domain for the torque measurements: using the two-dimensional approximation [Eq. (2)] justified below, the reversible domain (T,H) for torque can be found above the line in Fig. 1, when the field in this diagram is set as the component of the applied field transverse to the plane direction. We find that, above $T=9$ K and for fields greater than 5 kOe, the signal should not be affected by pinning for angles larger than about 2° from the plane direction.

The magnetization, after subtraction of the normal-state contribution, is shown in Fig. 2. Due to the increasing normal-state diamagnetic contribution with field, measurements were performed for fields less than 5 kOe only, in order to limit the uncertainty on the base line. A close inspection of the data (Fig. 2, inset) reveals the existence of the so-called crossing point where the magnetization, $M^*(T^*)$, is found independent of the applied field,[6,7] with $M^* \approx -4 \times 10^{-3}$ emu cm^{-3} and $T^* \approx 10.6$ K. The precision of the measurements and the magnitude of the effect do not allow for rescaling of the magnetization curves, as was done for high-T_c superconductors

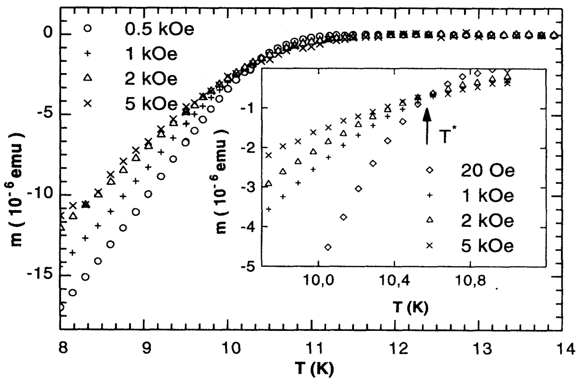

FIG. 2. Magnetization curves for field applied perpendicularly to the plane direction, using the SQUID magnetometer.

in Refs. 6, 8, and 9. We notice that the normal-state magnetization reaches about $0.5M^*$ at 5 kOe. Two different theoretical expressions have been given for the magnetization at the crossing point, in the case of two-dimensional (2D) materials. Bulaevskii, Ledvij, and Kogan,[10] considering the vortex-lattice state and including fluctuations at $H \ll H_{c2}$, obtained $M^* \approx T^*/s\phi_0 \ln(\eta\alpha/\sqrt{e})$, where $\alpha \approx 1$. Tesanovic[6] gave a similar expression, within the critical fluctuation regime, but not including the logarithmic term. Finally, Koshelev[11] argued that the contribution of the weak fluctuations should also be taken into account in addition to the critical ones, yielding $M^* = 0.346 T^*/s\phi_0$. Using this expression, we give in Table I the estimate for s found from the data in the literature. For the most anisotropic superconductors (Bi 2:2:1:2; Bi 2:2:2:3; Tl 2:2:2:3), the value obtained in this way is in reasonable agreement with the structural data (although, there is a tendency to underestimate this parameter). As noticed in Ref. 11, the use of Tesanovic's expression tends to overestimate the value for s, which has often been corrected in the analysis of experimental data by invoking some superconducting fraction less than one. Such an overestimation is found however for the

TABLE I. Selected values from the literature for the temperature, the magnetization at the crossing point, the separation of the coupled planes from structural data and the one inferred from Koshelev's result.

Compound	T^* (K)	$-M^*$ (emu cm^{-3})	s (structural) (Å)	s (calculated) (Å)	Reference
$Bi_2Sr_2CaCu_2O_8$	88.3	0.3	15	7	21
	80	0.25		8	22
	87	0.1		20	23
$Bi_2Sr_2Ca_2Cu_3O_{10}$	108.1	0.21	18	12	24
$YBa_2Cu_3O_{7-\delta}$	91.2	8×10^{-2}	12	27	8
$(La_{1-x}Sr_x)_2CuO_4$			13		
$x=0.046$		0.1		7	25
$x=0.059$		5×10^{-2}		14	
$x=0.077$	32.5	4×10^{-2}		19	
$x=0.090$	30	1.4×10^{-2}		50	
$Tl_2Ca_2Ba_2Cu_3O_{10+\delta}$	121	0.29	18	10	26
$HgBa_2CuO_{4+\delta}$	92.4	0.13	9.5	17	25
κ-(BEDT-ITF)$_2$ Cu[N(CN)$_2$]Br	11±0.3	0.02	15	13	This work

less anisotropic compounds in Table I (Y 1:2:3; Hg 1:2:1; La 2:1:2), using Koshelev's result also. It should be noticed at this point that the two-dimensional model is expected to be valid only when the transverse coherence length, $\xi_\perp(T^*)$, is smaller than the interlayer spacing, s: this might provide an explanation for the discrepancy observed for the less anisotropic compounds. In our case, taking $\gamma \approx 30$ and $H_{c2,\perp}(0) \approx 150$ kOe yields the condition $1-T^*/T_{c0} > 10^{-2}$. The subtraction of a large normal-state susceptibility—as compared to M^*—may cause a substantial error in the determination of this quantity and limits the magnitude of the applied field. The torque data is less sensitive to such an effect: only the anisotropic part of the normal-state susceptibility contributes to the signal. The study of the transverse magnetization has been widely used to infer the ratio of the effective masses for anisotropic superconductors, $\gamma=(m_c/m_{ab})^{1/2}$, within the anisotropic Ginzburg-Landau (AGL) model, since the early work of Kogan[12] and Farrell *et al.*[13] As pointed out by Martinez *et al.*, the determination of γ in this way can be invalidated for strongly anisotropic superconductors.[14] In a more general way, the usefulness of the expression for torque as given in Ref. 12 is limited to the AGL model, for fields $H_{c1} \ll H \ll H_{c2}$. For fields close to H_{c2} or for a strong contribution of the fluctuations to the reversible magnetization, this expression should be revised. As pointed out by Hao and Clem[15] and by Blatter, Geshkenbein, and Larkin,[16] a more general approach (scaling) can be used to analyze the angular dependence of the reversible magnetization, provided that the London model can be applied. Using the results of Refs. 15 and 16, the torque per unit volume in the London regime can be expressed as

$$\Gamma/V=\frac{H\sin(2\theta)(\gamma^2-1)}{2\varepsilon(\theta)\gamma}\tilde{M}[H\varepsilon(\theta)/\gamma]\ ,$$
$$\varepsilon(\theta)=[\sin^2(\theta)+\gamma^2\cos^2(\theta)]^{1/2}\ , \tag{1}$$

where $\tilde{M}(H)$ is the magnetization for the isotropic superconductor with superconducting parameters $\lambda_\parallel$ and $\xi_\parallel$ (where $\parallel$ stands for the plane direction), and θ is the angle between the applied field and the normal to the plane. As pointed out in Ref. 16, the regime of applicability is not restricted to temperatures close to T_c. The scaling approach should also be valid for layered superconductor, as long as the discreteness of the structure remains unimportant. An important limiting case for Eq. (1) is the one for a layered superconductor in the decoupled limit. Setting $\gamma=\infty$, yields the 2D scaling form, as used in Ref. 14:

$$\frac{\Gamma/V}{H_\parallel}=M_\perp(H_\perp)\ , \tag{2}$$

where $H_\parallel$ and $H_\perp$ are the components of the applied field, respectively, parallel and perpendicular to the layers and $M_\perp(H_\perp)$ is the perpendicular magnetization in response to the normal field component. An estimate for the anisotropy parameter, based on torque measurements at $T=4.2$ K and $H=30$ kOe—using the Ginzburg-Landau expression for $\tilde{M}(H)$—has been given in Ref. 17 for the material studied here: $\gamma \approx 30$. A similar value ($\gamma \approx 25$) was inferred in Ref. 18 from surface impedance measurements. We have checked that the two-dimensional expression is valid in our case within a good approximation. This is shown in Fig. 3 where the torque signal, once normalized by the parallel component of the field, is found to be dependent only on the transverse component of the field. Then, assuming a quasi-two-dimensional behavior, we can use the torque data (taken at $\theta=25°$) and Eq. (2) to infer $M_\perp(H)$ at fields up to 35 kOe. The result is shown in Fig. 4, for different temperatures, where we have subtracted the contribution of the anisotropic part of the normal-state susceptibility (roughly 20% of the smallest signal shown in Fig. 4). The existence of the crossing point at $T^* \approx 11$ K is visible, where the magnetization at high field tends toward a constant. The asymptotic value for M^* found in this way, $M^* \approx 2\times10^{-2}$ emu cm^{-3}, is larger than the one inferred from the SQUID data. Using Koshelev's result, we find $s \approx 13$ Å, which is reasonably close to the crystallographic data. Then, it is natural to attempt to fit the torque data, using the results in Ref. 11 including both the critical and weak fluctuations contributions to the magnetization [Eqs. (31)–(33) in Ref. 11]. Using the above value for s, there are three parameters left for such a fit: T_{c0}, the mean-field transition temperature at zero field, $(dH_{c2,\perp}/dT)_{Tc0}$ and τ_f, the critical fluctuations parameter. As shown in Fig. 5, a

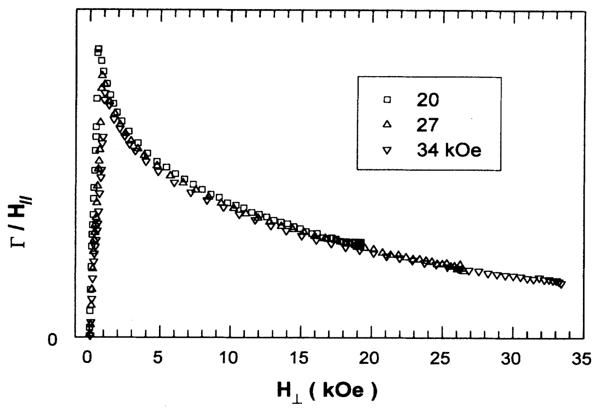

FIG. 3. Torque curves rescaled according to Eq. (2) ($T=9.5$ K).

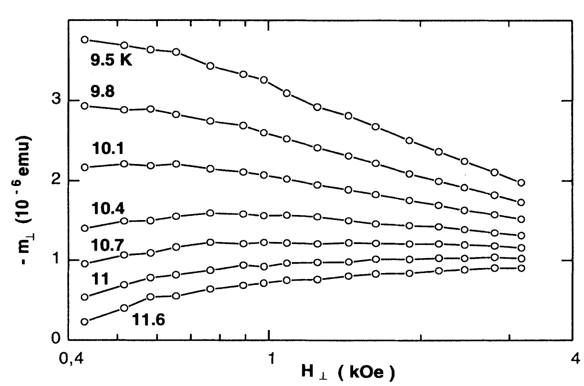

FIG. 4. Magnetization curves inferred from torque data at $\theta=25°$, using Eq. (2).

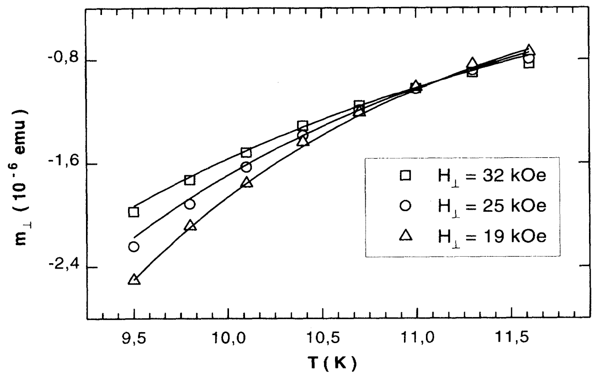

FIG. 5. Magnetization curves inferred from torque data at $\theta=25°$. Lines are the fit to the critical and weak fluctuations contributions, as given in Ref. 11.

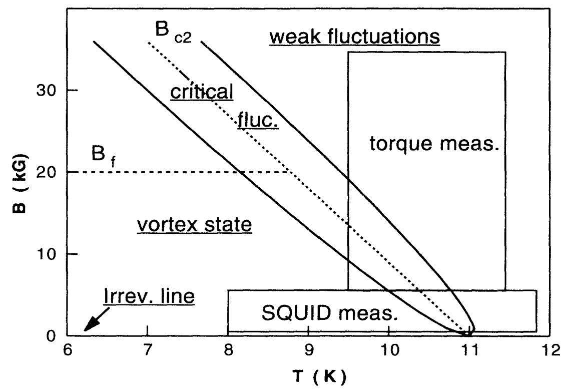

FIG. 6. Phase diagram, as derived from the analysis of the fluctuations magnetization.

reasonable agreement is found with our data, taking $T_{c0}=11.0$ K, $(dH_{c2,\perp}/dT)_{T_{c0}}=9$ kOe/K, and $\tau_f=0.025$. The slope of the upper critical field found in this way is smaller by a factor 2 than the one inferred by us in Ref. 17 and in Ref. 19 from dc magnetization measurements. In Ref. 17, the upper critical field was estimated at 4.2 K, i.e., far below T_c. In Ref. 19, a sharp decrease for $dH_{c2,\perp}/dT$ was observed at $T>0.9T_c$. This observation, associated to a decrease of the anisotropy in the critical fields from $\gamma\approx 80$ at low temperature to $\gamma\approx 13$ near T_c, was interpreted in this work as a dimensional crossover of the compound. If such a crossover does occur near T_c, it could explain that our value for the upper critical derivative is lower than the ones inferred from low-temperature measurements. However, we underline that our fit does include the contribution of fluctuations to the magnetization, while the simple derivation of the upper critical field from a linear extrapolation of the magnetization to zero (London limit), as was done in Ref. 19, can be impaired when fluctuations are present. This is the case for our field range and the upper critical field cannot be inferred from the data in Fig. 5 in this way. The critical fluctuation parameter is comparable to the one found in Ref. 6 for the high-T_c compound Bi 2:2:2:3. From this value for τ_f, one should have $T_{c0}-T^*\approx 0.3$ K. It is not possible to check with such accuracy the exact value for T^* from the data in Fig. 4, but the data for both dc magnetization and torque are compatible with such a value. Using the following estimate: $\tau_f\approx T_{c0}/\varepsilon_0(0)s$, where $\varepsilon_0(0)=[\phi_0/4\pi\lambda_{\parallel}(0)]^2$, yields $\lambda_{\parallel}(0)\approx 2500$ Å. This value is considerably smaller than the value inferred from low-temperature torque measurements on the same compound[17] ($\lambda_{\parallel}(0)\approx 4000$ Å) and from low-field dc magnetization measurements on a parent compound[20] [$\lambda_{\parallel}(0)\approx 5300$ Å]. Given the estimates obtained from the fit, it is possible to schematize the phase diagram for our compound. This is shown in Fig. 6. The critical reduced temperature width is given by $|T_{\rm crit}-T_c(B)|/T_c(B)\approx[\tau_f B/T_{c0}(dH_{c2}/ubT)_{T_{c0}}]^{1/2}$. The fluctuation field, $B_f\approx T_{c0}\tau_f(dH_{c2}/dT)_{T_{c0}}$, delimits the region above which the lowest Landau level only exhibits strong fluctuations (lower field for the validity of our fit in Fig. 5), which justifies the fit in Fig. 5 *a posteriori.*

In summary, we have used torque measurements to infer the magnetization of an organic compound at high field, using the quasi-two-dimensional approximation for the angular dependence of the magnetization. This technique allows one to avoid most of a large normal-state diamagnetism which limits the accessible field range in the case of standard magnetization measurements. We were able to detect the existence of a crossing point where the magnetization at high field is found independent of the applied field. The magnetization at the crossing point agrees with the predicted value reasonably. However, the penetration depth inferred from the Ginzburg parameter, $\tau_f\approx 0.025$, is smaller than expected from other measurements. The temperature derivative of the upper critical field at this point is found smaller than previous low-temperature determination, which could be due to a dimensional crossover near T_c.

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