Investigation of Vortex Behavior in the Organic Superconductor κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ Using Muon Spin Rotation

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(Received 14 January 1997)

Muon spin rotation (μSR) measurements have been performed on the organic superconductor κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ in order to investigate its exotic vortex behavior. Previously unobserved features of the μSR line shapes have been measured at low fields and temperatures. In the mixed state the existence of a lattice composed of linear vortices is demonstrated at low fields. The breakup of this well-ordered lattice has furthermore been observed as a function of both field and temperature. [S0031-9007(97)03800-3]

PACS numbers: 74.70.Kn, 74.25.Dw, 74.60.Ge, 76.75.+i

Charge transfer salts of the organic molecule BEDT-TTF [*bis(ethylenedithio)tetrathiafulvalene*] exhibit a wide range of properties, and the low temperature ground states include antiferromagnetic metallic conductors and superconductors [1]. This Letter concerns the vortex behavior in the superconducting salt κ-(BEDT-TTF)$_2$Cu(SCN)$_2$, (ET-SCN), which has a superconducting transition onset $T_c \approx$ 10.4 K. The quasi-2D Fermi surface topology of ET-SCN leads to highly anisotropic superconducting properties, so that in many ways it resembles some of the high-T_c compounds such as $Bi_{2.15}Sr_{1.85}CaCu_2O_{8+\delta}$ (BSCCO). The flux vortex arrangements in the mixed state of these other extremely anisotropic superconductors have recently been the subject of a great deal of discussion in the literature [2–6]. Compared with the high-T_c materials, ET-SCN has a significantly smaller T_c and a substantially longer in-plane penetration depth $\lambda_\parallel \sim 500$ nm, which might be expected to lead to significant differences in the low field phase diagram [3,7,8].

There have been several previous studies of ET-SCN using muon spin rotation (μSR) which have concentrated mainly on determining the value and temperature dependence of the in-plane superconducting penetration depth $\lambda_\parallel(T)$ [9–11]. A precise knowledge of $\lambda_\parallel(T)$ can give important information concerning the strength and symmetry of the superconducting pairing. For materials having an Abrikosov lattice composed of rigid, static vortex lines, the temperature dependence of the μSR linewidth is closely related to $\lambda_\parallel(T)$. Here we present μSR measurements which clearly demonstrate the existence in ET-SCN of a flux-line lattice at low fields, which has the characteristic field distribution expected from the presence of straight *vortex lines*. This is in contrast to previous studies which concentrated only on the high-field regime [9–11]. At high fields the probability distributions indicate, as we shall discuss further below, a quasi-two dimensional order, with reduced correlations of vortex segments along the field direction. By studying the evolution of the μSR line shapes with field and temperature, we are able to observe the breakdown of 3D order in the system. The measured dimensional crossover field $B_{\rm cr}(T)$ and the thermal breakup temperature T_b are in good agreement with expectations for extremely anisotropic systems [3,7,8,12].

Recent ac-susceptibility measurements on ET-SCN yielded an estimate of the superconducting anisotropy parameter $\gamma = \lambda_\perp/\lambda_\parallel \approx 160$–$350$, where $\lambda_\perp$ and $\lambda_\parallel$ are the superconducting penetration depths for currents flowing perpendicular and parallel, respectively, to the superconducting planes in a uniaxial system [13]. This estimate is comparable with values of γ obtained for the high-T_c material BSCCO [8], which exhibits an extremely rich magnetic phase diagram, largely as a result of its very large anisotropy [3–5,14,15]. In contrast to the rigid flux lines found in conventional isotropic superconductors, those in extremely anisotropic materials are very flexible. Consequently, the latter are highly susceptible to thermally induced and pinning-induced disorder [4,5,14,15]. In the extreme case of $\gamma = \infty$, a situation which may be realized in superconducting multilayer systems, an appropriate model is one based on quasi-2D vortices ("pancake" vortices) confined to the superconducting planes [12]. For systems of large but finite γ, such as BSCCO or ET-SCN, it can be instructive to think of the vortex lines as being composed of strings of weakly coupled pancake vortices [5]. For extremely anisotropic materials these may be coupled mainly by electromagnetic interactions, whereas for less anisotropic materials additional Josephson tunneling currents between the layers may play a more significant role [3,8]. The degree of flexibility of the vortex lines very much depends on the nature and strength of the coupling, as well as the temperature and the applied magnetic field. While at low fields, pancake vortices may couple to form fairly rigid linelike vortices; at higher fields the vortices in a given layer begin to overlap and interact more strongly. In this case the cost of in-plane shear deformations of the lattice is greater than that for short wavelength tilt deformations. When, in a Josephson coupled system, the wavelength of such tilt deformations becomes of the order of the separation of the superconducting planes s, the model reduces to one of quasi-independent 2D lattices, which occurs above a characteristic crossover field $B_{2D} \sim \phi_0/(s\gamma)^2$ (where $\phi_0 = h/2e$ is the flux quantum) [5]. A similar crossover behavior has been directly observed in BSCCO using μSR and other techniques [4,14,15].

The μSR experiments were carried out at the ISIS facility at the Rutherford Appleton Laboratory, UK, which provides pulsed beams of positive muons at a frequency of 50 Hz with a width of $\approx$70 ns. These muons, which are approximately 100% spin polarized antiparallel to their momentum ($\approx$29 MeV/c), are implanted into a mosaic of high-quality ET-SCN single crystals grown by an electrochemical oxidation method. The individual crystals are platelets of typical dimensions $1.0 \times 1.5 \times 0.5$ mm^3 with the largest face parallel to the superconducting planes ($\hat{b}$-$\hat{c}$ planes). The crystals were mounted with Apiezon grease on a plate of hematite (Fe_2O_3) to give a total area approximately 10×20 mm^2. The use of a hematite backing plate ensures that any muons not stopping in the sample are rapidly depolarized outside of the μSR time window, so that the resulting spectra contain information relating only to the sample. The experiments were conducted in the transverse-field geometry where the initial muon spin is perpendicular to the applied magnetic field. The sample was arranged so that the superconducting planes made an angle of 45° with both the field and with the muon momentum. The polarization of the muons at ISIS cannot be rotated, so this geometry was chosen to provide a reasonable cross-sectional area of sample to the beam, while also allowing the field to be directed at a large angle to the superconducting planes. The muons are assumed to stop at random positions within the vortex lattice, and, consequently, the spatial distribution of internal fields $B(r)$ leads to a distribution of muon precession frequencies characterized by a probability distribution $p(B)$ [9]. Hence, a measurement of $p(B)$ by μSR reveals important information regarding the vortex arrangements and dynamics in the superconducting sample [4,7–9,14].

Figure 1(a) is a μSR line shape measured at 1.8 K for a sample cooled in a field of 2.5 mT, which was derived from the muon time spectra using a maximum entropy technique [4,7,8,14]. The inset curve in Fig. 1(a) is the probability distribution $p(B)$ from a numerical simulation of a vortex-line lattice in a uniaxial superconductor at an angle of 45° to the superconducting planes, using a penetration depth $\lambda_{\parallel} = 4600$ Å. Such a $p(B)$ has several

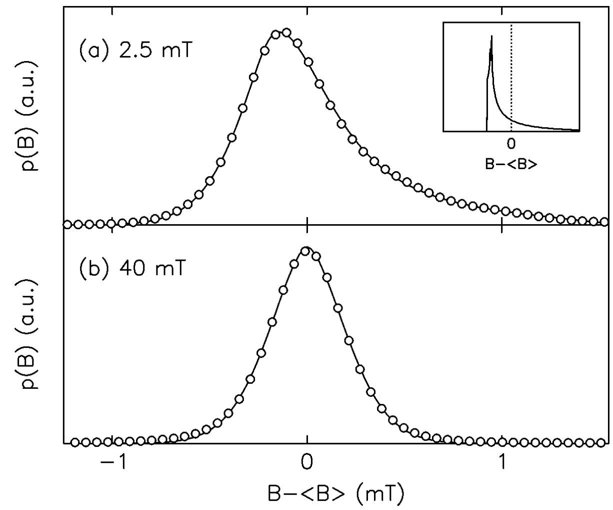

FIG. 1. Circles represent the measured μSR line shape for ET-SCN when field cooled to 1.8 K in a field applied at 45° to the superconducting planes. (a) At 2.5 mT the characteristic asymmetric vortex-line-lattice line shape is seen; the solid line is derived from a numerical simulation of an ideal 45° vortex-line lattice using a penetration depth $\lambda_{\parallel} = 430(4)$ nm to describe the data. The simulation has been convoluted with a Gaussian of rms width 0.23 mT, which represents the instrumental and dipolar broadening. The inset shows the line shape before convolution (see text). (b) At 40 mT a symmetric line shape is obtained which cannot be described using an ideal vortex-line lattice model [inset of (a)]. This is indicative of a more 2D arrangement of pancake vortices (see text); the data can be represented by a symmetrical vortex field distribution of rms width 0.06 mT broadened by 0.23 mT (solid line).

characteristic features, including the highly asymmetric shape arising from the long "tail" at high fields, which corresponds to regions of the lattice close to the vortex cores. The mode of the distribution occurs at B_{pk}, which lies below the mean field $\langle B \rangle$. The solid curve in the main part of Fig. 1(a) is the convolution of this asymmetric field distribution with a symmetrical Gaussian of rms width 0.23 mT, representing the instrumental and dipolar broadening. This describes the measured data well. It is important in this system to correctly allow for this latter contribution, since it is comparable to that arising from the superconductivity for $T < T_c$, due to the long penetration depth. The ISIS MuSR spectrometer is ideal for these types of measurements, since the very long time window available at a pulsed source reduces the contribution to the line shapes from instrumental broadening. Curves such as Fig. 1(a) are the first highly asymmetric μSR line shapes to be observed in ET-SCN, and the first confirmation of the existence of a *vortex-line* lattice at any applied field in this material. Figure 1(b) shows the μSR line shape taken for the same sample cooled in a field of 40 mT. This is very similar to previously published line shapes in ET-SCN [9–11], being highly symmetric, with a mode at B_{pk} close to the average field $\langle B \rangle$. This change of line shape with field is very similar to changes observed in the high-T_c

superconductor BSCCO, and indicates the loss of short-range correlations of the pancake vortices along the field direction [14,15]. It is attributed to the effective smearing out of the core fields due to the local tilt deformations of the pancake stacks.

The penetration depth may be extracted from the μSR data since features of the line shape scale with $\lambda_{\parallel}(0)^{-2}$ [14,16]. The model lattice which best describes the present data yields $\lambda_{\parallel} = 430(4)$ nm [Fig. 1(a)], assuming the usual angular scaling [17]. While this is somewhat smaller than previously published results [9–11], it must be remembered that the broader line shapes presented here at low fields are considerably closer in shape to those expected for an ideal lattice, and consequently lead to a more accurate assessment of $\lambda_{\parallel}$. We have also performed μSR measurements at PSI, Switzerland, using the alternative geometry with B perpendicular to the planes, which cannot be performed at ISIS [16]. From these measurements we estimated a value of 650 nm, which reduces to $\lambda_{\parallel} = 538(8)$ nm after full correction for dipolar and instrumental broadening. While exhibiting similar features, these data are not so well suited to the detailed analysis of the very narrow line shapes, due to the relatively short time window at PSI. However, it carries the advantage that the analysis for this geometry does not require any assumptions concerning the angular scaling, and so provides a more direct estimate of $\lambda_{\parallel}$. The discrepancy between the two might arise from deviations from the expected angular scaling which can occur in highly anisotropic systems, which are attributed to the effects of pinning-induced disorder which can affect the angular dependence of the field distribution [17–19]. The measurement is in good agreement with the value estimated from reversible magnetization $\lambda_{\parallel} = 535(20)$ nm [20].

It is useful to quantify the line shape by a dimensionless skewness parameter β. For the very narrow line shapes which occur in ET-SCN an appropriate expression is $\beta = (\langle B \rangle - B_{pk})/(\langle B^2 \rangle - \langle B \rangle^2)^{1/2}$. A value of $\beta = 0$ indicates a symmetric line shape, while a positive value reflects a weighting toward fields higher than B_{pk}, as is the case for a line shape arising from a vortex line lattice [Fig. 1(a)]. Figure 2(a) shows β (after correction for the nonsuperconducting contributions to the linewidth) as a function of applied field. This broadening was determined by measurements on the sample in the normal state ($T > T_c$), where there is no contribution to the line shape from the superconductivity. There is a broad crossover centered around $B_{\mathrm{cr},45^\circ} \sim 7$ mT going from the asymmetric line shape of Fig. 1(a) to the almost symmetric line shape of Fig. 1(b).

For a Josephson coupled superconductor ($\lambda_{\parallel} \gg \gamma s$) the dimensional crossover is expected at a field $B_{2\mathrm{D}} \sim \phi_0/(\gamma s)^2$. Taking the layer separation for ET-SCN as $s \sim 1.6$ nm together with recent estimates of $\gamma \sim 160$–350 [13] yields an estimate of $B_{2\mathrm{D}} \sim 7$–30 mT. It has recently been shown that for systems which obey $\lambda_{\parallel}(0) \lesssim$

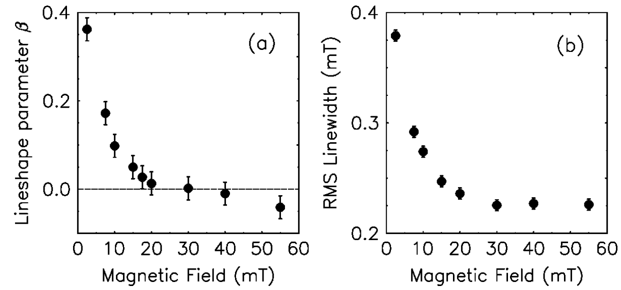

FIG. 2. (a) The skewness parameter β (see text) of the superconducting line shape, after field cooling to 1.8 K in a field applied at 45° to the superconducting planes. The values of β were calculated on line shapes which have been deconvoluted from the nonsuperconducting contributions. The β value observed at low fields is characteristic of a vortex-line lattice (asymmetric line shape), the disappearance of which is indicated by the fall of β above $B_{\mathrm{cr},45^\circ} \sim 7$ mT, leading to a fully symmetric line shape above $\sim$20 mT. (b) The linewidth $\langle \Delta B^2 \rangle^{1/2}$ as a function of field, where $\langle \Delta B^2 \rangle$ is the second moment of the field distribution. The linewidths have not been corrected for broadening effects, to allow comparison with previously published results. All line shapes were measured after cooling the sample from above T_c to 1.8 K in the applied field.

γs, a better estimate of the crossover field is given by $B_{\mathrm{cr}} \sim B_{\lambda} = \phi_0/\lambda_{\parallel}^2$ [7,8]. We may thus estimate $B_{\lambda} \sim 7$ mT. It is therefore not clear from the present data whether the experimentally determined $B_{\mathrm{cr}} \sim 7$ mT is best identified with $B_{2\mathrm{D}}$ or B_{λ}.

For a rigid vortex-line lattice the temperature dependence of the linewidth $\langle \Delta B^2 \rangle^{1/2}(T) \propto 1/\lambda^2$, where λ is the effective penetration depth in a plane perpendicular to the applied field and $\langle \Delta B^2 \rangle$ is the second moment of the field distribution. However, for unconventional flexible or quasi-2D lattices the $\langle \Delta B^2 \rangle^{1/2}(T)$ may be influenced by other temperature-dependent effects on the lattice structure, such as thermally induced fluctuations and vortex-lattice melting [4,9,11,14]. Figure 3(a) shows $\langle \Delta B^2 \rangle^{1/2}(T)$ for fields above and below B_{cr}. For $B > B_{\mathrm{cr}}$, $\langle \Delta B^2 \rangle^{1/2}(T)$ is not inconsistent with that expected for a conventional s-wave superconductor, as also found in Ref. [11]. However, for $B < B_{\mathrm{cr}}$ there is a dramatic increase in linewidth below $T^* \sim 5$ K. More significantly, Fig. 3(b) shows that the *line shape* also changes around T^*. While for $T < T^*$ the value of β approaches that expected for an ideal vortex-line lattice, this value falls rapidly with T and plateaus at a significantly reduced value. This reduction in β, reflecting a change in line *shape,* indicates a reduction of pancake vortex correlations along the field direction. Clem [12] has analyzed the behavior of electromagnetically coupled pancake stacks in the dilute limit, and found that the characteristic temperature for the thermally induced breakup of such a stack is given by

$$T_b = \frac{\phi_0^2 s}{k_B \mu_0 (4\pi)^2 2 \lambda_{\parallel}^2} \approx 4.5\ \mathrm{K}. \qquad (1)$$

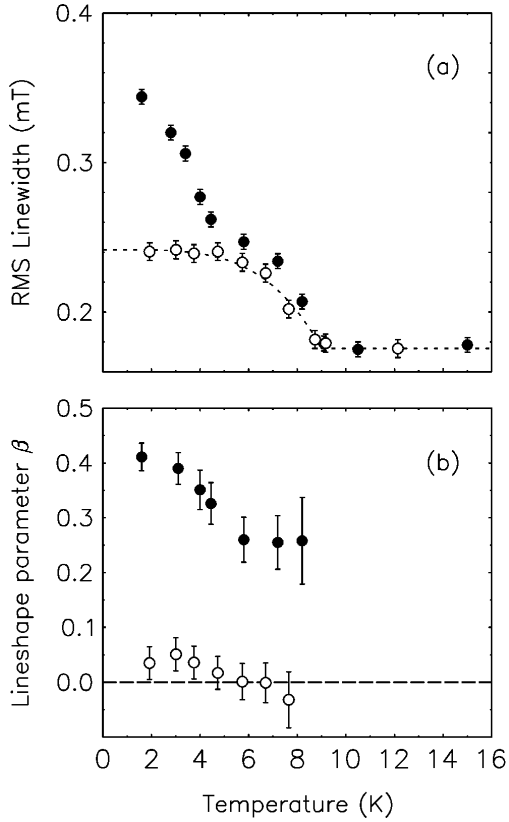

FIG. 3. The temperature dependence of (a) the total μSR rms linewidth $\langle \Delta B^2 \rangle^{1/2}$ and (b) the skewness β, for applied fields of 2.5 mT (solid circles) and 30 mT (empty circles), after field cooling to 1.8 K in a field applied at 45° to the superconducting planes. At 30 mT, which is above B_{cr}, the temperature dependence of the linewidth is consistent with a two-fluid-like dependence of λ (dashed line) with the narrow linewidth expected for a quasi-2D flux arrangement. For 2.5 mT, which is below B_{cr}, the line shape rapidly becomes narrower between 3 and 5 K, and above 5 K it follows a similar dependence to that seen above B_{cr}. At 2.5 mT (solid circles) the narrowing of the linewidth is accompanied by a significant fall in β, centered around a crossover temperature $T^* \sim 5$ K, which indicates a reduction of pancake vortex correlations along the field direction above T^*.

The reduction in the positional correlations of pancake vortices at $T^* \sim 5$ K is thus consistent with the predicted breakup of vortex lines at T_b.

In conclusion, we have provided the first clear evidence for the existence of a vortex-line lattice in ET-SCN, which is found at low fields and temperatures as expected. There exists a crossover to a quasi-2D lattice at $B_{\mathrm{cr},45^\circ} \sim 7$ mT. At low fields a further crossover occurs with increasing temperature at $T_b \sim 5$ K. This is consistent with estimates for the thermally induced breakup of a vortex line composed of weakly coupled pancake vortices [12].

We thank the staff of the ISIS MuSR facility for assistance. We also acknowledge the financial support of the EPSRC of the United Kingdom and the Swiss National Science Foundation.

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