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## Angle-dependent vortex structure in a high anisotropy superconductor

F.L. Pratt<sup>a,\*</sup>, I.M. Marshall<sup>b</sup>, S.J. Blundell<sup>b</sup>, A. Drew<sup>c</sup>, S.L. Lee<sup>c</sup>, F.Y. Ogrin<sup>c</sup>,  
N. Toyota<sup>d</sup>, I. Watanabe<sup>e</sup>

<sup>a</sup>ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, UK

<sup>b</sup>Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, UK

<sup>c</sup>School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY16 9SS, UK

<sup>d</sup>Department of Physics, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan

<sup>e</sup>RIKEN-RAL, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, UK

### Abstract

Angle-dependent muon spin rotation measurements have been made on the organic superconductor  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(SCN)<sub>2</sub>. Oscillations are observed in the width of the internal field distribution, which are periodic in the perpendicular component of the applied magnetic field,  $B_z = B \cos \theta$ , with a uniform period over a range of angles and fields. These oscillations are superimposed on the standard  $\cos \theta$  scaling expected for the width in a highly anisotropic superconductor. The oscillation period is of order 2 mT and the amplitude is particularly strong at fields comparable with the period. The origin of this novel phenomenon is discussed in terms of the low field instabilities of tilted vortices in this highly anisotropic superconductor.

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When a magnetic field is applied at an angle to the normal to the layers in an anisotropic superconductor, the vortex structure can become strongly modified by anisotropic interactions. In low fields, vortex lattice shear modes [1] produce regions of instability [2]. The unstable tilted vortex lattice can then be replaced by a combined lattice,

consisting of an Abrikosov vortex array perpendicular to the layers, combined with a Josephson vortex array parallel to the layers [3]; the stability region for the combined lattice has been investigated for the London model [4] and for the pancake vortex model [5]. Recently, various distortions of the combined lattice structure were found to reduce the free energy still further [6], suggesting that the details of the actual vortex structures in these high anisotropy systems may be extremely complex.

A further factor governing vortex structure is the phenomenon of vortex attraction, which was

\*Corresponding author. Muon Science Laboratory, The Institute of Physical and Chemical Research (RIKEN) Chilton, Didcot OX11 0QX, UK. Tel.: +44-1235-445135; fax: +44-1235-445720.

E-mail address: [f.pratt@rl.ac.uk](mailto:f.pratt@rl.ac.uk) (F.L. Pratt).

predicted for anisotropic superconductors in tilted fields [7], leading to non-uniform chain-like vortex structures [8]. These chains would be expected to disappear at fields well above  $H_{c1}$  [9]. Such chains were observed using flux decoration techniques in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  [10]. Similar decoration experiments on the higher anisotropy system  $\text{Bi}_{2.1}\text{Sr}_{1.9}\text{Ca}_{0.9}\text{Cu}_2\text{O}_{8+\delta}$  (BSCCO) showed mixed-phase behaviour, with chains embedded in a more uniform flux lattice [11]. Interaction between the components of the combined lattice has been suggested to be important in this case, providing a second mechanism for chain formation [12]. Subsequent decoration studies on BSCCO suggested two non-orthogonal vortex phases with different tilt angles to the applied field [13]. Having two stable vortex orientations for a particular angle of applied field had previously been shown to be theoretically possible when the anisotropy of the vortex core was taken into account [14].

The organic superconductor  $\kappa$ -(BEDT-TTF) $_2\text{Cu}(\text{SCN})_2$  (ETSCN), has  $T_c \sim 10$  K and a superconducting anisotropy comparable to that of BSCCO. Previous  $\mu\text{SR}$  work [16] has shown that a static flux line lattice exists in this system at temperatures well below  $T_c$  and fields well below  $H_{c2}$ . For fields oriented perpendicular to the layers both  $\mu\text{SR}$  [16] and flux decoration studies [17] have shown that a triangular Abrikosov vortex structure is present for fields down to well below 1 mT. We report here detailed angle-dependent studies of the internal field distribution in ETSCN using  $\mu\text{SR}$  in which we have observed an oscillatory instability in the flux arrangement. The oscillation is manifested through the second moment of the field distribution and is periodic in the perpendicular component of the applied magnetic field.

Crystals of ETSCN were prepared electrochemically and were fully deuterated to reduce the relative contribution to the linewidth from nuclear dipoles compared to the contribution from the vortex structure. The sample consisted of a mosaic of  $\sim 150$  mg of crystals aligned with their highly conducting planes parallel. The orientation of the field with respect to the sample was varied using two perpendicular pairs of Helmholtz coils. By placing the sample plane at  $45^\circ$  to the incoming

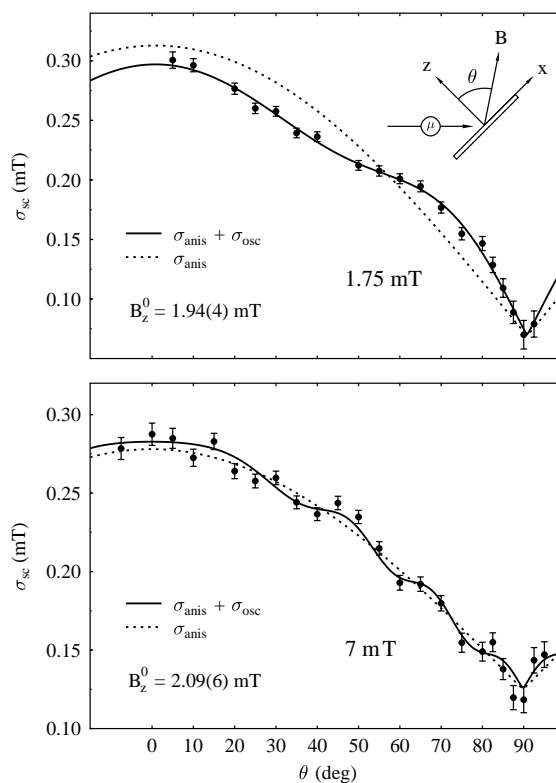


Fig. 1. The angular dependence of  $\sigma_{\text{SC}}$  in ETSCN at 1.8 K for two fields. The lines are the fits discussed in the text. The inset shows the  $45^\circ$  sample geometry used for the experiments: the incoming muon beam and its spin polarization are horizontal and the field direction is varied between the  $z$  and the  $x$  axes.

muon beam, the field orientation could be varied from  $0^\circ$  to  $90^\circ$  while maintaining sufficient muon spin rotation amplitude for the measurement (Fig. 1 inset). This arrangement allowed the sample to maintain a constant cross-section to the muon beam during the angular scan, contributing to the accuracy and stability of the measurement by eliminating any angle-dependent non-sample fraction in the measured asymmetry.

Measurements were carried out on the ARGUS spectrometer of the RIKEN-RAL Muon Facility at ISIS and the GPS spectrometer at PSI. For each point at a specific field and angle, the amplitude and phase of the transverse oscillatory component of the muon polarization was calibrated in the normal state at 12 K for each detector group. The sample was then field cooled to 1.8 K and the

increase in the Gaussian damping rate was recorded, providing a measurement of the superconducting contribution to the RMS width of the internal field distribution  $\sigma_{SC}$ .

The superconducting anisotropy  $\gamma$  is defined as the ratio of the out-of-plane to in-plane penetration depths, i.e.  $\gamma = \lambda_{\perp}/\lambda_{\parallel}$ . For this sample, detailed studies of the vortex phase diagram [18] suggest that  $\gamma \sim 100$ . Fig. 1 shows measurements of the angular dependence of  $\sigma_{SC}$  at two different field values. From anisotropic London theory, the linewidth is expected to vary as

$$\sigma_{anis} = \sigma_0(\cos^2 \theta + \sin^2 \theta/\gamma^2)^{1/2} + \sigma_c, \quad (1)$$

where  $\theta$  is the angle of the applied magnetic field with respect to the normal to the sample layers,  $\sigma_0$  is the vortex contribution at normal orientation and  $\sigma_c$  represents angle-independent sources of broadening in the superconducting state, that are unrelated to the vortex structure. For the high anisotropy system studied here, this dependence closely follows a  $\cos \theta$  form, except very near  $90^\circ$ . Attempts to fit the data using Eq. (1) are shown as dotted curves in Fig. 1, where it can be seen that although the anisotropic London expression describes the general behaviour, there is a marked oscillatory variation superimposed on it. In order to adequately describe the angular dependence of the linewidth we must add an oscillatory term to Eq. (1)

$$\sigma_{SC} = \sigma_{anis} + \sigma_{osc}. \quad (2)$$

At 7 mT the width oscillates rapidly with angle, whereas at 1.75 mT the oscillation is much slower. When analysing data from a range of angular scans at different fields, it is found that the oscillations scale linearly with the perpendicular component of the applied field  $B_z = B \cos \theta$  and  $\sigma_{osc}$  takes the form

$$\sigma_{osc} = \sigma_1 \sin(2\pi B_z/B_z^0). \quad (3)$$

The oscillation period  $B_z^0$  obtained from fitting angular scans to Eq. (2) is found to be described by a single value of  $B_z^0$  for all values of applied field (Fig. 2a). In contrast, the oscillation amplitude is particularly strong at low field and falls off at higher fields (Fig. 2b), suggesting that increasing

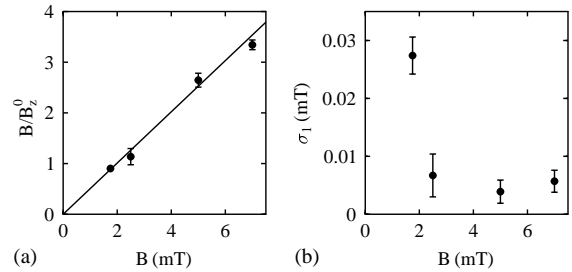


Fig. 2. Field-dependent parameters derived from fitting the data to Eq. (2): (a) the ratio  $B$  to  $B_z^0$  with the fitted line corresponding to the field independent value 1.98(4) mT for the parameter  $B_z^0$  in Eq. (3); (b) the oscillation amplitude  $\sigma_1$ .

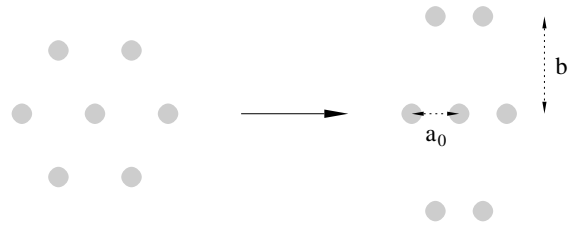


Fig. 3. Effect of chain formation on the in-plane vortex lattice;  $a_0$  is the equilibrium intra-chain vortex separation and the inter-chain spacing is  $b$ .

inter-vortex interactions tend to suppress the oscillations.

In order to try and explain the departures from the standard angular scaling of Eq. (1), we consider here two possible mechanisms: (1) flux lines deviating from the applied field direction [13,14] and (2) formation of vortex chains parallel to the tilt plane [8–12] (Fig. 3).

For the first mechanism, it is the angle of the vortex line with respect to the  $z$ -axis that determines the width rather than the applied field angle. Since it has been suggested that two different vortex line directions may be stable for a given applied field direction [13,14], it is possible that two differently aligned phases may be present at the same time. The measured width would then be the weighted average between the two phases and oscillations in weight between these two phases or in their angle of deviation from the applied field could explain the oscillations in the observed width. The angular deviations of the average vortex line direction from the applied field direction in this model can be estimated from

Fig. 1 to be about  $\pm 5^\circ$  at 7 mT and  $\pm 10^\circ$  at 1.75 mT. For the second mechanism, calculations of the effect of chain formation on the muon linewidth suggest that distortion of the vortex lattice constants by 20–50% would be sufficient to explain the observed amplitude of the oscillations.

Although both mechanisms can give a modulation amplitude consistent with experiment, the question remains as to what is driving the oscillations and how the oscillations become periodic in  $B_z$ . Although a clear answer to this question has not yet emerged, we believe that one possible explanation is that a geometric matching effect is taking place, where a length scale of the magnetic vortex structure periodically becomes commensurate with some other fixed length scale in the system. The vortex length scales for a normal tilted vortex array vary as  $B^{-1/2}$  [8]. In contrast, for the chain state (see Fig. 3) the intra-chain spacing within the high conductivity plane  $a_0$  is constant at low fields [8] and the intra-chain spacing in the direction perpendicular to the field varies as  $a = a_0 \cos \theta$ . The corresponding inter-chain separation  $b$  varies inversely with  $B$ , following  $b = \phi_0 / (a_0 \cos \theta B) = \phi_0 / (a_0 B_z)$  with  $\phi_0$  the flux quantum. Hence, if the oscillations have a geometric origin related to commensurability effects, it is clear that the chain state can provide a linear periodicity with  $B_z$  which matches experiment, whereas the normal tilted vortex array cannot. The corresponding commensurability matching condition for the chain state would be  $nb = x$  where  $x$  is the fixed length scale and  $n$  is an integer. The periodicity in  $B_z$  becomes  $\Delta B_z = \phi_0 / (a_0 x)$ . For our high anisotropy case, we expect  $a_0 \sim \lambda_{\parallel}$  and since  $\lambda_{\parallel} \sim 0.5 \mu\text{m}$ , we estimate  $x \sim 2 \mu\text{m}$  from the measured periodicity. It is not clear at present where such a periodicity might originate. Further work is under way to better understand the oscillatory effect and to identify more precisely the regions of the phase diagram where the oscillatory vortex structure is occurring.

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