Physica B 294–295 (2001) 422–426

PHYSICA B

www.elsevier.com/locate/physb

Vortex structure and dynamics in κ-$(ET)_2Cu(NCS)_2$

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Abstract

We use Josephson plasma resonance (JPR) and DC torque magnetization measurements to investigate the vortex structure and dynamics in the layered organic superconductor κ-$(ET)_2Cu(NCS)_2$. JPR studies probe the AC response of this extreme type-II superconductor for currents driven along the low conductivity axis. An observed crossover in the magnetic field and temperature dependence of the JPR frequency, in close proximity to the irreversibility line, is attributed either to a melting or a depinning transition in the quasi-two-dimensional (Q2D) vortex structure. Angle and temperature dependent magnetization studies reveal the rich dynamical process by which magnetic flux enters this highly anisotropic superconductor, as well as information on the structure of the vortices once inside. At extremely low temperatures ($T < 100$ mK), the sudden cessation of avalanching flux jumps above an angle- and temperature-dependent critical field $B^*(\theta, T)$, is indicative of Q2D vortex lattice melting. Thus, using these two very different techniques, we are able to probe the vortex phase diagram over a wide temperature and magnetic field range.

Keywords: Organic superconductors; Magnetic measurements; Flux jumps; Superconducting phase transitions

1. Introduction

In recent years, there has been intense investigation into the structure and dynamics of the magnetic flux lattice (FL) in extreme type-II superconductors [1–3]. A model system for these types of study is the 10 K organic superconductor κ-$(ET)_2Cu(NCS)_2$, where ET denotes bis-ethylenedithio-tetrathiafulvalene. Like the high-temperature superconductors (HTS), it has a highly anisotropic layered structure with the superconducting ET planes separated by insulating anion layers. However, unlike many of the HTS, this material is extremely clean, possessing far fewer crystal defects or pinning sites for magnetic flux. Furthermore, because of the reduced T_c and H_{c2} ($T_c = 10$ K and $\mu_0 H_{c2} = 4$ T for the field perpendicular to the layers), we are able to probe much more of the field/temperature parameter space within the superconducting state than is currently possible for the HTS. To this end, we have used two very different techniques – torque magnetometry (TM) and Josephson plasma resonance (JPR) – to investigate the vortex phase diagram over a wide range of temperatures and magnetic fields in this particular organic superconductor.

2. Experimental

Several different single crystals of κ-$(ET)_2Cu(NCS)_2$, of approximate dimensions $0.75 \times 0.5 \times 0.2\,\text{mm}^3$,

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0921-4526/01/$ - see front matter
PII: S0921-4526(00)00691-8

were used in this study; all of the samples were grown in the same batch using standard techniques [4]. JPR measurements were performed using a cavity perturbation technique in the frequency range from 28 to 153 GHz; for experimental details, see Ref. [5]. Magnetic torque measurements were carried out at the National High Magnetic Field Laboratory using a dilution refrigerator in conjunction with a 20 T superconducting magnet. The torque cantilever was mounted on a single-axis rotator, allowing for angle dependent measurements. An angle of $\theta = 0^\circ$ corresponds to the magnetic field applied parallel to the least conductive $\boldsymbol{a}$-axis, while $\theta = 90^\circ$ refers to the field applied within the highly conducting $\boldsymbol{bc}$-plane. Temperature-dependent torque measurements were performed at two angles ($\theta = 47^\circ$ and 74°) away from $\theta = 0^\circ$, where the torque is zero in this setup. Subsequent analysis of the angle dependent torque (from $\theta \approx 0$–90°) enabled us to scale the temperature dependence back to $\theta = 0^\circ$ [6]. The external magnetic field was swept at a constant rate of 1.5 and 0.5 T/min for JPR and TM measurements, respectively.

3. Results and discussion

In highly anisotropic superconductors, it is expected that the plasma mode for the low conductivity direction ($\sigma_{\boldsymbol{a}}$) lies below the superconducting gap. Below the critical temperature T_c, this plasma mode dominates the $\boldsymbol{a}$-axis microwave response with frequency ω_p, which depends on the maximum inter-layer (or Josephson) current $J_m(B, T)$, through the expression:

$$\omega_p^2(B, T) = \frac{8\pi^2 cs}{\varepsilon_c \Phi_0} J_m(B, T),$$

where

$$J_m(B, T) = J_0 \langle\langle \cos \varphi_{n,n+1}(\boldsymbol{r}) \rangle_t \rangle_d,$$

s is the inter-layer crystal spacing, ε_c is the high-frequency permittivity, Φ_0 is the flux quantum, $\varphi_{n,n+1}(\boldsymbol{r})$ is the gauge-invariant phase difference between layers n and $n+1$ at a point $\boldsymbol{r} = x, y$ in the $\boldsymbol{bc}$ (high conductivity) plane, and $\langle \dots \rangle_t$ and $\langle \dots \rangle_d$ denote thermal and disorder averages. J_0 is the maximum inter-layer Josephson current density at zero field ($B_{DC} = 0$), and $\lambda_\perp$ is the inter-layer London penetration depth.

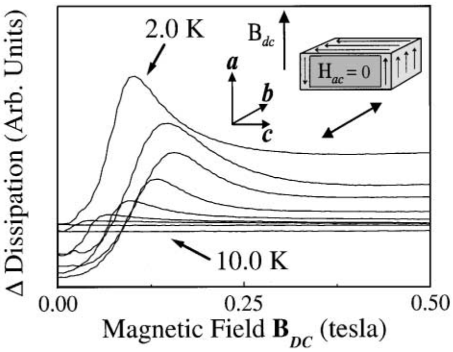

Fig. 1. Temperature dependence of the JPR at 111 GHz, from 2 to 10 K in integer steps. The inset depicts the DC and AC field geometries relative to the sample's crystallographic axes, as well as how AC currents flow within the sample (see Ref. [5]).

An important property of the Josephson coupling is the influence of an externally applied magnetic field on the collective plasma frequency ω_p. When the applied field is parallel to the least conductive axis, a mixed state is created in which the field penetrates the sample in quantized flux tubes, generating vortices in the superconducting layers. $\varphi_{n,n+1}(\boldsymbol{r})$ depends explicitly on the vortex structure within this mixed state and is thus responsible for the field dependence of the resonance frequency ω_p. If the flux tubes form straight lines along the $\boldsymbol{a}$-axis, $\langle \cos \varphi_{n,n+1} \rangle = 1$, and maximum Josephson coupling occurs. However, in the presence of disorder, e.g., as a result of crystal defects which create vortex-pinning sites, or through thermal fluctuations, the flux tubes may deviate from straight lines. This suppresses the maximum Josephson current. It is when ac currents are excited between the layers, at a frequency which corresponds to the natural frequency of the plasma oscillation, that a sharp resonance is observed. This resonance provides a direct probe of vortex structure in the mixed state.

Fig. 1 shows the microwave dissipation versus magnetic field at a frequency of 111 GHz, for temperatures between 2 and 10 K at intervals of 1 K. Notice, as the temperature is increased from 2 to

4 K the peak feature, which we associate with a JPR [5], moves to higher field. Such an increase corresponds to an increase in the inter-layer Josephson tunneling probability for cooper pairs (see discussion in Ref. [5]). In this field and temperature regime, the vortices are thought to exist in a quasi-two-dimensional (Q2D) solid phase, exhibiting long range order within each superconducting layer [7]. However, due to the random pinning of each Q2D "pancake" vortex solid, together with the weak inter-layer coupling, little or no correlation in the locations of vortices in adjacent layers is expected – hence, a low inter-layer Josephson tunneling probability results. The observed JPR temperature dependence is, thus, thought to be the result of increased thermal fluctuations, which actually promote inter-layer Josephson tunneling, i.e. increasing the temperature increases the mean deviations of individual vortices from their random positions relative to vortices in adjacent layers.

Upon increasing the temperature above 4 K (see Fig. 1), the JPR position begins to move back to lower fields, corresponding to a decrease in the inter-layer Josephson tunneling probability. One of two possible explanations for this "cusp" — the point in the B, T phase diagram at which the temperature dependence of the resonance changes – involves a melting of the Q2D vortex lattice. Above a field dependent critical temperature $T^*(B)$ [or temperature-dependent critical field $B^*(T)$], thermal fluctuations enhanced by low dimensionality will ultimately cause the mean vortex displacements to exceed some critical value (Lindemann criterion), at which point Q2D long-range order is lost, i.e. the Q2D vortex lattice melts. Above this critical temperature $T^*(B)$ [or critical field $B^*(T)$], increased thermal fluctuations actually result in the opposing effect of decreasing the inter-layer Josephson tunneling probability, hence, the opposite temperature dependence of the JPR.

Another possible explanation for the behavior observed in Fig. 1 involves a depinning transition. In this scenario, instead of melting, increased thermal fluctuations cause the individual pinned Q2D vortex lattices to undergo larger and larger collective oscillations about their mean positions, up to some critical depinning threshold, whereupon they become completely depinned, or mobile. Such a transition is indistinguishable from melting using this technique and, in either case, one would expect opposing temperature dependences for the JPR position above and below the transitions $T^*(B)$ and $B^*(T)$, i.e. as observed experimentally.

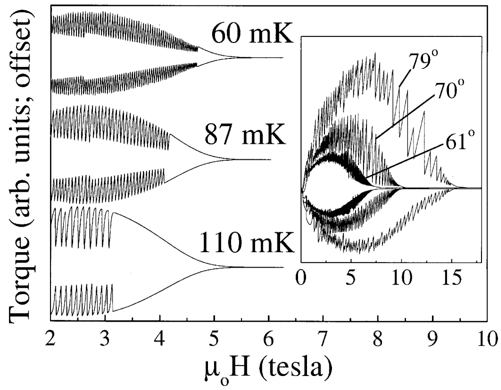

Fig. 2. Up (upper trace) and down (lower trace) field sweeps (at 0.5 T/min) for the torque measurements made at 60, 87, and 110 mK; the angle θ is 47° in the main panel of the figure. Inset: torque measurements (at 0.5 T/min) made at $\theta = 61°$, 70° and 79° and a temperature of 25 mK.

In the main panel of Fig. 2, torque is plotted versus magnetic field for $\theta = 47°$; both up and down sweeps are shown for various temperatures. The flux jumps are obvious, and have been observed previously in this and other materials [8,9]. We discuss the origin of this phenomenon briefly below; however, for a more detailed account, see Refs. [8–10]. Notice that the flux jumps stop abruptly above a temperature-dependent hysteretic critical value of the magnetic field, $B^*(T)$. This behavior is seen for all temperatures where flux jumps are visible, and for all angles (see also inset of Fig. 2) up to about 80°, above which this feature moves beyond the available magnetic field range.

Flux jumps are due to an avalanche behavior associated with the reorganization of magnetic flux as it enters the sample, which is presumed to be in the vortex solid phase. Crystal defects collectively pin each rigid Q2D vortex solid, thus causing a build up of flux near the sample surface, i.e. a critical Bean state [11]. At extremely low temperatures, a thermal boundary resistance tends to isolate the sample from the surrounding cryogen bath. Thus,

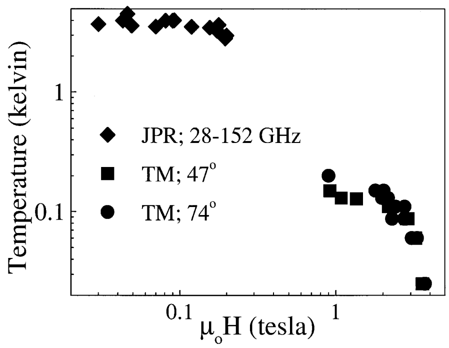

Fig. 3. A plot of the temperature dependent critical fields, $B^*(T)$, corresponding to observed transitions in the vortex structure by each technique, i.e. JPR and TM. The torque data have been scaled to $\theta = 0°$ to match the JPR data. Note the different parts of the available phase space probed by each technique.

runaway thermal instabilities due to viscous transport of vortices across the sample edge may cause large portions of the crystal to suddenly enter the normal metallic phase, at which point, magnetic flux is able to enter the sample in a catastrophic avalanche of vortices. Once the flux enters the sample, the metallic portions of the crystal once again become superconducting and the process starts anew. The existence of a vortex solid phase is essential for this behavior. In contrast, vortices will slip easily past each other in a vortex liquid phase, thereby allowing flux to enter the sample freely, i.e. the rigidity of a vortex solid, in combination with pinning, is necessary to establish the Bean state. Hence, we propose that the cessation of the flux jumps represents a melting transition into the liquid state from the Q2D vortex solid phase, driven either by thermal or quantum fluctuations.

Finally, a plot of $B^*(T)$ versus temperature is shown in Fig. 3, for both experiments. It is clear that the data obtained from the two techniques are appropriate for vastly different regimes of the field/temperature phase diagram. In fact the two data sets show quite different temperature dependences. This would seem to confirm that the two phenomena correspond to different transitions in the vortex structure, i.e. a depinning transition in the high temperature/low-field regime, and a melting transition in the mK regime. However, it is also possible that the two phase lines meet at a critical point corresponding to a crossover from a first order to a second-order transition, as is believed to be the case for a melting transition in highly anisotropic layered systems [6]. Furthermore, we cannot yet rule out the possibility that the low temperature transition is due to depinning. Clearly, further data obtained in the intermediate field/temperature regime should resolve this issue.

4. Conclusions

We have used two very different techniques to study transitions in the vortex structure in the mixed state of κ-$(ET)_2Cu(NCS)_2$. At moderate fields and high temperatures, a JPR has proven to be a useful tool for studying flux structure and associated transitions. While at very low temperature and high fields, torque magnetometry is a highly sensitive technique which enables a study of the vortex dynamics, as well as the melting of the Q2D vortex lattice.

Acknowledgements

This work was supported by NSF-DMR 0071953 and the Office of Naval Research (N00014-98-1-0538). Work carried out at the NHMFL was supported by a cooperative agreement between the State of Florida and the NSF under DMR-95-27035.

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