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Direct observation of vortices in the organic superconductor κ-$(\mathrm{BEDT\text{-}TTF})_2\mathrm{Cu}(\mathrm{NCS})_2$

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Results of direct imaging of Abrikosov vortices in an organic superconductor by means of the decoration technique are presented. The vortices have been observed in the title compound at temperature of $\simeq 0.5T_c$ at the magnetic fields 3.3–9.4 Oe directed perpendicular to the two-dimensional plane of the crystal. The decoration patterns reveal the sixfold coordination short-range order or slightly distorted triangular lattice of the vortices with the magnetic flux per vortex equal to the flux quantum, $hc/2e$. The slight deviation of the vortex lattice from the hexagonal symmetry is attributed to the in-plane anisotropy of the penetration depth. The average size of the individual vortex images on the sample surface yields the upper limit estimation of the London penetration depth as 0.4 μm at the given temperature. Pronounced effects of pinning on the crystal defects and thermally activated mobility of the vortices have been observed.

Layered organic superconductors with relatively high critical temperatures (T_c about 10 K) have been known for more than ten years.[1,2] They are characterized by an extremely high anisotropy, relatively strong electron correlations and, consequently, considerable fluctuation effects, resembling in these respects the metal-oxide high-T_c superconductors. Both families are well established strong type II superconductors whose magnetic and resistive properties below T_c are mainly determined by the vortex structure. However, no experimental data on direct observation of the Abrikosov vortices in the organic superconductors have been available so far. Such experiments could provide important information on the magnetic field penetration, vortex structure and dynamics, influence of surface and crystal lattice defects on the vortex pinning, and other properties of the mixed state.

The present work reports on direct imaging of the Abrikosov vortices in a single crystal of the organic superconductor (OS) κ-$(\mathrm{BEDT\text{-}TTF})_2\mathrm{Cu}(\mathrm{NCS})_2$ [where BEDT-TTF stands for bis(ethylenedithio)tetrathiafulvalene] by means of the so-called decoration technique which was successfully applied earlier to a number of conventional and high-T_c superconductors.[3–5]

Single crystals of κ-$(\mathrm{BEDT\text{-}TTF})_2\mathrm{Cu}(\mathrm{NCS})_2$ were obtained by the electrochemical oxidation of BEDT-TTF in 1.1.2-trichloroethane under a constant current regime at 20 °C. During first seven days the constant applied current was 1.75 μA then it was decreased to 1.5 μA. Under this current the crystals continued to grow for another three weeks. The complex of cyclic 18-crown-6 ether with Cu(SCN) and K(SCN) (1:1:1) was used as an electrolyte. The electrolyte was prepared in the electrochemical cell by dissolution of Cu(SCN) in the presence of K(SCN) and 18-crown-6 ether. The compound is a radical-cation salt characterized by a layered crystal structure:[2] conducting layers parallel to the crystal bc plane are formed by mutually orthogonal dimers of the radical cations of the BEDT-TTF molecule and separated from each other by insulating anion layers. The crystals used in the experiment were thin (<0.1 mm) platelets with the largest face being parallel to the bc plane and having typical size of about 1×0.5 mm^2. The superconducting critical temperature of the samples determined from the ac-susceptibility transition was $T_c \approx 10-11$ K, with the transition width of ≈ 1 K at the earth magnetic field. A single-crystal sample of the OS was glued by a conducting silver paste to a massive copper substrate which served as a thermal anchor to minimize the sample overheating during the decoration procedure. The temperature was measured by a resistive thermometer fixed to the substrate near the sample. The decoration was performed by means of sputtering of iron in helium atmosphere at pressure ~ 0.1 Torr onto the sample surface at temperatures below T_c. The tiny magnetic particles (5–10 nm)—''*magnetic smoke*,'' were formed directly near the superconducting surface during the sputtering process. The magnetic particles concentrated at the areas with higher local gradient of magnetic field which, in the mixed state, should mainly correspond to the positions of the Abrikosov vortices on the surface.

In more detail the decoration technique is described elsewhere.[3–5] The experiments were carried out in the field-cooling regime at temperatures 4–6 K and magnetic fields below 20 Oe directed perpendicular to the highly conducting bc plane. Since the applied magnetic field was small in our experiments the vertical component of the earth magnetic field, $H_E = 0.49$ Oe was always taken into account in the

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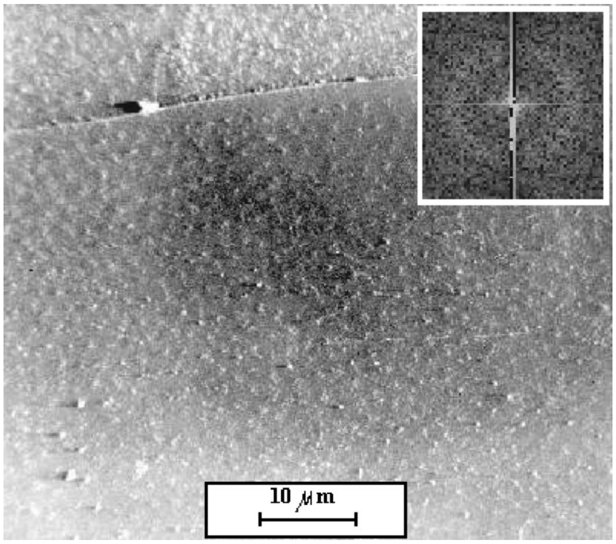


FIG. 1. The vortex structure in the κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ single crystal at the field of 4.1 Oe. Inset: 2D Fourier transform pattern in an arbitrary scale.

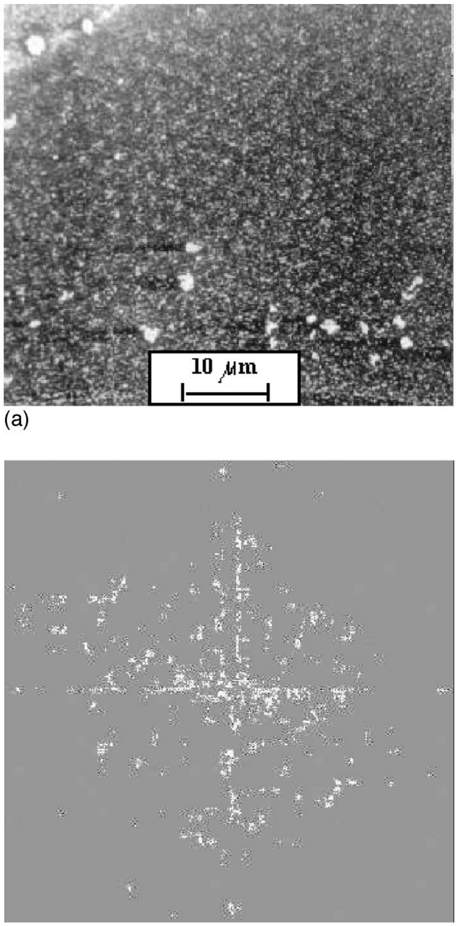
analysis. After decoration, the Cu substrate with the sample was warmed up to room temperature and transferred to SEM in order to study the distribution of the magnetic particles on the sample surfaces. The variation of intensity on SEM microphotographs was digitalized and investigated by applying two-dimensional fast Fourier transformation (2DFFT).[4] The main difficulties in the vortex decoration of the OS come from the high London penetration depth, relatively low T_c, poor thermal conductivity and wide region of reversible magnetization caused by the very high anisotropy.[2,6]

Successful imaging of the Abrikosov vortices was achieved on four different samples at the magnetic fields of 3.3, 3.7, 4.1, and 9.4 Oe, respectively. Figure 1 shows a nonhomogeneous distribution of the magnetic particles on a central part of the sample surface at magnetic field of $H = 4.1$ Oe. The bright areas correspond to a higher concentration of the particles.

Despite a relatively weak sharpness of the picture, one can resolve separated conglomerations of the magnetic particles corresponding to the positions of the Abrikosov vortices. The averaged magnetic induction B_{av}, estimated from the density of the conglomerations (i.e., their number per area unit) under the assumption of one flux quantum $\phi_0 = \pi\hbar c/e \approx 2\times10^{-7}$ G cm^2, equals to 4.3 G. This is in good agreement with the applied magnetic field.

The 2DFFT pattern shown in the insert in Fig. 1 reveals a blurred slightly distorted ring with the radius inversely proportional to the mean intervortex distance. The deviation of the ring from the ideal circular shape indicates the anisotropy of the vortex distribution. The absence of sharp maxima in the 2DFFT reflects the absence of the long-range order in the vortex structure. Nevertheless a short-range order with the six-fold coordination can be resolved in the vortex structure in the main panel of Fig. 1.

Figure 1 also illustrates pronounced interactions between the vortices and crystal defects: In the upper part one can see a step on the sample surface. At the pattern below the step, a narrow ($\simeq 2$ μm) Meissner band and a row of vortices parallel to the step are observed. A broader region of the Meissner state, more than 10 μm, was observed at the sample edges. Qualitatively, the pictures are similar to those obtained earlier on the high-T_c BSCCO crystals.[7] They are generally associated with a geometrical barrier[7] and/or magnetic flux creep.[8]



(a)

(b)

FIG. 2. (a) The vortex lattice in the κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ crystal at the field of 3.3 Oe; (b) the 2D Fourier transform pattern obtained from the picture in (a) in an arbitrary scale.

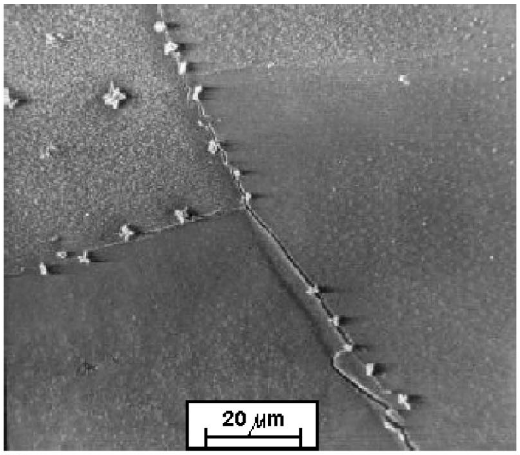
Figure 2(a) shows a decoration pattern for the magnetic field of 3.3 Oe. A part of the surface reveals a triangular lattice of vortices. The existence of the long range order is confirmed by relatively weak, however resolvable maxima at the 2DFFT displayed in Fig. 2(b). The estimated period of the vortex lattice $a \approx 2.4$ μm is in reasonable agreement with the expected value $a_0 = (2\phi_0/\sqrt{3}\mu_0 H)^{1/2} \approx 2.7$ μm. Thus, the inhomogeneous distributions of the magnetic particles in the presented patterns undoubtfully correspond to vortex structures. Below we discuss some features of the OS vortex structure in more detail.

A rough but direct estimation of the London penetration depth λ can be made based on the average size of individual conglomerations of the magnetic particles. Assuming that the vortex diameter equals 2λ, we obtain the upper limit for the penetration depth as 0.8 μm at the temperature of our experiment, 4–6 K. However, one should take into account that the vortices expand at the vicinity of the surface.[9] The diameter of a single isolated vortex is $d_{\mathrm{single}} \simeq 4\lambda$.[10] At the low magnetic field, $d \gg \lambda$, the vortex can be treated as almost isolated and our estimation of the penetration depth reduces to $\lambda \simeq 0.4 \pm 0.1$ μm. Despite the roughness of such estimation, we believe it is still useful, taking into account a strong scattering of the reported values: Our value is similar to those obtained by Lang *et al.*[11] and Lee *et al.*[12] [$\lambda(0) \simeq 0.5$ μm but significantly smaller than 1.4 μm reported by Dressel *et al.*[17]]. The corresponding estimation of the lower critical field, $H_{c1} = (\phi_0/4\pi\lambda^2)\ln\kappa$, yields 50 Oe for $\lambda = 0.4$ μm and 12 Oe for $\lambda = 0.8$ μm (the Ginzburg-Landau parameter is taken as $\kappa = 150$).[6] The fact that the decoration pattern at 9.4 Oe (corresponding to the intervortex distance of 1.6 μm) has revealed well separated vortices, the lower estimate for the penetration depth, 0.4 μm, looks more preferable.

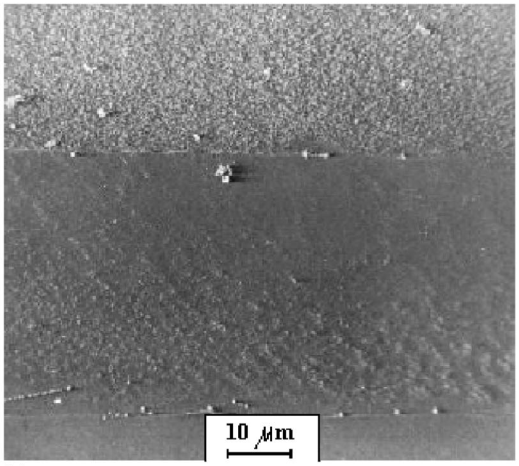
As seen from the 2DFFT pattern displayed in Fig. 2(b), the vortex lattice deviates from the regular 60° triangular shape. This is most likely caused by the anisotropy of the penetration depth within the bc plane. Further decoration experiments at lower temperature are expected to allow a quantitative evaluation of this anisotropy.

Perhaps the most interesting pattern of the OS surface is displayed in Fig. 3. It reveals, simultaneously, the areas with well resolved vortices and those in which the magnetic particles are distributed homogeneously either completely absent. The latter are understood as Meissner areas free of Abrikosov vortices. As to the regions with apparently homogeneous magnetic flux, their interpretation is not clear so far. In principle, they could correspond to either normal (nonsuperconducting) domains or to superconducting areas with delocalized vortices. The latter case can be realized if the vortex displacements during the decoration time [typically several hundred milliseconds and about 500 milliseconds for the experiment relating to Fig. 3(a)] exceed the inter-vortex spacing. If the vortex motion is chaotic and sufficiently fast (vortex liquid), it results in a homogeneous distribution of the decorating particles such as in the upper left corner of Fig. 3(a) or upper side of Fig. 3(b). If the vortices move along fixed direct trajectories, one can expect to resolve ''vortex tracks.'' This is likely the case at the lower side of the pattern shown in Fig. 3(b). Similar phenomena were observed earlier in some conventional superconductors.[13–15] We note that the decoration experiments were all carried out at temperatures about $0.5T_c$ at constant magnetic field. Therefore the sharp change in the magnetic structure taking place at the distance of several micrometers [Figs. 3(a), 3(b)] can hardly be caused by local overheating of strictly confined domains to temperatures above T_c. On the other hand, imperfect thermal contact with the Cu substrate could lead to slight variations of the temperature over the sample surface, thus inducing the vortex liquid state in the ''hot'' parts. The role of the thermal fluctuations is certainly enhanced due to the quasi-two-dimensional character of the superconductivity in the OS κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. In particular, this is the reason for an extended region of reversible magnetization in the H-T phase diagram of this compound (see, e.g., Ref. 6). It should be noted that the irreversibility field, $H_{\mathrm{irr}} \simeq 200$ Oe at $T \simeq 6$ K,[6] is much higher than the fields applied in the present experiment. Nevertheless, as seen from Fig. 3, the mobility of the vortices becomes considerable at temperature of ~ 6 K and fields as low as 4 Oe. This result is likely associated with the thermally induced breakup of the three-dimensional vortex lines into uncorrelated two-dimensional pancakes. A similar effect with the threshold temperature of $T^* \sim 5$ K was observed in μSR experiments by Lee *et al.*[12] The zero-temperature in-plane penetration depth estimated from the breakup temperature, $T_{\mathrm{bu}} = 5-6$ K, according to Clem[16] equals 590 ± 30 nm that is consistent with our above estimation as well as with the values given by Lang *et al.*[11] and Lee *et al.*[12]

Summarizing, in this paper we have presented the results of direct imaging of Abrikosov vortices in an organic superconductor by means of the decoration technique. The decoration patterns have revealed the sixfold coordination short-range order or slightly distorted triangular lattice of the vortices with the magnetic flux per vortex equal to the flux quantum $\phi = hc/2e$. Effects of pinning on the crystal defects



(a)



(b)

FIG. 3. The magnetic structure in the κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ crystal at the field of 4.1 Oe caused by the vortices motion: (a) the homogeneous magnetic particles distribution in the left top corner; the region without any particles in the center; the vortex structure marked by particles conglomerates on the right-hand side; (b) the homogeneous magnetic particles distribution at the top; blurred strings: ''the tracks of the moving vortices,'' at the bottom.

and thermally activated mobility of the vortices have been observed. With further progress in the decoration of the organic superconductors, more precise quantitative information on the vortex structure, in particular the penetration depth and its in-plane anisotropy will become available.

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