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Observation of the Fulde-Ferrell-Larkin-Ovchinnikov state in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$.

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Abstract

Magnetic measurements show that the quasi-two-dimensional superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ enters a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state below ~ 5.5 K in exactly in-plane magnetic fields $\sim 19 - 24$ T.

Key words: Organic superconductors; superconducting phase transitions; magnetic measurements.

There is great interest in the possibility of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) [1] state in organic superconductors [2]. In a metal in a magnetic field, the normal quasiparticles have separate spin-up and spin-down Fermi surfaces (FSs) which are displaced due to the Zeeman energy. In the FFLO state, attractive interactions of quasiparticles with opposite spin on opposite sides of the two FSs lead to the formation of pairs with nonzero total momentum [1,2] and hence an inhomogeneous superconducting state. It was suggested that the quasi-two-dimensional (Q2D) superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ in exactly in-plane magnetic fields is a possible candidate for the FFLO [3,4]; in this paper we report a direct observation of this state.

The experiments employed single crystals of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ ($\sim 1 \times 0.5 \times 0.1$ mm^3; mosaic spread $\lesssim 0.1°$). The procedures for preparing electrical contacts and orientating the crystals in the magnetic field are the same as those used in our study of the upper critical field [3]. In the current experiments, the crystal was also mounted on or in the coil of a tuned-circuit differential susceptometer (TCDS) [5]. The coil was mounted in a cryostat which allowed it (and the sample) to be rotated to all possible orientations in the magnetic field **B** [3]. The orientation of the

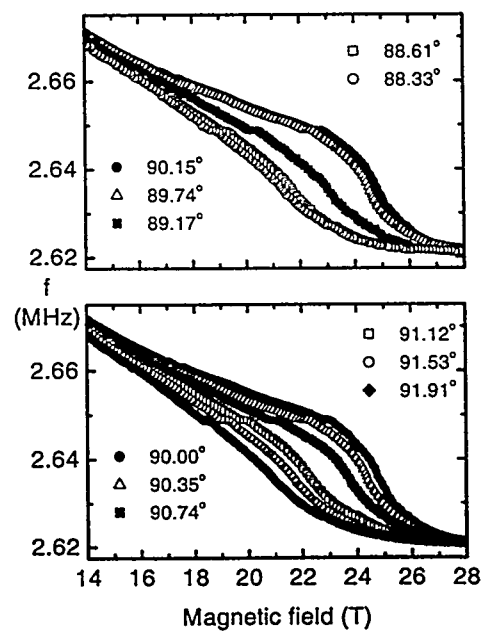

Fig. 1. TCDS frequency versus magnetic field for several different values of θ ($\phi = -45°$; $T = 1.39$ K). The elbow at $B_L \sim 22$ T disappears when the angle differs from 90° by more than about $\sim 1.5°$.

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sample is defined by the polar angle θ between **B** and the normal to the sample's **bc** planes and the azimuthal angle ϕ; $\phi = 0$ is a rotation plane of **B** containing **b** and the normal to the **bc** plane. The cryostat was placed in 31 and 33 T magnets at NHMFL, Tallahassee.

In order to detect the FFLO, we examine the *rigidity* of the vortex arrangement, which is predicted to change on going from mixed state to FFLO [6]. The sample is mounted with its Q2D planes perpendicular to the axis of the TCDS coil. When the quasistatic field is in the sample planes ($\theta = 90°$), the TCDS coil provides an oscillating magnetic field *perpendicular* to the static field (and the vortices) which exerts a torque on the vortices. The coil in the TCDS forms part of a tank circuit, so that changes in the rigidity of the vortices affect the effective "stiffness" of the circuit and therefore shift its resonant frequency f [5].

Figure 1 shows f for for several angles $\theta = 90° \pm \Delta\theta$. Superimposed on the gentle downward trend, due to the growing flux penetration of the sample [5], is an "elbow" at $\sim$ 22 T for values of $\Delta\theta$ close to zero. The elbow indicates a change in the vortex rigidity which we associate with the onset of the FFLO state; we label the corresponding magnetic field B_L. It is obvious that the elbow only occurs for $|\Delta\theta| <\sim 1.5°$; this is in good agreement with the calculations of Refs. [4,6], which predict that the FFLO is only stable in typical organic conductors for $|\Delta\theta| < 0.3 - 2.3°$. For bigger deviations, a substantial number of closed orbits will be possible on the FS [3], leading to suppression of the superconductivity due to orbital effects [3].

Fig. 2 compares B_L with the calculated FFLO phase diagram of Ref. [7], derived for a Q2D metal; also plotted are values of the resistive upper critical field B_p ($\theta = 90.00°$), defined in the inset to Fig. 2. The theoretical curves have been scaled using a $T = 0$ B_{c2} of 35 T and $T_c = 10$ K. Even though there are (not unexpected [3]) deviations of B_p from the theoretical dependence of B_{c2}, the data in Fig. 2 bear a striking similarity to the calculations of Ref. [7]. In particular, B_L follows the phase boundary between the Type-II superconducting state and the FFLO state (dotted curve) closely, extrapolating to B_p at $T \sim T^* = 0.56T_c$. The meeting of the two phase boundaries at $T^* = 0.56_c$ is a robust feature of models of the FFLO, irrespective of dimensionality [7]. Note that the effect is very reproducible; data for different samples and different cooling and bias conditions are shown in Fig. 2; all follow the same trends.

In summary, when the field lies in the Q2D planes of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, there is evidence for a transition into a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state. The data are in good agreement with theoretical predictions.

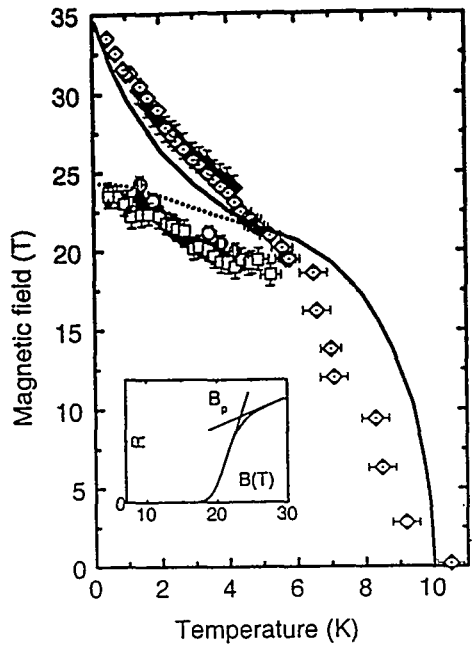

Fig. 2. Temperature dependence of the fields B_L (squares, circles) and B_p (diamonds; the field B_p is defined in the inset as the intersection of the two extrapolations); the different symbols indicate data from different samples and experimental runs. The data are compared with the FFLO phase diagram of Shimahara; the solid curve separates the superconducting and normal states. The boundary between the mixed state and the FFLO is shown as a dotted line.

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