Anisotropy of the Superconducting Order Parameter in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$

In a recent Letter, Schrama *et al.* [1] presented millimeter-wave cavity absorption measurements that claimed to show strong support for a *d*-wave symmetry of the superconducting wave function in the organic metal κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. The issue concerning the symmetry of the superconducting order parameter is of crucial importance in organic superconductors, since it has a direct bearing on the fundamental question as to whether the superconductivity is mediated ordinarily by phonons or more exotically by spin fluctuations.

While the results of Schrama *et al.* [1] are certainly intriguing, albeit that the measurements were made only over a restricted $<180°$ range in ϕ (as evidenced by the perfect 180° rotational symmetry of the plot in Fig. 3 of their paper), their conclusions are based on assumptions that are largely at odds with the findings of earlier experimental investigations. Most notably, in contrast to nonsuperconducting α-(BEDT-TTF)$_2$KHg(SCN)$_4$ [2], which is referenced by Schrama *et al.* as an example of a material for which the oscillating magnetic field penetrates completely into the sample, in the case of the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, it is well documented that the penetration depth is, in fact, rather small [3]. For interplane currents, $\lambda \sim 40\ \mu$m, while for in-plane currents, $\lambda \sim 1\ \mu$m (for $T < T_c/2$). Thus, for samples of the geometry and size used by Schrama *et al.*, the millimeter waves could not have penetrated deep into the bulk of the superconducting state (see [4]). The assertion made by Schrama *et al.* that they measure bulk conductivity is therefore very misleading. To further complicate matters, the electrodynamics change radically from one extreme (skin depth regime) to another (depolarization regime) on passing from the superconducting state into the normal state [3]. In the skin depth regime, in particular, both the sample geometry and its orientation within the cavity profoundly affect the degree of dissipation. What is more, the symmetry of the oscillating electromagnetic fields in the vicinity of the sample will be greatly distorted by the presence of such an anisotropic conductor; this is especially true for a platelet-shaped sample placed in an H-field antinode of rectangular TE_{102} cavity [1]. The exact orientation of the oscillatory magnetic field component with respect to the sample is therefore largely unknown, and is made still worse by the additional uncertainties caused by rotation of the sample within the cavity. With all of these factors taken into consideration, the assumption made by Schrama *et al.* that the ϕ-dependent changes in absorption between the two regimes are intrinsically related to the in-plane conductivity in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is not well founded. Given that both the phase and amplitude of the Shubnikov–de Haas oscillations are extremely sensitive to the orientation of the current path within the sample [5], the process by which the degree of absorption within the superconducting state is renormalized by the amplitude of these oscillations can then only make matters worse.

It is further worth noting that, while Schrama *et al.* [1] notionally associate the increase in absorption on entry into the superconducting state with the formation of a gap in the quasiparticle spectrum along certain directions in **k** space, other groups (which were not cited) have substantiated quite a different interpretation [6]. This absorption feature has, instead, been shown to result primarily from the effects of Josephson coupling of the superconducting condensate between the layers [6]. Besides this, since it has now been established, both experimentally and theoretically [7], that the quasiparticle spectrum is always essentially gapless within the vortex state, irrespective of the symmetry of the order parameter, it is unwise to assume that a gapped quasiparticle spectrum exists along any particular direction in **k** space, at any finite magnetic field. The question of the symmetry of the order parameter is therefore most reliably investigated at zero magnetic field. The absence of nodes in the superconducting wave function of other κ-phase BEDT-TTF salts (i.e., from the same family) has been demonstrated quite vividly by measurement of the specific heat [8]. While the large jump in the specific heat on cooling is indicative of strong coupling, the temperature dependence of the specific heat within the superconducting state reveals an exponential vanishing of the number of quasiparticles at low temperatures.

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