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Superconductivity and magnetism in organic layered superconductors

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Abstract

The present state of knowledge in the field of layered organic superconductors based on κ-(BEDT-TTF) is reviewed. Normal and ground states inside the temperature versus pressure phase diagram are discussed. The question of unconventional versus conventional superconducting pairing is addressed. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Organic superconductivity; Superfluid density; Spin-density waves

1. Introduction

It is remarkable that magnetic insulators are found in the vicinity of superconductors (SCs) not only for high T_c cuprates but also for layered organic SCs κ-(BEDT-TTF)$_2$X, where X stands for Cu(NCS)$_2$, Cu[N(CN)$_2$]Br and Cu[N(CN)$_2$]Cl [1,2]. The κ phases of BEDT-TTF (abbreviated ET) are materials in which orthogonally aligned ET dimers form 2D conducting layers sandwiched between the polymerised anion layers (Fig. 1). One hole per dimer hopes within ET layer much easier than between layers yielding the in-plane to out-of-plane conductivity anisotropy of the order of 10^3. In the organic SCs, the competition between these two ground states is governed by pressure and choice of the anion or the isotope substitution. Magnetic order is of an insulating antiferromagnet with mildly canted spins [3]. An indirect evidence of local charge ordering has been obtained recently by low frequency dielectric spectroscopy [4,5]. The experimental situation regarding the symmetry of the order parameter is still unclear and rather controversial. Approximately, a half of the performed studies gave evidence for non-s-wave pairing, while the other half showed the exponential dependence expected for s-wave pairing [6,8]. In this paper we overview essential features of the phase diagram, as well as main results we have obtained in the studies of the magnetic penetration depth that clearly indicate d-wave nature of the SC order parameter.

2. Phase diagram

Phase diagram is shown in Fig. 2 [9]. AF and SC phases occur next to one another. At the low

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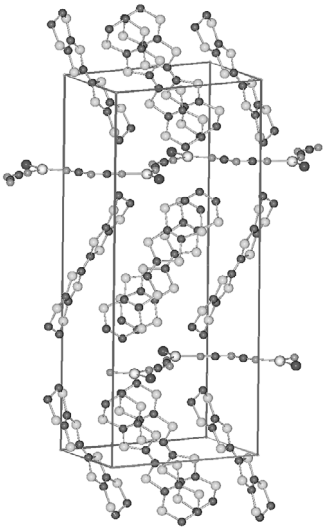

Fig. 1. Crystal structure of κ-(BEDT-TTF)$_2$X, where X stands for Cu[N(CN)$_2$]Br and Cu[N(CN)$_2$]Cl.

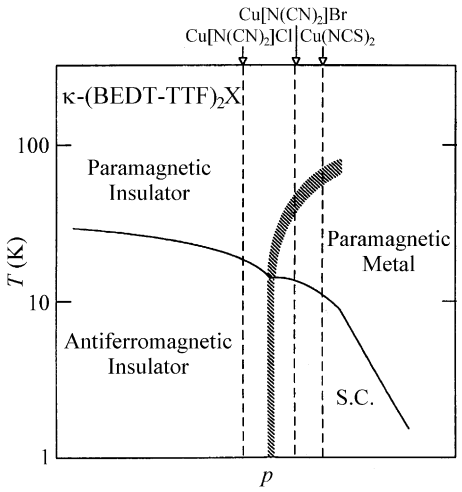

Fig. 2. Phase diagram of κ-(BEDT-TTF)$_2$X.

pressure side of the phase diagram, the DC resistivity increases gradually below room temperature due to very strong on-site Coulomb interactions. A commensurate spin-density wave (SDW) modulation of localised spins, which are canted by an angle of 0.06 degrees from the easy axis is established at $T_c = 22$ K [4]. The low temperature (LT) state is therefore, a weak ferromagnetic state divided in domains with equivalent spin configurations. The low frequency dielectric spectroscopy data indicate that SDW charge relaxation originates from domain wall pairs, whose size is about 0.1 μm [5]. A random charged domain wall pattern divides ET planes into randomly coupled AF regions. At very LT, a regular ferromagnetic domain structure is established that might imply the existence of static charge order. In addition, charged wall pair correlations are observed in the 10 K wide region above T_c.

Metallic phase, at the high pressure side of the phase diagram, is in some aspects distinct from conventional metal. Knight shift decreases significantly below 50 K, suggesting a suppression of the density of states [10]. An additional evidence for that was obtained by STM experiments [11]. Further, a peak in NMR spin–lattice relaxation rate at about 50 K suggests the appearance of short range AF correlations [10]. However, Fermi liquid-like behaviour, in particular Schubnikov–de Haas oscillations can be induced by moderate magnetic fields [2]. A disorder–order transition bearing glassy features happens at 75 K [12]. The transition is caused by gradual freezing down of the motion of the ethylene groups of the ET molecules that are thermally activated at high temperatures between two possible conformations. At LT in the SC state, the level of the residual disorder is critically determined by the time scale of the experiment. When the applied time scale is much longer than the mean relaxation time, the measured single crystal is in Relaxed state. A remenant disorder is harmless and a full Meissner state is achieved with the LT saturation value of χ' of almost -1 (Fig. 3). The latter corresponds to the measurement geometry when applied AC field is perpendicular to ET planes, whereas for the parallel field the SC volume amounts to 90%. Since the penetration depth anisotropy ratio is much larger than the aspect ratio of measured single crystals, the sample susceptibility is dominated by the in-plane and the out-of-plane penetration depth in the former and the latter geometry, respectively. The obtained $T = 0$ values for $\lambda_{in}(0) = 1.5$ μm and $\lambda_{out}(0) = 53$ μm correspond to the values reported in the literature. The in-plane and out-of-plane superfluid density, calculated according to $\rho_{s,in}(T) = (\lambda_{in}(0)/\lambda_{in}(T))^2$

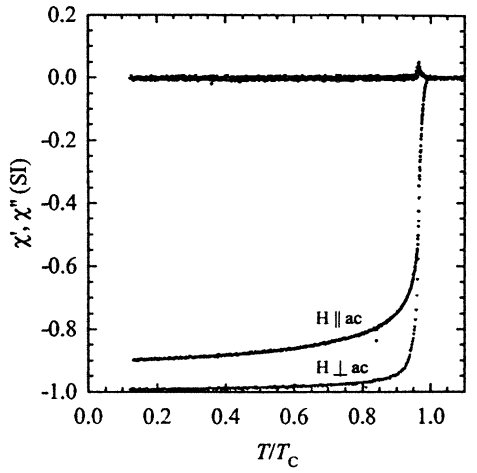

Fig. 3. Real and imaginary parts of the AC susceptibility in an AC field of 14 mOe parallel and perpendicular to crystal *ac* plane for the Relaxed state.

and $\rho_{s,out}(T) = (\lambda_{out}(0)/\lambda_{out}(T))^2$ for the Relaxed state are shown in Fig. 4. The leading term which describes the LT behaviour of $\rho_{s,in}$ and $\rho_{s,out}$ is T and T^2, respectively. Note that the shapes of both curves are qualitatively different from the s-wave BCS result.

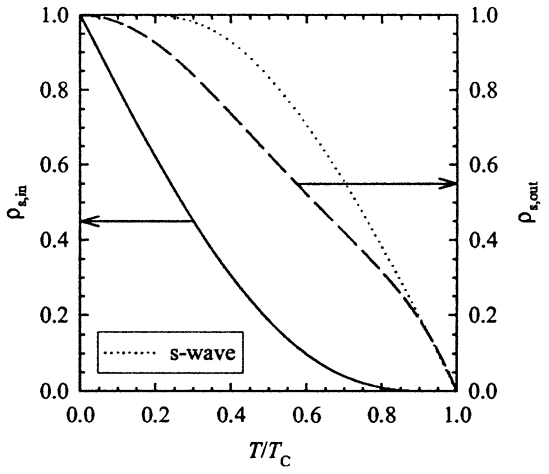

Fig. 4. In-plane superfluid density $\rho_{s,in}$ (—) and out-of-plane superfluid density $\rho_{s,out}$ (– – –) versus reduced temperature for the Relaxed state. The s-wave BCS result (···) is shown for comparison.

3. Summary

The observed power laws in the LT behaviour of the penetration depth are fully consistent with those expected in the d-wave model, in which the bulk SC state is created by antiparamagnon exchange and stabilised by the Josephson coherent tunnelling between SC layers [6,7]. Further, the T^3 dependence of the spin–lattice relaxation rate found in ^{13}C NMR measurements, the T^2 dependence of the electronic specific heat and the temperature linear planar thermal conductivity also confirm d-wave symmetry (see references in Ref. [6]). Nevertheless, even recently, there are experiments reported in the literature, which claim to find behaviour consistent with the s-wave SC [13]. In attempt to reconcile the existing contradictions, we have undertaken an investigation that covers a broad range of single crystals of various syntheses and in which the influence of thermal cycling and sample history is checked in carefully designed experiments. The full account of our work will be published elsewhere [14].

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