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Superfluid density and pairing in planar organic superconductors

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Abstract

We calculate self-consistently in-plane superfluid density $\rho_{\parallel}^{s}$ in planar organic superconductors for three types of pairing symmetry: $d_{x^2-y^2}$, d_{xy}-like and extended s-wave pairing. Assuming strong dimerization, we consider a single band model with elliptical Fermi surface to evaluate the temperature dependence $\rho_{\parallel}^{s}(T)$ in κ-(BEDT–TTF)$_2X$ compounds for those types of pairing. The obtained results, compared with measurements of the superfluid density in κ-(BEDT–TTF)$_2$Cu[N(CN)$_2$]Br, strongly suggest that in these compounds the gap symmetry, influenced by cooling history, in the "ground state" with the least disorder is of the extended s-wave type. Possible physical reasons for agreement of $d_{x^2-y^2}$, and/or isotropic s-wave pairing with experimental results in more disordered "intermediate state" are discussed.

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Keywords: Planar organic superconductors; Order parameter symmetry; Superfluid density

1. Introduction

The question of pairing symmetry in planar organic superconductors, like κ-(BEDT–TTF)$_2X$ (abbreviated in the following as κ-(ET)$_2X$), where X can be I_3, Cu[N(CN)$_2$]Br or Cu(SCN)$_2$, is still open. Theoretical studies of spin-fluctuation induced superconducting state have suggested a superconducting order parameter d-wave symmetry, similar as in cuprate high-T_c superconductors. This was found using the Hubbard model with fluctuation exchange (FLEX) approximation [1–3] or quantum Monte Carlo calculations [4]. However, whereas many experiments give evidence of unconventional pairing with nodes in the

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superconducting gap, others give results consistent with a fully gaped state [5,6]. In particular, measurements of the magnetic penetration depth indicate the presence of nodes [7], whereas those of the specific heat are consistent with a conventional gap [8]. To resolve this controversy, the Zagreb group (M. Pinterić, S. Tomić and coworkers) has performed a detailed analysis of the sample preparation and cooling procedure influence on the ac susceptibility and on corresponding temperature dependence of the magnetic penetration depth $\lambda(T)$ and the superfluid density $\rho^{s}(T)$ in planar organic superconductor κ-$(ET)_2Cu[N(CN)_2]Br$ [7,9,10]. They have shown that depending on the relaxation time of the terminal ethylene groups in ET molecules and of the cooling rate, one can reach the "ground state", with the least disorder, or some more disordered state. This "ground state" is not the state at $T = 0$; at $T < T_c$ it is characterized by almost complete diamagnetic response to magnetic field perpendicular to conduction planes, and is achieved in a sample dependent way, in annealed or in relaxed state [7]. In particular, there is a striking difference between low temperature dependence of in-plane superfluid density $\rho^{s}_{\|}(T)$ in the "ground state" where it is linear, and in the more disordered "intermediate state", where it is quadratic. In the "ground state" the leading term in $\rho^{s}_{\|}(T)$ is linear, as expected for the standard d-wave pairing (e.g. in the high-temperature superconductors). However, the coefficient in front of T/T_c found by the fit to experimental curves is much higher that in the d-wave model, and in the whole temperature region the curvature of $\rho^{s}_{\|}(T)$ is positive. Similar behavior of $\rho^{s}_{\|}(T) = \lambda^2_{\|}(0)/\lambda^2_{\|}(T)$ is found by Carrington et al. [11] in ac inductance measurements on slowly cooled samples of κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ taking $\lambda_{\|}(0) \sim 1$ μm, as given in the literature. They pointed out that only for $\lambda_{\|}(0) \geqslant 1.8$ μm the slope of $\rho^{s}_{\|}(T)$ becomes similar as in high-T_c cuprates, whereas in Ref. [7] this crossover value is even larger, $\lambda_{\|}(0) \sim 3$ μm. In both cases, it is evident that the pairing is unconventional, but it is possible that the pairing symmetry is not simply d-wave. The authors of the references [7,10] have considered a mixture $d + s$ of the order parameters, $\Delta(\mathbf{k}) = \Delta(\cos 2\phi + r)$, where ϕ is the angle between one principal axis in the crystal plane and the quasiparticle momentum $\mathbf{k}$. Changing the s-wave contribution r, one gets different curvatures of $\rho^{s}_{\|}(T)$. A good fit to experimental data (with $\lambda_{\|}(0) \sim 1$ μm) is obtained for rather large s-wave contribution, $r = |0.7|$, which is theoretically unlikely [7] and in contradiction with magnetothermal conductivity measurements, which suggest much smaller $|r|$ [12]. A similar mixed $(d + is)$ order parameter was proposed by Xu et al. [13]. In the present article we consider, instead, three types of pairing proposed theoretically for planar organics, two types of d-wave and the extended s-wave [6,14].

2. Model

We use the same model as applied previously [15] to calculate some bulk properties (the density of states, the tunnelling conductance, the electronic specific heat and the spin susceptibility) of κ-$(ET)_2X$ superconductors.

For d-wave pairing, we take $d_{x^2-y^2}$,

$$\Delta_{\mathbf{k}} = \Delta(T)(\cos k_x a - \cos k_y a), \tag{1}$$

and d_{xy}-like form

$$\Delta_{\mathbf{k}} = \Delta(T)[\cos k_x a + \cos k_y a - \alpha \cos(k_x a + k_y a)], \tag{2}$$

with $\alpha = 0.8$, as used by Tanuma et al. [14]. By applying the fluctuation exchange method to the Hubbard model, Kuroki et al. [16] have found that these pairing symmetries compete with each other, this competition being sensitive to the parameters values (see Fig. 7 of Ref. [16]).

For anisotropic s-wave pairing, we take the extended s-wave of the form

$$\Delta_{\mathbf{k}} = \Delta(T)(\cos k_x a + \cos k_y a). \tag{3}$$

An extended s-wave pairing has been proposed by Brandow [6] for κ-$(ET)_2X$ family to explain the presence of gap nodes observed in some experiments and their absence in others. The removal of nodes from an anisotropic gap may be related to various perturbations in these compounds, like the disorder between the ethylene groups and others sample defects. In the standard d-wave case,