Gossamer superconductivity in \( \kappa-(BEDT-TTF)_2X \)?

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Abstract

It has been recently established that the superconductivity (SC) in \( \kappa-(BEDT-TTF)_2Cu[N(CN)_2]Br \) is extremely sensitive to the cooling procedure through 100–70 K, where the ethylene groups attached to the BEDT-TTF molecules order with very slow relaxation times (the glass transition). In particular, the superconductivity in the annealed samples is described in terms of d\(_{x^2-y^2}\)-wave, while the quenched samples have very small (by a factor of \( 10^{-2} \) or less) superfluid density with the different temperature dependence. In spite of this dramatic difference, the SC transition temperature of these samples is practically unchanged. We propose that this dramatic decrease in the superconductivity is due to the appearance of spin-density wave order parameter in the quenched samples.

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PACS: 74.70.Kn; 74.25.Ha; 74.20.Rp

Keywords: Organic superconductors; Magnetic properties; Pairing symmetries

Among the organic superconductors three \( \kappa-(BEDT-TTF)_2X \) (abbreviated as \( \kappa-(ET)_2X \)) with \( X = Cu(NCS)_2, Cu[N(CN)_2]Br \) and Cu[N(CN)_2]Cl occupy the central position in part due to their relatively high SC transition temperatures (\( T_C = 9–13 \) K) and in part due to many parallels to the high-\( T_C \) cuprate superconductors: the layered structure and the proximity of the spin-density wave (SDW) phase [1]. In the past decade, a heated debate has been going on as to the nature of SC in \( \kappa-(ET)_2X \) salts: s-wave or d-wave. Finally, this question appears to be settled in favor of d\(_{x^2-y^2}\)-wave SC by two definitive experiments: the angular dependent STM [2] and the angular dependent magnetothermal conductivity [3], where the magnetic field is rotated within the conducting plane [4].

In spite of this new development, there still remains the controversy on the symmetry. In a recent paper we have established that the cooling procedure modifies dramatically the SC properties of \( \kappa-(BEDT-TTF)_2Cu[N(CN)_2]Br \) (abbreviated as \( \kappa-(ET)_2Br \)) [5]. Surprisingly, the SC transition temperature is little affected, while both the superfluid density \( \rho_s \) and its temperature \( (T) \) dependence strongly differ. For example, in the annealed samples, kept three days at the liquid nitrogen temperature before final slow cooling, the in-plane magnetic penetration depth \( \lambda_{in}(T) \) is of the order of micrometer and its \( T \) dependence is consistent with d-wave SC as in earlier works [6,7]. On the other hand, in the quenched samples \( \lambda_{in}(T) \) is 10 \( \mu \)m or larger implying that the in-plane superfluid density \( \rho_{s, in} \) is 1% or less of \( \rho_{s, in} \) in the annealed samples. Further, its \( T \) dependence can be interpreted in terms of s-wave SC (see Fig. 1). Since \( T_C \) decreases for the highest cooling rates at most for 1 K, there is no way to interpret these differences in terms of impurity scattering models [8,9]. So, what is going on? It is well known that for the \( \kappa-(ET)_2Br \) material the glass transition connected to the ordering of ethylene groups is located in the temperature region between 100–70 K [10]. Further, in the same temperature region the cooling rate strongly affects electronic properties, which can be clearly seen in the \( T \)
dependence of resistivity \cite{11}. But, the actual effect of remnant disorder on the low-temperature electronic properties of $j$-(ET)$_2$X remains unclear. Earlier, Kanoda’s group has studied both the effect of deuteration and the cooling rate on the SC in $\kappa$-(ET)$_2$Br \cite{12,13}. As in undeuterated samples, they found an enormous decrease in $\rho_{\text{in}}$ in the quenched samples and interpreted as the reduction in the volume fraction occupied by SC. Then presumably the remaining volume fraction should be occupied by SDW. Further, the broadening of the initial decrease of the diamagnetic response in the quenched samples suggests that the SDW and SC are distributed inhomogeneously all over the space. Does this model apply to undeuterated $\kappa$-(ET)$_2$Br as well? The competition between SC and SDW remains very plausible. However, the sharp decrease in the diamagnetic response even in the quenched samples \cite{5} is against inhomogeneous mixture of SDW and SC.

Another possibility is the spatially homogeneous distribution of the SC and SDW order parameters. There has been a long controversy as to the nature of the pseudogap in the underdoped region in the high-$T_C$ cuprate superconductors. Recently, a few people proposed that the pseudogap phase is described in terms of d-wave density wave \cite{14–16}. Then the SC state in the underdoped region consists of two order parameters: d-wave density wave and d-wave SC. Since SDW in $\kappa$-(ET)$_2$X salts looks conventional, the SC state in the quenched samples may be characterized as the state with two competing order parameters: SDW and d-wave SC. A model similar to “gossamer superconductivity” proposed by Laughlin \cite{17} may describe this state. Further experimental and theoretical work is needed to elucidate our proposal.

References