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LETTER TO THE EDITOR

On the relationship between the critical temperature and the London penetration depth in layered organic superconductors

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Abstract

We present an analysis of previously published measurements of the London penetration depth of layered organic superconductors. The predictions of the BCS theory of superconductivity are shown to disagree with the measured zero temperature, in plane, London penetration depth by up to two orders of magnitude. We find that fluctuations in the phase of the superconducting order parameter do not determine the superconducting critical temperature as the critical temperature predicted for a Kosterlitz–Thouless transition is more than an order of magnitude greater than is found experimentally for some materials. This places constraints on theories of superconductivity in these materials.

In this letter we consider the layered organic superconductors such as κ -ET₂X and λ -BETS₂Y¹. Most theories of superconductivity in these materials are based on BCS theory with either phonons [1] or spin fluctuations [2, 3] providing the attractive interaction. However, we will show that simple BCS theory is inconsistent with the measured London penetration depth [4–6]. Layered organic superconductors are, in many ways, similar to the cuprates [7]. Both classes of materials are quasi-two dimensional (q2D) and have phase diagrams which include antiferromagnetism, a Mott transition, unconventional metallic states and superconductivity. The superconducting state of the cuprates has d-wave symmetry [8] and, although there is, as yet, no consensus [9] on the pairing symmetry in the organics, several authors have presented evidence for d-wave pairing [9]. NMR experiments on the layered organic superconductors are suggestive of a pseudogap [10] similar to that observed in the cuprates [11]. It has been suggested that the Hubbard model is a minimal model for both of these systems [12, 13]. The most notable difference between the two classes of materials is that in the cuprates doping

¹ ET is bis(ethylenedithio)tetrathiafulvalene, BETS is bis(ethylenedithio)tetraselenafulvalene, X and Y are anions, e.g., $X = Cu(NCS)_2$ or $Y = GaCl_4$, and the Greek indices indicate crystal structure; see [12] for a recent review.

Table 1. The electron and superfluid densities of various layered organic superconductors. $m_{\beta}^*/m_{\rm e}$ is the effective mass of the magnetic breakdown (β) orbit determined from quantum oscillation experiments. $n_{\rm e}$ is the electron density calculated via the Onsager relationship from quantum oscillation experiments. The quasi-two-dimensional area occupied by a dimer is $A_{\rm d} = V_{uc}/N_{\rm d}d$, where $V_{\rm uc}$ is the volume of the unit cell, $N_{\rm d}$ is the number of dimers per unit cell, and d is the average interlayer spacing. Thus for a quasi-two-dimensional tight binding model of dimers at half filling one expects $n_e A_d = 1$. This is indeed observed experimentally. This shows that there is no correlation between the band filling and the many-body effects responsible for the mass renormalization. We have taken both T_c and λ_0 from the same experiments as both quantities can be sample dependent [9]. The superfluid density defined by $n_{\rm s} = m^* c^2/4\pi e^2 \lambda_0^2$. Note that $n_s/n_{\rm e}$ varies approximately linearly with T_c and that the BCS prediction that $n_{\rm s} = n_{\rm e}$ is strongly violated by the low T_c materials: β -(ET)₂IBr₂, α -(ET)₂NH₄Hg(NCS)₄ and κ -(BETS)₂GaCl₄.

Material	$n_{\rm e} \ ({\rm nm}^{-2})$	$A_{\rm d}~({\rm nm}^2)$	$n_{\rm e}A_{\rm d}$	$T_{\rm c}$ (K)	$\lambda_0 \; (\mu m)$	$m_{\beta}^{*}/m_{\mathrm{e}}$	$n_{\rm s} \ ({\rm nm}^{-2})$	$n_{\rm s}/n_{\rm e}$
κ -(ET) ₂ Cu[N(CN) ₂]Br	1.83 [19]	0.552 [20]	1.01	11.6 [21]	0.78 [21]	6.4 [22]	0.64	0.35
κ -(ET) ₂ Cu(NCS) ₂	1.83 [23]	0.519 [24]	0.95	9.4 [4]	0.54 [4]	6.5 [23]	1.0	0.55
λ -(BETS) ₂ GaCl ₄	1.95 [25]	0.484 [26]	0.94	5.5 [4]	0.72 [4]	6.3 [25]	0.63	0.32
β -(ET) ₂ IBr ₂	1.91 [27]	0.549 [28]	1.05	2.21 [4]	0.90 [4]	4.0 [27]	0.21	0.11
α -(ET) ₂ NH ₄ Hg(NCS) ₄	1.95 [29]	0.488 [30]	0.95	1.12 [4]	1.09 [4]	2.0 [29]	0.098	0.050
κ -(BETS) ₂ GaCl ₄	2.11 [31]	?	?	0.16 [4]	2.26 [4]	2.4 [31]	0.025	0.012

changes the charge carrier density, whereas the organics are, as we will confirm, half filled for all the anions that we consider here.

It has been suggested [14] that fluctuations in the phase of the superconducting order parameter determine the superconducting critical temperature, T_c , in both the (underdoped) cuprates and the layered organic superconductors. In this letter we will show that recent experimental data [4–6] disprove this conjecture in the case of the layered organic superconductors, and discuss which theories are consistent with these experiments.

It is widely believed that the anion layers of the layered organic superconductors are insulating and that at low temperatures the metallic phase of the organic layers can be described by a Fermi liquid tight-binding model that is half filled [12]. A check of this model is to compare its predictions with the size of the orbits observed in quantum oscillation experiments. The area, A, enclosed by an orbit in a quantum oscillation experiment is related to the observed frequency, F in 1/B, where B is the magnetic field strength, by the Onsager relationship, $A = \frac{2\pi e}{\hbar}F$. Thus for a q2D Fermi liquid it follows from Luttinger's theorem that the electron density $n_e = \frac{\hbar}{4\pi^3 e}F$.

The q2D area occupied by a dimer, A_d , can be calculated from crystallographic measurements (see the caption to table 1). If one assumes that each dimer donates exactly one electron to each anion then the product $n_e A_d$ is predicted to be unity. It can be seen from table 1 that this prediction is in excellent agreement with experiment.

In general, a state is deemed superconducting if it breaks gauge symmetry and displays a Meissner effect in weak magnetic fields. It follows directly from these very general requirements that a supercurrent, $\mathbf{j} = -D_s 2e\mathbf{A}/\hbar \equiv -c^2\mathbf{A}/4\pi\lambda^2$, is induced by a magnetic vector potential \mathbf{A} . λ is the London penetration depth and D_s is the superfluid stiffness. In BCS theory and its extensions one can separate D_s into a superfluid density and an effective mass, $(D_s \propto n_s/m^*)$. Here m^* is the effective mass of the quasiparticle excitations and n_s describes the proportion of electrons in the condensate in the terms of the two-fluid model. However, this separation is *not* a necessary feature of a superconducting state [15].

In London theory the zero temperature superfluid density is defined as [16] $n_s = m^* c^2 / 4\pi e^2 \lambda_0^2$, where λ_0 is the average London penetration depth parallel to the q2D planes at zero temperature. BCS theory [16] predicts that $n_s = n_e$ and Eliashberg theory [17] predicts

$$\frac{n_{\rm s}}{n_{\rm e}} = \frac{1 + \frac{1}{3}F_1^s}{m^*/m},\tag{1}$$

where F_1^s is a Landau Fermi liquid parameter. For a Galilean invariant system $1+F_1^s/3 = m^*/m$ and so $n_s = n_e$. But for systems with broken translational symmetry, such as the crystals that we consider here, there is no *a priori* relationship [18] between F_1^s and m^* .

It can be seen from table 1 that the predictions of BCS theory are in disagreement with experiments on the layered organic superconductors by up to two orders of magnitude. It has been suggested that only the q2D pocket of the Fermi surface of κ -ET₂Cu[N(CN)₂]Br is involved in superconductivity [19]. Such Fermi surface sheet dependent superconductivity can be ruled out as the explanation of the reduced superfluid density because, for example, the Fermi surface of β -ET₂IBr₂ ($n_s/n_e = 0.11$) has only one sheet [27]. Corrections due to the variation in the Fermi velocity around the Fermi surface [32] may be able to explain small deviations from $n_s/n_e = 1$, but are certainly not large enough to explain the extremely small superfluid density observed in the low T_c materials.

The simplest explanation of the penetration depth measurements is that not all of the electrons participate in the superconducting condensate. This would lead to many observable predictions. For example, thermodynamic indications of the superconducting state would be expected to show a 'mixed' behaviour, e.g. the specific heat anomaly should be extremely small in low T_c , low n_s compounds. Thus the observation of a clear anomaly in the heat capacity [34] in α -ET₂NH₄Hg(NCS)₄ ($n_s/n_e = 0.05$) appear to rule out scenarios in which only a fraction of the conduction electrons enter the condensate. Another possibility that retains the independent concepts of the effective mass and the superfluid density is to allow the Cooper pair to have an effective mass that is not simply $2m^*$. This has been discussed elsewhere and we will not dwell on this idea here as it was shown [35] that even in these scenarios it is still necessary to set $n_s/n_e \neq 1$ to explain the observed behaviour of the layered organic superconductors.

Note that for the organics the superfluid density is smallest for those materials with the lowest T_{cs} and the smallest effective masses, i.e., those materials that are the least strongly correlated. This is in direct contradiction with the predictions of the simple interpretations of the BCS and Eliashberg [17] theories where as the electron–phonon (or indeed electron–electron) couplings increase so do m^* and T_c . In the underdoped cuprates the pseudogap is associated with low critical temperatures and small superfluid densities, whereas in the organics the pseudogap-like features are associated with high critical temperatures and large superfluid densities. However, in both classes of materials the pseudogap is found close to the Mott transition.

It has been suggested [18] that in the cuprates F_1^s increases as m^* increases, rather than in the decreasing as is the case for a Galilean invariant Fermi liquid. Could a similar, albeit significantly stronger, effect be at play here? If $n_s \rightarrow 0$ as $T_c \rightarrow 0$ (while at the same time m^* decreases) then (1) requires that $F_1^s \rightarrow -3$ as $T_c \rightarrow 0$. For a momentum independent self energy $F_1^s = 0$ (see [33]); therefore for either the BCS or Eliashberg theories to be consistent with the data would require a strong momentum dependence in the self energy. Electron phonon coupling can only generate a momentum dependent self energy if Migdal's theorem is strongly violated². However, a strong momentum dependent self energy may be a more natural feature of spin fluctuation mediated superconductivity [2, 3].

² Note that none of the phonon mechanisms that have been discussed in the context of the organics [1] have proposed that Migdal's theorem is broken.

In the case of very strong electron–electron interactions equation (1) may not be valid. However, importantly, unlike underdoped cuprates, in the organics the normal state at temperatures only slightly above T_c appears to be a good Fermi liquid [12]. Hence, there is a need to calculate D_s for the models and approximations that have been proposed for the organic superconductors [1–3] to see if they predict the observed variation in D_s with T_c .

A possible explanation of the measured penetration depths is that the microscopic theory of superconductivity in the layered organics, whatever it may be, does not admit the separation of the superfluid stiffness into parts that correspond naturally to a superfluid density and an effective mass. This has the advantage of allowing the observation of a small superfluid stiffness to be reconciled with evidence that all of the electrons participate in the condensate.

To explain the Uemura relation [36], namely that in the underdoped cuprates $T_c \propto 1/\lambda_0^2$, Emery and Kivelson [14] proposed that phase fluctuations can limit the transition temperature of a q2D superconductor. The limit on T_c due to phase fluctuations, T_{θ}^{max} , is given by

$$k_{\rm B}T_{\theta}^{\rm max} = A \frac{\hbar^2 c^2 a}{16\pi e^2 \lambda_0^2},\tag{2}$$

where *a* is the larger of *d*, the average spacing between the q2D planes, and $\sqrt{\pi}\xi_{\perp}$, where ξ_{\perp} is the coherence length perpendicular to the planes. *A* is a constant of order 1. In the case of vanishingly small coupling between the planes we have a genuinely two-dimensional system and therefore the superconducting transition is a Kosterlitz–Thouless phase transition. In the underdoped cuprates further support for these ideas comes from measurements of the optical conductivity [37] of Bi₂Sr₂CaCu₂O_{8+ δ} for $T > T_c$ which are consistent with the predictions of Kosterlitz–Thouless theory and the observation of vortex-like excitations [38] above T_c in La_{2-x}Sr_xCuO₄. However, we should note that the evidence of phase fluctuations in these experiments did not extend to temperatures as high as those at which the onset of the pseudogap is observed [39].

Emery and Kivelson [14] suggested that the data of Uemura *et al* [36] implies that the critical temperatures of the layered organic superconductors in general and of κ -ET₂Cu(NCS)₂ in particular are also limited by phase fluctuations. In figure 1 we plot T_c as a function of λ_0 for a variety of layered organic superconductors. It can clearly be seen that T_{θ}^{max} is more than an order of magnitude larger than T_c for some of the materials considered (see [35]).

Further evidence that T_c is limited by the temperature at which pairing occurs and not by the energy scale of phase fluctuations comes from the ratio of the zero temperature superconducting order parameter, $\Delta(0)$, to T_c . For κ -ET₂Cu[N(CN)₂]Br and κ -ET₂Cu(NCS)₂ it has been found that [40] that $\Delta(0)/k_BT_c = 2.5-2.8$. These values of $\Delta(0)/k_BT_c$ seem more consistent with strong coupling superconductivity than with the expectation [41] that, if T_c is limited by phase fluctuations, $\Delta(0)/k_BT_c \gg 2$, which is indeed found for the underdoped cuprates. Measurements of $\Delta(0)/k_BT_c$ in low T_c materials may be expected provide a more stringent test of this criterion; however, we are not aware of any such measurements.

The destruction of superconductivity by phase fluctuations is strongly linked with the idea that preformed pairs are responsible for the pseudogap in the cuprates [11, 14, 37, 38]. Therefore the observation that T_c is not limited by phase fluctuations in the layered organics makes it unlikely that preformed pairs are responsible for the pseudogap-like features observed by NMR [10].

For the cuprates several theories have been proposed that may admit an increase in the superfluid stiffness as one moves away from the Mott insulating phase by increasing the doping from half filling. Examples of these include the RVB state [13] and its generalization gossamer superconductivity [43], the SU(2) slave-Boson model [44], and the two-species treatment of the t-J model [45]. Thus the observation that the superfluid stiffness varies as one moves away



Figure 1. Variation of the superconducting critical temperature, T_c , with the zero temperature penetration depth, λ_0 . The experimental data are taken from Pratt *et al* [4], Lang *et al* [5] and Larkin et al [6] and show data for κ -ET₂Cu[N(CN)₂]Br (open diamond) [5] κ -ET₂Cu(NCS)₂ both at ambient pressure (circle) [4] and under pressure (open squares), [6] λ -BETS₂GaCl₄ (square), β -ET₂IBr₂ (diamond), [4] α -ET₂NH₄Hg(NCS)₄ (triangle) [4] and κ -BETS₂GaCl₄ (empty circle) [4]. The empirical fit, $T_c \lambda^3 = 2.0 \text{ K} \mu \text{m}^3$, to the data from Pratt *et al* is also reproduced (dotted dashed line). Note that the data of Larkin et al (open squares) are actually for the penetration depth at $T = 0.35T_{\rm c}$. This means the data should be shifted somewhat to the left. However, even given this caveat the pressure dependence data of Larkin et al are in broad agreement with the ambient pressure data of Pratt et al. The upper limit imposed on T_c by phase fluctuations, $T_{\rm d}^{\rm max}$ (2), is shown for both the three-dimensional (dashed line, A = 2.2) and two-dimensional (solid line, A = 0.9) cases. Although it is possible that the details of the short-range interactions of the layered organic superconductors change the exact numerical values of A (see [42]), it is difficult to imagine that this effect is large enough to account for the order of magnitude difference between the predictions of the phase fluctuation model and the observed variation of T_c with λ_0 . For the phase fluctuation curves (solid and dashed lines) we take a = d = 18 Å, where a is the length parameter in equation (2) and d is the interlayer spacing which approximately 18 Å for all of these materials.

from the Mott insulating phase in the layered organic superconductors may indicate that one of these theories provides the correct microscopic description of these materials. Clearly detailed calculations are required to discover whether any of these models agree with the experimentally measured penetration depth.

It appears then that the key to understanding the microscopic details of the superconducting state in the organic superconductors is the low T_c materials. In addition to the need for a detailed systematic study of the thermodynamics of the low T_c materials discussed here, there are several other materials with low ambient pressure T_c s that should be investigated, such as β -ET₂AuI₂ ($T_c = 4.9$ K), κ -ET₂I₃ ($T_c = 3.6$ K), λ -BETS₂GaCl₃F ($T_c = 3.5$ K), κ -DMET₂AuBr₂ ($T_c = 1.9$ K), BO₂Re₄·H₂O ($T_c = 1.5$ K) and β -BO₃Cu(NCS)₃ ($T_c = 1.1$ K).

We have shown that the zero temperature superfluid stiffness of the layered organic superconductors is up to two orders of magnitude smaller than is predicted by simple BCS theory. We have also shown that phase fluctuations do not limit T_c in these materials as the transition temperature is more than an order of magnitude smaller than is predicted for a Kosterlitz–Thouless phase transition. This places constraints on theories of superconductivity in layered organic superconductors. It is therefore clear that the unusual behaviour of the penetration depth is a key experimental result which any theory of the layered organic superconductors must explain.

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