

The origin of the difference in the superconducting critical temperatures of the β_{H} and β_{L} phases of $(\text{BEDT-TTF})_2\text{I}_3$

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Incommensurate lattice fluctuations are present in the β_{L} phase ($T_c \sim 1.5$ K) of ET_2I_3 (where ET is BEDT-TTF - bis(ethylenedithio)tetrathiafulvalene) but are absent in the β_{H} phase ($T_c \sim 7$ K). We propose that the disorder in the conformational degrees of freedom of the terminal ethylene groups of the ET molecules, which is required to stabilise the lattice fluctuations, increases the quasiparticle scattering rate and that this leads to the observed difference in the superconducting critical temperatures, T_c , of the two phases. We calculate the dependence of T_c on the interlayer residual resistivity. Our theory has no free parameters. Our predictions are shown to be consistent with experiment. We describe experiments to conclusively test our hypothesis.

It has long been known that the application of hydrostatic pressure, P , to $\beta\text{-ET}_2\text{I}_3$ has a dramatic effect on the superconducting critical temperature, T_c . At ambient pressure $T_c \sim 1.5$ K but when the applied pressure reaches $P \sim 1$ kbar a discontinuous increase in T_c (~ 7 K) is observed. The low T_c state ($P \lesssim 1$ kbar) is denoted the β_{L} phase and the high T_c state ($P \gtrsim 1$ kbar) is labelled the β_{H} phase. When the pressure on the β_{H} phase is decreased the material does not return to the β_{L} phase but rather T_c is seen to further increase. Below $T \sim 130$ K the resistivity of the β_{H} phase is found to undergo a discontinuous decrease while no such anomaly is found in the β_{L} phase [1]. Incommensurate lattice fluctuations have been observed in the β_{L} phase but they are absent in the β_{H} phase below $T \sim 130$ K [2]. The incommensurate lattice fluctuations are stabilised by variations in the conformational ordering of the terminal ethylene groups of the ET molecules and thus can only exist in the presence of disorder [3]. For a recent review of this phenomenology see [4].

Although the change in resistivity between the β_{L} and β_{H} phases and the incommensurate lattice fluctuations have been studied both theoretically [3] and experimentally [1, 2, 4], no explanation of the change in T_c has been forthcoming. In this paper we propose that difference in the T_c 's of the two phases is due to disorder, we will show that current data is consistent with our explanation and suggest experiments which could clearly determine whether or not this is the correct explanation

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of the difference in T_c .

In a recent paper it was shown [5] that intrinsically non-magnetic disorder decreases the T_c of a wide range of the superconducting salts of ET in line with the Abrikosov–Gorkov (AG) formula:

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \psi\left(\frac{1}{2} + \frac{\hbar}{4\pi k_B T_c \tau}\right) - \psi\left(\frac{1}{2}\right), \quad (1)$$

where T_{c0} is the superconducting transition temperature of a pure sample, $1/\tau$ is the quasiparticle scattering rate and $\psi(x)$ is the digamma function. There are two scenarios compatible with this observation: either there is opposite spin pairing and the impurities induce localised magnetic moments, or else there is a finite angular momentum pairing state (most probably d-wave pairing). In this paper we will not discuss which of these scenarios is realised, but rather make use of this observation of the effect that increasing $1/\tau$ has on T_c .

In a quasi-two dimensional metal, such as β -ET₂I₃, the residual interlayer resistivity is given by

$$\rho_0 = \frac{\pi\hbar^4}{2e^2 m^* c t_\perp^2 \tau} \quad (2)$$

where m^* is the quasiparticle effective mass, c ($= 15.291$ Å for β -ET₂I₃ [4]) is the interlayer lattice constant and t_\perp is interlayer hopping integral. Substituting (2) into (1) one finds that

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \psi\left(\frac{1}{2} + \frac{e^2 m^* c t_\perp^2}{2\pi^2 \hbar^3 k_B T_c} \rho_0\right) - \psi\left(\frac{1}{2}\right). \quad (3)$$

from quantum oscillation Wosnitza *et al.* [6] found that $m^*/m_e = 4.2 \pm 0.2$, where m_e is the electronic rest mass, $t_\perp/E_F = 1/(175 \pm 10)$, where E_F is the Fermi energy, and the Fermi wavevector, $k_F = (3.4 \pm 0.2) \times 10^9$ cm⁻¹. Thus taking $E_F = \hbar^2 k_F^2 / 2m^*$ one finds that $t_\perp = 0.59 \pm 0.08$ meV. $T_{c0} = 7.75$ K is found from fast electron irradiation experiments [5, 7].

In figure 1 we plot T_c as a function of ρ_0 . We stress that there are no free parameters in this plot once the data from Wosnitza *et al.* and Forro *et al.* [5, 7] is considered. For comparison we also plot the data of Tokumoto *et al.* [8] who deliberately induced impurities in β_H -ET₂I₃ by fabricating the alloy β -ET₂(I₃)_{1-x}(IBr₂)_x. The data is consistent with our theory although clearly more data is required to properly test our prediction.

We propose that the disorder in the conformational degrees of freedom of the terminal ethylene groups of the ET molecules which is required to stabilise incommensurate lattice fluctuations in the β_L phase [3], but absent in the β_H phase, increases the quasiparticle lifetime. (Either by inducing localised magnetic moments or causing variation in the site energy, depending on which of the scenarios proposed in [5] is realised). Note that conformational disorder of the terminal ethylene groups of κ -ET₂Cu[N(CN)₂]Br can be controlled by varying the cooling rate and this leads to variations in T_c which are well described by the AG formula [5].

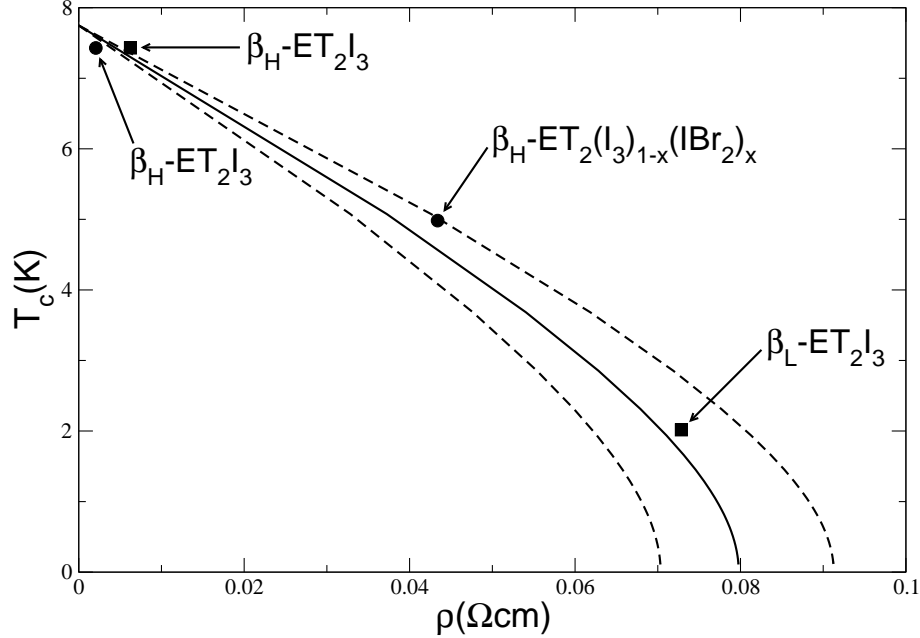


FIG. 1: The variation of the superconducting transition temperature of $\beta_{\text{H}}\text{-ET}_2\text{I}_3$ with the interlayer resistivity. The prediction (solid line) is made from equation (3) taking the measured interlayer hopping integral, t_{\perp} [6]. The dotted lines show the effect of changing t_{\perp} by one standard deviation. The critical temperature of a pure sample, T_{c0} , is that found from fast electron irradiation experiments [5, 7], we do not include the effects of the errors in this measurement. By way of comparison we show data for impurities induced in $\beta_{\text{H}}\text{-ET}_2\text{I}_3$ by fabricating the alloy $\beta\text{-ET}_2(\text{I}_3)_{1-x}(\text{IBr}_2)_x$ [8] (circles). This data is consistent with our prediction. The squares are the data [1] for both the β_{H} and β_{L} phases. However, as the size of the samples was not reported the value of the resistivity contains one free parameter, the relative dimension of the sample, A/d . However, this data does show that the relative change in resistivity between the β_{L} and β_{H} phases is consistent with our hypothesis that the disorder required to stabilise the incommensurate lattice fluctuations observed in the β_{L} phase lowers T_c by increasing the quasiparticle scattering rate, $1/\tau$.

Unfortunately, we are not aware of any reports of the resistivity of $\beta_{\text{L}}\text{-ET}_2\text{I}_3$ with which to test our hypothesis. For example Ginodman *et al.* [1] reported the change in resistance observed between the β_{H} and β_{L} phases, but did not report the size of their crystals. Taking the relative dimensions of their sample as a fitting parameter we find good agreement with our prediction (see figure 1; based on $A/d = 0.4$ cm, where A is the cross-sectional area of the sample and d is the length of the sample in the c -axis, this is actually a fairly typical relative dimension for samples of this material). However, as we do not know the size of the sample this does not represent a very stringent test of our hypothesis. The resistance data of Ginodman *et al.* is well described by the Fermi liquid form $R(T) = R_0 + AT^2$ in both the β_{L} and β_{H} phases with the same value of A in

both phases. This suggests that major difference between the two phases is the scattering rate due to impurities, in support of our hypothesis.

Systematic measurements of the resistivity of β -ET₂I₃ in both the β_L and β_H phases are required to test our hypothesis. If this were done for several samples with varying amounts of disorder due to random imperfections produced during the fabrication process then it would be possible to map out the entire curve of figure 1 for the β_H phase. Clearly only the lower part of the curve could be mapped out for the β_L phase because of the disorder in the terminal ethylene groups.

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