The origin of the difference in the superconducting critical temperatures of the

$\beta_H$ and $\beta_L$ phases of (BEDT-TTF)$_2$I$_3$

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Incommensurate lattice fluctuations are present in the $\beta_L$ phase ($T_c \sim 1.5$ K) of ET$_2$I$_3$ (where ET is BEDT-TTF - bis(ethylenedithio)tetrathiafulvalene) but are absent in the $\beta_H$ 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 $\beta$-ET$_2$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 $T_c$ ($\sim 7$ K) is observed. The low $T_c$ state ($P \lesssim 1$ kbar) is denoted the $\beta_L$ phase and the high $T_c$ state ($P \gtrsim 1$ kbar) is labelled the $\beta_H$ phase. When the pressure on the $\beta_H$ phase is decreased the material does not return to the $\beta_L$ phase but rather $T_c$ is seen to further increase. Below $T \sim 130$ K the resistivity of the $\beta_H$ phase is found to undergo a discontinuous decrease while no such anomaly is found in the $\beta_L$ phase.$^1$. Incommensurate lattice fluctuations have been observed in the $\beta_L$ phase but they are absent in the $\beta_H$ 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 $\beta_L$ and $\beta_H$ 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 \cite{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:

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

(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 $\beta$-ET$_2$I$_3$, the residual interlayer resistivity is given by

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

(2)

where $m^*$ is the quasiparticle effective mass, $c$ (= 15.291 Å for $\beta$-ET$_2$I$_3$ \cite{4}) is the interlayer lattice constant and $t_\perp$ is interlayer hopping integral. Substituting (2) into (1) one finds that

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

(3)

from quantum oscillation Wosnitza et al. \cite{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 $E_F = \hbar^2k_F^2/2m^*$ one finds that $t_\perp = 0.59 \pm 0.08$ meV. $T_{c0} = 7.75$ K is found from fast electron irradiation experiments \cite{5, 7}.

In figure 1 we plot $T_c$ as a function of $\rho_0$. We stress that there are no free parameters in this plot once the data from Wosnitza et al. and Forro et al. \cite{5, 7} is considered. For comparison we also plot the data of Tokumoto et al. \cite{8} who deliberately induced impurities in $\beta$-H-ET$_2$I$_3$ by fabricating the alloy $\beta$-ET$_2$(I$_3$)$_{1-x}$(IBr$_2$)$_x$. 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 $\beta_L$ phase \cite{3}, but absent in the $\beta_H$ 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 \cite{5} is realised). Note that conformational disorder of the terminal ethylene groups of $\kappa$-ET$_2$Cu[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 \cite{5}. 

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FIG. 1: The variation of the superconducting transition temperature of $\beta_H$-ET$_2$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 $\beta_H$-ET$_2$I$_3$ by fabricating the alloy $\beta$-ET$_2(I_3)_{1-x}(IBr_2)x$ (circles). This data is consistent with our prediction. The squares are the data [1] for both the $\beta_H$ and $\beta_L$ 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 $\beta_L$ and $\beta_H$ phases is consistent with our hypothesis that the disorder required to stabilise the incommensurate lattice fluctuations observed in the $\beta_L$ phase lowers $T_c$ by increasing the quasiparticle scattering rate, $1/\tau$.

Unfortunately, we are not aware of any reports of the resistivity of $\beta_L$-ET$_2$I$_3$ with which to test our hypothesis. For example Ginodman et al. [1] reported the change in resistance observed between the $\beta_H$ and $\beta_L$ 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 $\beta_L$ and $\beta_H$ 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 $\beta$-ET$_2$I$_3$ in both the $\beta_L$ and $\beta_H$ 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 $\beta_H$ phase. Clearly only the lower part of the curve could be mapped out for the $\beta_L$ phase because of the disorder in the terminal ethylene groups.

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