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Bulk superconducting specific-heat anomaly in β-di[bis(ethylenedithio) tetrathiafulvalene] diiodoaurate [β-$(ET)_2AuI_2$]

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In order to determine how universal the lack of a specific-heat anomaly at T_c is in the β-$(ET)_2X_3$ [β-$(C_{10}S_8H_8)_2X_3$, where X_3 is the linear anion] ambient-pressure organic superconductors [no anomaly has been found in β-$(ET)_2I_3$], we have measured the low-temperature specific heat of β-$(ET)_2AuI_2$, with $T_c = 5$ K. Using a high-precision, low-addendum specific-heat platform we have found a distinct, although somewhat broadened, anomaly at T_c in β-$(ET)_2AuI_2$ indicative of bulk superconductivity. Also, C/T $(T \rightarrow 0)$ extrapolates, within the accuracy of our measurements, to zero, indicative of a fully opened gap in the electronic energy levels.

The study of organic superconductors, with the hope of obtaining higher superconducting transition temperatures T_c, has become increasingly active.[1] The highest T_c known for these materials has grown by ~6 K since the discovery[2] of superconductivity ($T_c = 1.5$ K) in β-$(ET)_2I_3$ in 1984, with most of this increase coming in the organic materials classified as the β-$(ET)_2X_3$ salts, which are derived from the bis(ethylenedithio)-tetrathiafulvalene electron-donor system.

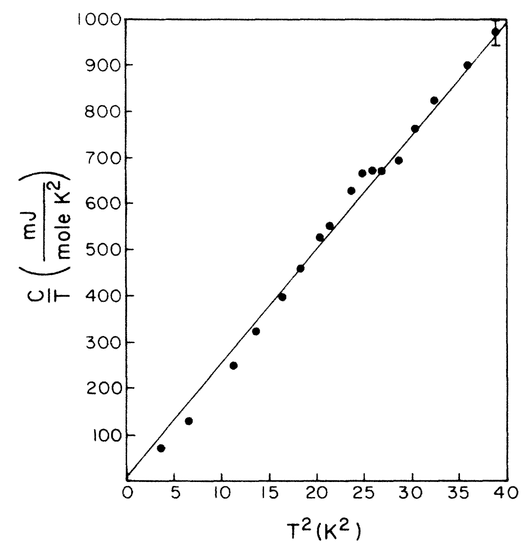

FIG. 1. Specific heat divided by temperature vs temperature squared of three single crystals of β-$(ET)_2AuI_2$ for 1.9 K $< T <$ 6.2 K. The precision of the data is better than ±1%, achieved through a low addendum platform in the calorimeter and extensive signal averaging at each temperature. The absolute accuracy (see error bar) is ±3%. The extrapolation of the normal state data is only approximate and serves more as a guide to the eye to point out the nonlinearity of the superconducting state data. Although the precise transition width is difficult to determine from specific heat data, the data are consistent with the 1-K transition width observed by ac susceptibility.

One of the surprises in the study of the ambient-pressure superconductors of this class has been that, although β-$(ET)_2I_3$ shows convincing evidence (i.e., bulk Meissner effect[3]) of bulk superconductivity, our previously reported specific-heat studies on the *same* crystals showed no corresponding anomaly[4] at $T_c = 1.5$ K or for temperatures as low as 0.7 K.

In the present work we report the low-temperature specific heat C of β-$(ET)_2AuI_2$, $T_c \sim 5$ K (the highest-temperature ambient-pressure superconductor known to date)[5,6] in order to determine whether or not this lack of a specific-heat anomaly is a universal feature in the β-$(ET)_2X_3$ superconductors. The crystals used in these measurements, with a total mass of 5.70 mg, were found to have $T_c = 4.88 \pm 0.05$ K, as determined by the onset in the change in the real part of the ac susceptibility, and a transition width of 1 K. The small mass calorimeter used has been described previously;[7,8] the fraction of the total measured specific heat due to the addenda was 40% at 1.9 K, falling to 12% at 6 K. As seen in Fig. 1, the specific heat of β-$(ET)_2AuI_2$ shows a broadened, but distinct, anomaly at $T \sim 5$ K, and C/T extrapolates to zero as $T \rightarrow 0$, indicating a gap in the electronic energy levels. The absolute value of C/T $(T \rightarrow 0) \equiv \gamma$ in the normal state, proportional to the electronic density of states at the Fermi energy above T_c, is difficult to extrapolate from the normal-state data ($T > 5$ K) due to the extremely large lattice contribution to C in these materials.[4] However, the anomaly observed at T_c in the C/T data is consistent with a γ approximately the size of that observed[4] for β-$(ET)_2I_3$, i.e., $\gamma \sim 20$ mJ/mole K^2. Measurements in a field to suppress T_c and measure the normal state C/T to 1.4 K are planned.

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