Observation of a Non-Magnetic Impurity Effect in the Organic Superconductor (TMTSF)2ClO4

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Abstract. We report a non-magnetic impurity effect on the superconductivity observed in (TMTSF)2ClO4. By varying the sample cooling rate in the vicinity of an anion ordering transition at \( T_{AO} = 24 \) K, we can systematically control the amount of disorder in the sample. We then use dc transport and microwave periodic orbit resonance measurements (closely related to cyclotron resonance) to determine the critical temperature \( T_c \) and the transport scattering time \( \tau \). We find a simple relationship between the two quantities, which strongly suggests that the superconductivity is suppressed by non-magnetic impurities, thus supporting the scenario for unconventional \( p \)-wave superconductivity in this material.

Keywords: triplet superconductivity, non-magnetic impurity effect, anion ordering, TMTSF and quasi-one-dimensional.

PACS: 74.70.Kn, 74.62.Dh, 74.20.Mn, 72.15.Gd

INTRODUCTION

The nature of the superconductivity in the quasi-one-dimensional (Q1D) organic superconductor (TMTSF)2X (X=ClO4, PF6 etc) remains an open-question even though it was first discovered 26 years ago [1]. It was believed for a long time that (TMTSF)2X was a conventional BCS superconductor. However, recent experiments have shown evidence for possible triplet superconductivity, e.g. a divergence of \( H_{c2} \) for X = ClO4 and PF6 [2], no NMR Knight shift through \( T_c \) for X = PF6 [3], and a non-magnetic impurity effect for the X = (ClO4)1-x(ReO4)x alloy [4]. Here we report on a non-magnetic impurity effect associated with disordered ClO4 anions, which undergo an order-disorder transition at a temperature \( T_{AO} = 24 \) K in the X = ClO4 salt. The degree of ordering is sensitive to the cooling rate at \( T_{AO} \). Our investigation is focused on the effect of this disorder on the superconducting critical temperature \( T_c \). We characterize the disorder using a periodic-orbit resonance (POR) technique, which enables us to determine both the Fermi velocity \( v_F \) and the quasiparticle scattering time \( \tau \) in the normal state [5]. We find a direct correlation between \( T_c \) and \( \tau \), suggesting that pair-breaking due to non-magnetic impurities is responsible for the suppression of \( T_c \).

EXPERIMENTS

We separately performed POR and dc transport measurements in order to determine the cooling rate dependence of \( \tau \) and \( T_c \), respectively.

Study of the Periodic-Orbit Resonance

The POR measurements were performed using a millimeter vector network analyzer and a cavity perturbation technique [6]. A 17 T superconducting solenoid with a He flow cryostat was used for magnetic field and temperature control. The field orientation was varied using a rotating cavity [7]. We first used the angle-dependence of the POR to align the sample with the applied field parallel to \( c^* \) [8]. This was carried out in the relaxed state achieved via the slowest cooling rate (0.01 K/min). We then studied...
the cooling rate dependence of the POR, particularly the resonance linewidth from which we determine $\tau$. Prior to each cooling, the sample was heated to 50 K, and then cooled to 15 K at the appropriate rate. The range of cooling rates achievable in the flow cryostat was 0.01 to 37 K/min. In addition to determining $\tau$, we studied the angle-dependence of the POR to ensure that the cooling rate did not affect $v_F$, i.e. the band structure. Fig. 1 shows microwave absorption as a function of magnetic field at different cooling rates; the data were obtained at 1.4 K and 51.8 GHz. The Lorentzian-like POR is centered at $\sim 2$ T. As seen in the figure, the position of the POR is independent of the cooling rate. However, the resonance becomes broader with increasing cooling rate. This confirms that faster cooling leads to increased anion disorder and to an enhancement of the quasiparticle scattering rate. Thus, we can control the concentration of non-magnetic impurities in this way.

**dc Transport Measurements**

The interlayer resistance $R_{zz}$ was measured for various cooling rates using a four-probe low-frequency (17 Hz) lock-in technique. In order to measure the superconducting critical temperature, $T_c$ (onset $T_c \sim 1.2$ K in the relaxed state), we employed a home-built $^3$He cryostat [7]. We employed an identical cooling procedure as used for the POR measurements, achieving cooling rates between 0.03 and 28 K/min in the $^3$He cryostat. The resistance was measured from 50 K down to 0.5 K. Each trace (not shown) displayed a kink at around 24 K indicative of the anion ordering. The resistance data were identical between 50 K and $T_A = 24$ K for different cooling rates. However, they became distinguishable below $T_A$, with higher resistances measured for faster cooling rates. The temperature dependence of $R_{zz}$ was nearly linear below 8 K. Therefore, we estimated the residual resistance $R_{zz}(T = 0 K)$ using a polynomial fit. More importantly, $T_c$ shifted to lower temperatures as the cooling rate was increased. We determined $T_c$ at the completion temperature, where the maximum $dR_{zz}/dT$ and zero resistance lines intersect; the transition width varied from 0.1 to 0.3 K. These tendencies were completely systematic and reproducible for all cooling rates.

**DISCUSSION AND SUMMARY**

By comparing the dc transport and POR data, we find that changes in $R_{zz}$ and $R_{xx}$ can be attributed to changes in the transport scattering time for all but the fastest cooling rates (we speculate that regions of the sample may become insulating for the fastest cooling rates, causing a departure from this behavior). We plot $T_c$ as a function of the pair breaking strength $\alpha$ in Fig. 2. Here we make the assumption that the pair-breaking strength $\alpha (= h/2 \tau_0)$, which is inversely related to the pair breaking time $\tau_0$, is proportional to the transport scattering rate, i.e. $\alpha \propto 1/\tau$. The data in Fig. 2 have been fit to the universal function in the Abrikosov-Gor’kov theory for the impurity effect on $T_c$ [9],

$$\ln \frac{T_c}{T_{c0}} = \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{\alpha}{2\pi K T_{c0}}\right),$$

where $\psi$ is the digamma function and $T_{c0}$ is the critical temperature in the absence of impurities. The good agreement between the data and the fit suggests that the anion disorder suppresses $T_c$, supporting the scenario for unconventional spin-triplet ($p$-wave) superconductivity in (TMTSF)$_2$ClO$_4$.

**ACKNOWLEDGMENTS**

This work was supported by the National Science Foundation (DMR0196461 and DMR0239481).

**REFERENCES**

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