Field-Induced Dimensional Crossover and Anomalous Superconducting States in the Quasi-One-Dimensional Superconductor (TMTSF)$_2$ClO$_4$

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Abstract. Quasi-one-dimensional (Q1D) superconductivity becomes stable in high magnetic fields due to the field-induced dimensional crossover (FIDC) and a stabilization of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) states. We have studied the field-direction and field-amplitude dependence of the onset temperature $T_{c\text{onset}}$ of the superconducting state of the organic superconductor (TMTSF)$_2$ClO$_4$. Comparing the in-plane field-angle dependence of $T_{c\text{onset}}$ and the behavior of the inter-plane resistance, we show that the FIDC stabilizes the superconductivity when the field is more than 19° away from the a axis. In addition, we demonstrate that $T_{c\text{onset}}$ remains finite for $H \parallel a$ up to $H = 50$ kOe, as well as for $H \parallel b'$. The high-field states are anomalous in nature, suggesting that different FFLO states are realized.

1. Introduction

It has been predicted that quasi-one-dimensional (Q1D) superconductors are rich in anomalous superconducting (SC) phenomena. For example, as first predicted by Lebed [1], a 2D electronic state confined in the conductive planes is realized if a strong magnetic field is applied parallel to the sheet-like Fermi surface [2], even though a small 3D coherence may exist in the electronic state in zero field. This phenomenon is called the field-induced dimensional crossover (FIDC). The FIDC weakens the orbital pair-breaking effect of superconductivity when the field is exactly parallel to the conducting plane, allowing superconductivity to survive in higher magnetic fields. Another interesting issue is that a spatially-modulated SC state, called a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [3, 4], can be highly stabilized in Q1D systems. The stabilization of FFLO states is attributed to the nesting property of the sheet-like Fermi surface [5]. The realization of FFLO states can lead to a strong enhancement of superconductivity even for singlet pairing in magnetic fields far above the Pauli-limited field $H_P$ [1, 6], $H_P/T_{c0} = 18.4$ kOe/K [7] for an isotropic gap, where $T_{c0}$ is the zero-field critical temperature.

The organic superconductor (TMTSF)$_2$X [8, 9], where TMTSF stands for tetramethyl-tetraselena-fulvalene and X is an anion such as ClO$_4^-$, PF$_6^-$, is an archetypal Q1D system being the most conductive along the a axis and the second most conductive along the $b'$ axis. The
The extrinsic increase of temperatures is subtracted. at 30 kOe for various field directions (in Fig. 1. Recent studies have revealed that the SC state in (TMTSF) \textsubscript{2}PF\textsubscript{6} and (TMTSF)\textsubscript{2}ClO\textsubscript{4} survives in magnetic fields much higher than \(H_P\) [10, 11, 12, 13].

We have recently discovered the unconventional field-angle dependence of the onset temperature of superconductivity \(T_c^{\text{onset}}\) in (TMTSF)\textsubscript{2}ClO\textsubscript{4}, for magnetic fields parallel to the \(ab\) plane [12]. In addition, we have revealed an anomalous dependence of \(T_c^{\text{onset}}\) on the impurity scattering and on a small tilt of the magnetic field out of the \(ab\) plane [13]. These findings can be explained by the scenario of a FIDC stabilizing superconductivity when the magnetic field is more than 19-degree away from the \(a\) axis and of a realization of two different FFLO states for \(H \sim a\) and \(H \sim b'\).

In this article, we present several new figures supporting the FIDC and FFLO scenarios.

2. Experiment
We measured the \(c^*\)-axis (perpendicular to the \(ab\) plane and along the least-conducting direction) resistance of high-quality single crystals of (TMTSF)\textsubscript{2}ClO\textsubscript{4}. The magnetic field is applied using the “Vector Magnet” system [14], which allows a precise 3D control of the field direction. The direction of the crystalline axes are determined from the anisotropy of \(H_{c2}\) and from the angular magnetoresistance oscillations (AMRO). Details of the experimental process have been presented in Refs. [13, 15]. We define by \(\phi\) the azimuthal angle within the \(ab\) plane measured from the \(a\) axis, as illustrated in Fig. 1.

We present here data for Sample #1 in Ref. [13], which is very clean as evidenced by its large mean free path \((l \sim 1.6 \text{ \mu m})\), as well as data for Sample #2, which is not as good in quality since \(l \sim 0.6 \text{ \mu m}\).

3. Field-Induced Dimensional Crossover and Superconductivity
First, we present in Fig. 1 the temperature dependence of the out-of-plane resistance \(R_{c^*}\) of Sample #1 at 30 kOe for various magnetic field directions within the \(ab\) plane. We note that, in this figure, the extrinsic small increase of resistance at low temperatures [12, 13], possibly due
to small cracks in the sample, is subtracted. Let us first focus on the temperature dependence of \( R_c(T) \) in the normal state, i.e. above 0.7–0.8 K. The normal-state inter-layer resistance \( R_c(T) \) exhibits a metallic temperature dependence for \( H \parallel a \) (i.e. \( \phi = 0^\circ \)) indicating an anisotropic 3D conducting state, whereas it shows non-metallic behavior for \( H \parallel b' \) (\( \phi = 90^\circ \)) reflecting the realization of a 2D electronic state confined in the \( ab' \) planes. It is clear that the crossover between the metallic and the non-metallic behavior takes place around \( \phi = 18–20^\circ \). Therefore, this angle, \( \phi_{3D-2D} \equiv 18–20^\circ \), can be regarded as the onset angle of the FIDC.

In order to compare the dimensionality of the electronic state and the field-angle dependence of \( T_c^{\text{onset}} \), whose definition is given in Refs. [12, 13, 15], we present \( T_c^{\text{onset}}(\phi) \) with the contour plot of \( R_c(T, \phi) \) at 30 kOe in Fig. 2(a), and \( T_c^{\text{onset}}(\phi) \) with the contour plot of the temperature derivative \( dR_c/dT(T, \phi) \) divided by \( R_c \) in Fig. 2(b). The contour plots shows convincingly that the behavior of \( R_c(T) \) in the normal state is different between \( \phi < \phi_{3D-2D} \) and \( \phi > \phi_{3D-2D} \). Interestingly, \( T_c^{\text{onset}}(\phi) \) displays unusual dips around \( \phi \sim \pm 17^\circ \). It is evident that the dips locate close to \( \phi_{3D-2D} \) and that \( T_c^{\text{onset}} \) is enhanced in the 2D region. The comparison between \( T_c^{\text{onset}}(\phi) \), \( R_c(T, \phi) \), and \( dR_c/dT(T, \phi) \) strongly support the following scenario for an enhancement of \( T_c^{\text{onset}} \) in high fields. First, when the angle \( \phi \) is larger than \( \phi_{3D-2D} \), the FIDC stabilizes superconductivity. Secondly, another stabilization comes into play as long as the field is closely aligned along the \( a \) axis. Combining two effects leads in turn to the dips of \( T_c^{\text{onset}} \) around 18°.

4. High-Field Superconducting States

We have observed that the SC state for both \( H \parallel a \) and \( H \parallel b' \) remains stable far above \( H_P \), which we estimated as 23–26 kOe for the present sample [13]. We revealed that the SC states in high fields are anomalous in nature based on the following observations displayed in Fig. 3:

(i) For \( H \parallel b' \), the \( T_c^{\text{onset}}(\phi) \) curve looses its mirror symmetry with respect to the \( b' \) axis above 30 kOe, and a new principal axis \( X \) of \( T_c^{\text{onset}}(\phi) \) appears [12].

(ii) For both \( H \parallel a \) and \( H \parallel b' \), the SC state above 40 kOe is destroyed by little impurities [13].

(iii) For \( H \parallel a \), the SC state above 30 kOe is easily suppressed by a small (\( \sim 3^\circ \)) tilt of the magnetic field out of the \( ab' \) plane whereas the SC state for \( H \parallel b' \) is only weakly unstable against a tilt of the magnetic field out of the \( ab' \) plane [13].

It is evident in Fig. 3 that the SC state above \( H_P \) is highly anomalous. The result (i) indicates a change in the spatial symmetry of the SC state. For an FFLO state in a Q1D system, it is likely that the wavevector of the modulation of the order parameter, \( q_{\text{FFLO}} \), is fixed to a certain direction determined by the Fermi surface geometry. This fixed \( q_{\text{FFLO}} \) can be a reason for the change of the spatial symmetry. The result (ii) is consistent with the prediction that an FFLO state is sensitively destroyed by a small amount of impurities [16]. Different behaviors against a tilt of the magnetic field (iii) suggests that the high-field states for \( H \parallel a \) and \( H \parallel b' \) have different natures, possibly due to the difference of the angle between \( H \) and \( q_{\text{FFLO}} \). It is worth noting that \( T_c^{\text{onset}} \) below which the anomalous behavior was observed is close to the theoretical tricritical temperature of an FFLO state \( T^\dagger \approx 0.56T_{c0} \) [16], approximately 0.8 K in the present case as shown in Fig. 3. This fact may also support FFLO scenarios.

5. Summary

To summarize, we have established a correspondence between the FIDC and the enhancement of superconductivity at low temperatures. The existence of an other enhancement of superconductivity when \( H \) is close to the \( a \) axis in the \( ab' \) plane results in dips of \( T_c^{\text{onset}}(\phi) \) near \( \phi_{3D-2D} = 18–20^\circ \). The stabilization of superconductivity for \( H \parallel a \), likely due to an FFLO state, is easily suppressed by a tilt of the field off the \( ab' \) plane. In conclusion, we have revealed that the SC states above \( H_P \) and below \( T^\dagger \) are highly anomalous. The most plausible explanation so far is that different FFLO states are realized for \( H \parallel a \) and for \( H \parallel b' \).
Figure 3. Summary of our experimental results for the SC states for $H \parallel a$ (left panel) and for $H \parallel b'$ (right panel). The estimated values of $H_p \sim 26$ kOe and the theoretical tricritical temperature $T^\dagger \simeq 0.56T_{c0} \sim 0.8$ K are shown with the arrows.

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