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Resonant angular magnetoresistance in the ac-plane of the Bechgaard salts: comparative studies

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Abstract. Lebed resonance phenomenon which manifests sharp resistance minima at angles where the magnetic field imposes electrons a periodic motion on the Fermi surface, is readily observed in most of Bechgaard salts such as $(TMTSF)_2ClO_4$, $(TMTSF)_2PF_6$, $(TMTSF)_2ReO_4$. On the other hand, another type of oscillatory behavior, observed when the magnetic field rotates from the most conducting direction towards the least conducting direction, in the *ac*-plane, is found to vary much from one compound to another, having been clearly observed only in $(TMTSF)_2ClO_4$. We have performed a systematic study of both the *bc*- and *ac*-rotation measurements and report that the *ac*-plane resonances are also omnipresent and universal in all the three compounds. Difference among compounds, notably the strength of the coherence peak may be used to account for the evolution of coherence from one compound to another.

1. Introduction

Physical properties of low-dimensional metal are very sensitive to the Fermi surface morphology. Angle-dependent resistance under a constant magnetic field, or angular magnetoresistance, is particularly sensitive to the details of the Fermi surface such as amplitude and direction of warping, electronic dimension, etc. Various oscillatory and resonant angular magnetoresistance has attracted great attention especially in the quasi-one-dimensional electron system of the Bechgaard salts, $(TMTSF)_2X$. The best studied is the Lebed resonance phenomenon which manifests sharp resistance minima at angles where the magnetic field imposes electrons a periodic motion on the Fermi surface[1]. It is readily observed in most of Bechgaard salts such as $(TMTSF)_2ClO_4$[2, 3], $(TMTSF)_2PF_6$[4], $(TMTSF)_2ReO_4$[5]. On the other hand, another type of oscillatory behavior is also observed when the magnetic field rotates from the most conducting direction towards the least conducting direction, in other words, in the *ac*-plane[6]. It has been interpreted as a topological effect of quasi-one-dimensional Fermi surface and could be used to estimated the band parameter t_b. However, distinctively from the Lebed resonance, the oscillatory behavior in the *ac*-plane is found to vary much from one compound to another[7].

There are three interesting members of Bechgaard salts in which the resonant effects of angular magnetoresistance are extensively studied. $(TMTSF)_2ClO_4$ becomes superconducting at ambient pressure, which is unique in Bechgaard salts. $(TMTSF)_2PF_6$ needs about 6.5 kbar to avoid the spin-density-wave formation and to become a superconductor. $(TMTSF)_2ReO_4$ needs even higher pressure of about 8.5 kbar to avoid an insulating state derived from the three dimensional anion ordering.

Although Lebed resonances are omnipresent in three compounds at angular positions predicted from the various calculations including Lebed's original prediction[1], the resonances in the ac-rotation is clearly reported only for $(TMTSF)_2ClO_4$[6]. We report in this paper that the ac-plane resonances are also omnipresent and universal in all the three compounds. Difference among compounds, notably the strength of the coherence resistance peak may be used to account for the evolution of coherence from one compound to another.

2. Experiments

A unique and fully motorized double axis rotator probe was built to allow a moderate sized pressure cell (14 mm in diameter and 20 mm long) to turn three-dimensionally in an also home-built variable temperature insert (32 mm in sample space diameter), which was in turn introduced into a standard superconductor solenoid with 52mm bore. The lowest temperature obtained in a pumped helium-4 bath was 1.5 K. Tiny fragments of samples (about 0.5 mm long along the a-axis) cut from a bar-like crystal were used. Electrical contacts with annealed gold wires of 20 μm in diameter were made with the carbon paste. Ac current of 10 μA was applied along the interlayer direction (along the c^*-axis) and the voltage across the sample was measured using a lock-in amplifier tuned to low frequency (between 70 and 80 Hz). Pressure was generated in a clamp-type miniature BeCu cell with the Daphne 7373 oil as a pressure transmitting medium. In this way, the samples, contacts, and pressure were stable for more than a year even at ambient temperature. The pressure of each sample at low temperature was estimated from the superconductivity transition temperature of high-purity (99.999 %) tin wire sample embedded near the samples. Smooth rotation could be monitored through two Toshiba-116 Hall sensors mounted on the pressure cell. Temperature increase due to friction is less than 0.01 K and returned fast to equilibrium in the pumped liquid helium bath. In most cases, the sample resistance was an excellent indicator of the angular position, showing well defined resistance minima at required positions whenever the magnetic field lay in the conducting plane.

3. Results

Figure 1 shows angular magnetoresistance of three different Bechgaard salt samples, $(TMTSF)_2ClO_4$, $(TMTSF)_2PF_6$ and $(TMTSF)_2ReO_4$ under the pressure of 1 bar, 8.4 and 10.3 kbar, respectively. For the last two compounds, pressure values are selected to maintain the samples in the metallic state for the entire angular range in the bc-rotation at given magnetic fields. Left panel shows data from the bc-rotation and reflects anomalies mainly due to Lebed resonances. Angles for resonances are similar between $(TMTSF)_2PF_6$ and $(TMTSF)_2ReO_4$, but much larger for $(TMTSF)_2ClO_4$ due to the superstructure formed by anion ordering. As is evident from the figure, resistance dips of Lebed resonances is the weakest in $(TMTSF)_2ClO_4$, and the largest in $(TMTSF)_2ReO_4$. This observation is valid even if we consider the difference of magnetic field used. What is interesting in this figure is that the background magnetoresistance has universal behavior irrespectively of samples, that is, it has a broad minimum around the c^*-axis, increases as $|\theta|$ increases, and develops a sharp minimum as the magnetic field approaches the conducting planes. None of the current model has attempted to account for this abnormal enhancement of interlayer conduction.

The right panel of Fig. 1 shows the ac-rotation of angular magnetoresistance. Larger field than that for the bc-rotation is intentionally used to observe clearer structure. The high magnetic field does not matter because the feature we are interested in the ac-rotation is restricted in the narrower angular range around $\theta = 90$ degree than those for the bc-rotation. First, the data for $(TMTSF)_2ClO_4$ corresponds well with the published data at 8 T. Both the resonance peaks and the coherence peak are well developed. The latter signifies that the interlayer transport in this compound is coherent at 1 bar and at low temperature. This point also agrees with the monotonous decrease of interlayer resistance with temperature over the whole temperature range. In comparison, the interlayer resistance of $(TMTSF)_2PF_6$ at 1 bar has a broad maximum whose temperature and amplitude decreases with pressure before a monotonous decreasing behavior

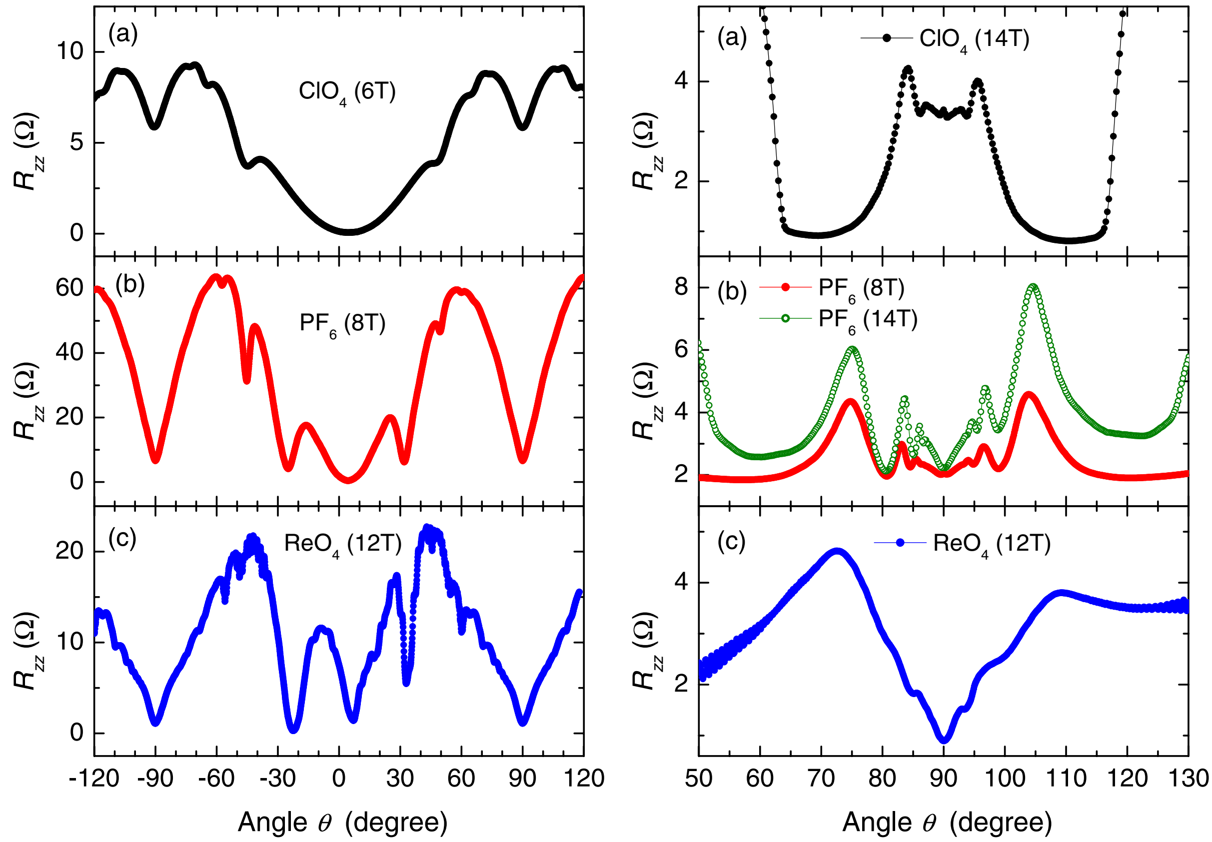

Figure 1. Comparison of angular magnetoresistance with a magnetic field in the *bc*-plane (left panel) and in the *ac*-plane (right panel). In each panel, curves are for (a) $(TMTSF)_2ClO_4$ ($P =$ 1 bar), (b) $(TMTSF)_2PF_6$ ($P = 8.4$ kbar) and (c) $(TMTSF)_2ReO_4$ ($P = 10.3$ kbar) are shown with the magnetic field strength that was used. For the *bc*-rotation, the magnetic field strength was maintained so that the samples lie in the metallic phase for the entire angular range. For the *ac*-rotation, high magnetic field was intentionally used to reveal the detail more clearly. Slight asymmetry, notably in the *ac*-rotation is due to the low symmetry of the crystal lattice. Sharp increase of resistance at positions indicated with arrows represents the sample enters into the nonmetallic FISDW states.

settles at high pressure. It is striking that the *ac*-rotation anomalies are far better developed in $(TMTSF)_2PF_6$ than in $(TMTSF)_2ClO_4$. The quality of our data is far superior to what was first claimed as such result in $(TMTSF)_2PF_6$, where only the second derivative showed some structures. But, the coherence peak is very much reduced and can barely be detected in the V-shaped angular resistance around $\theta = 90$ degree.

By the way, the cooling curve of $(TMTSF)_2ReO_4$ shows well established monotonic temperature decrease over the whole temperature range and at the same time shows the largest Lebed resonances. However, it shows a pronounced V-shaped angular resistance around $\theta = 90$ degree on which weak anomalies, presumably the same resonances as $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$ in the *ac*-rotation, are barely visible. No coherence peak was observed up to 14 T. Nonetheless, the Lebed resonances are the most pronounced among the three compounds.

4. Discussions

It is shown that the Lebed resonances arise both in coherent and in incoherent interlayer transport regime[8, 9, 10, 11]. On the other hand, the existence of the coherence peak has been suggested as a concrete evidence of the coherent interlayer transport[8]. In this sense, our data suggest that the interlayer transport is the most coherent in $(TMTSF)_2ClO_4$ and the least coherent in $(TMTSF)_2ReO_4$ over the temperature and magnetic field range of interest as far as only the coherence peak is concerned. But, this also suggests that the *bc*-rotation anomalies develops better in a less coherent sample, which is not so easy to reconcile with the existing theory. On the other hand, the *ac*-rotation anomalies are the best developed in $(TMTSF)_2PF_6$, which is in the middle in coherence. Any of the current models discussed quantitatively the relation between the degree of coherence and the strength of the resonance. So, it is too premature to determine the order of coherence among the three samples studied.

It would be interesting to observe the trend of angular magnetoresistance and compare it with other parameters like critical pressure for superconductivity. It would also be necessary to apply or adjust pressure to compare the data at the same magnetic field strength to draw a meaningful conclusion. However, it is prerequisite to have crystal lattice parameters and band parameters under pressure and at low temperature to complete the analysis. Such data are hardly available at present.

The extremely high quality *ac*-rotation oscillations of the $(TMTSF)_2PF_6$ sample allow us to verify its mechanism suggested in Ref. [6]. They suggested that the condition of the maximum satisfies $\tan\theta_n = 2t_b c/\gamma_n \hbar v_{\mathrm{F}}$ where θ_n is the angular position of n-th maximum measured from the a-axis and γ_n n-th zero of the Bessel function $J_0(x)$. Preliminary analysis shows that $\tan\theta_n$ increases linearly with $1/\gamma_n$ with a coefficient 0.59–0.65, which will give t_b provided the Fermi velocity is known.

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