

**Pressure dependence of the angular magnetoresistance of (TMTSF)<sub>2</sub>PF<sub>6</sub>**

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An anomalous crossover has been reported in the background of angular magnetoresistance of (TMTSF)<sub>2</sub>PF<sub>6</sub> as pressure increases. When a magnetic field rotated in the plane perpendicular to the *a*-axis, the background of the interlayer resistance  $R_{zz}$  was concave around *c'*-axis at low pressure but became convex above 10 kbar. However, our extensive experiments revealed that only one form of background resistance occurred in a sample regardless of pressure. The deceptive crossover may be attributed to the possible mixing of intralayer resistivity component(s) to the interlayer resistivity component.

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**I. INTRODUCTION**

Extensive experimental and theoretical studies have been made on the angular magnetoresistance oscillations in various organic conductors with low-dimensional electronic structures. Several distinct resonance effects have been reported depending on the rotating plane of magnetic field in a sample and on the sample dimensionality,<sup>1-4</sup> and have given deeper understanding of electron transport.

One of them, the Lebed resonance has been the most studied in the series of Bechgaard salts where the resistance shows dips when the field is parallel to one of the lattice vectors in the *b'**c*\* plane.<sup>1,2,5</sup> Although it was initially suggested to be a commensurability effect between two magnetic lengths along *k<sub>b</sub>* and *k<sub>c</sub>*, which is anticipated in the longitudinal resistance  $R_{xx}$ ,<sup>6,7</sup> the resonance is usually better resolved in the interlayer resistance  $R_{zz}$ .<sup>8</sup> Several scenarios have been suggested in terms of semiclassical Boltzmann transport theory,<sup>9,10</sup> electron-electron interaction,<sup>11</sup> hot<sup>12</sup> or cold spots<sup>10</sup> on the Fermi surface, Fermi-liquid–non-Fermi-liquid (FL-NFL) transition,<sup>13</sup> etc.

Experimentally, an anomalous pressure dependence has been reported in the background of  $R_{zz}$  in (TMTSF)<sub>2</sub>PF<sub>6</sub>. Considering only the background,  $R_{zz}$  at relatively low pressure has a minimum around *c*\* direction, increases as the field moves toward  $\pm b'$  direction, and develops a sharp minimum around the  $\pm b'$  directions.<sup>14,15</sup> (We will call such a behavior as type I.) On the other hand, at high pressure, angular magnetoresistance has a  $(\cos)^\alpha$ -like ( $1 < \alpha < 2$ ) background superposed by resonance dips around *c'* direction,<sup>15,16</sup> much similar to that of the intraplane resistance  $R_{xx}$ . (Type II behavior.) A crossover from type I to type II has been reported to take place somewhere between 6.0 and 8.3 kbar<sup>14,16</sup> or between 8.5 and 9.2 kbar.<sup>15</sup>

Chashechkina and Chaikin suggested that the zero-field interlayer conductivity could be described as the sum of contribution from all hopping directions between neighboring molecular chains which is independently killed by a perpendicular field.<sup>15</sup> This model is strongly supported by the observation of only odd-indexed Lebed resonance in (TMTSF)<sub>2</sub>ReO<sub>4</sub>.<sup>17</sup> With an additional anomalous in-plane contribution of the order of  $t'_b$ , the unnested bandwidth, which is destroyed by a field perpendicular to the *b* axis, they

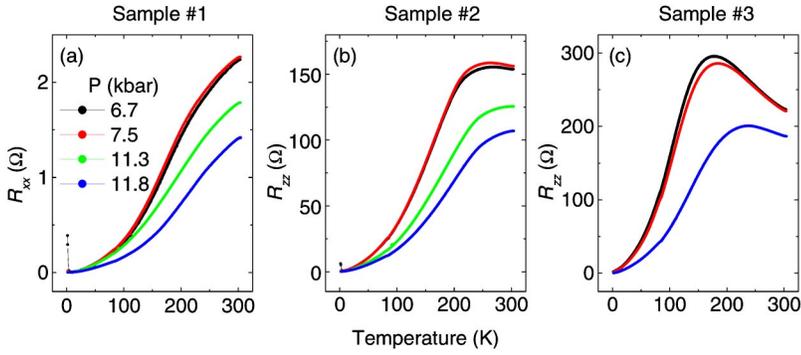
could not only successfully fit the angular dependence at fixed fields but also explain the crossover from type I to type II background magnetoresistance.<sup>15</sup>

However, the type II background of  $R_{zz}$  does hardly conciliate any semiclassical theory,<sup>9,10,18</sup> while it is at the center of the FL-NFL transition theory.<sup>13,19</sup> In the simplest form, the Lorentz force is always zero for field parallel to a current and maximum for field perpendicular to the current, which favors the type I behavior for  $R_{zz}$ . Such an effect can be ignored only when the coherence in the *c* direction is wiped out. Therefore, the crossover of background magnetoresistance signifies a drastic change of electron-transport mechanism even at a moderate pressure.

We performed a systematic study of angular dependent interlayer resistance  $R_{zz}$  under various pressures as well as magnetic fields to elucidate the mechanism of the crossover. Special attention has been paid to perform as many different pressure experiments as possible with the same sample in order to exclude any ambiguity which might be caused by sample dependence. Up to three samples were measured together in the same pressure cell. The experimental result describes that the sample dependence rather than the pressure dependence is more plausible for the observed crossover.

**II. EXPERIMENTS**

Small pieces of samples were cut from high-quality single crystals. Dupont 4929N silver paste assured electric connections between 20  $\mu\text{m}$  gold wires and samples. A miniature self-clamped pressure cell made of CuBe alloy has been used through experiments with a pressure medium of 1:1 mixture of Daphne 7373 oil and Kerosene oil.<sup>20</sup> The lowest temperature of 1.5 K was realized in the pumped liquid helium bath. The electrical resistance was measured by the standard four-probe method with lock-in amplifiers tuned at low frequency. In order to avoid any ambiguity due to sample dependence and pressure uncertainty, three samples, one with  $R_{xx}$  and two with  $R_{zz}$  configurations, were put side by side in the same pressure cell and measured simultaneously. Additional verification was made with two more samples, one for  $R_{xx}$  and the other for  $R_{zz}$  in another pressure cell. Four different pressure experiments were performed with the first set of samples, and three with the second set. Pressures at low temperature were calibrated independently using the supercon-



ducting transition of pure tin. The pressure values throughout this paper are those at low temperature. The pressure applied at room temperature was higher by 1.5–2 kbar depending on pressure range.

### III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of resistance of the first three samples, one for  $R_{xx}$  and two for  $R_{zz}$ . Measurements were performed in the order of 11.3, 7.5, 6.7, and 11.8 kbar.  $R_{xx}$  decreases monotonically with temperature regardless of pressure while  $R_{zz}$  of the sample no. 3 develops a pronounced maximum at an intermediate temperature. As pressure increases, the amount of resistance increase lessens and the temperature of maximum resistance ( $T_{max}$ ) shifts toward higher temperature with a good agreement with the results in Refs. 21 and 22. Although resistances of both  $R_{zz}$  samples rise with cooling, the ratios of resistance at maximum to that at room temperature are very much different from each other, by 1.02 and 1.30, respectively at 7.5 kbar. Electron motions along two conducting chains are incoherent above  $T_{max}$  and reduction of the interchain electron tunneling along  $c$  axis is responsible for the resistance increase on cooling.<sup>22</sup> Therefore, it is bizarre that  $R_{zz}$  of sample no. 2 shows only a little change with lowering temperature.

At 6.7 kbar, the lowest pressure studied, the resistance shows slight upturn at low temperature, which indicates that the pressure is very close to the border between spin-density wave and superconducting states. Superconducting transition was not verified because the lowest temperature was 1.5 K in this study. However, the occurrence of the field-induced spin-density wave states was checked at the pressure of 6.7 kbar. At other pressures, the threshold field for the field-induced spin-density waves was higher than 8 T, the maximum field of the current study.

Presented in Fig. 2 is angular magnetoresistance of the three samples measured with a field rotating in the  $b'c^*$  plane at four different pressure values. All the data except the first row were obtained at a fixed field of 8 T. At 6.7 kbar, the pressure is slightly so low that a field-induced spin-density wave state is beginning to develop already at 8 T and the angular magnetoresistance under 6 T is presented instead. Difference of angular magnetoresistance among samples is obvious. While the type II background (superposed by Lebed resonance dips) is dominant for the first two samples, resistance for the sample no. 3 is dominated by the type I back-

FIG. 1. (Color online) Temperature dependence of resistance for the first three samples under four different pressures.  $R_{xx}(T)$  decreases monotonically with temperature while  $R_{zz}(T)$  is characterized with a maximum at intermediate temperature. Two graphs of  $R_{zz}(T)$  are contrasted by the amount of resistance increase at maximum.  $R_{max}/R_{RT}=1.02$  and 1.30, respectively for (b) and (c) under 7.5 kbar. Although the curves at 6.7 kbar and at 7.5 kbar are quite similar, the latter could be distinguished by the absence of the field-induced spin-density-waves until 8 T.

ground. At this point, we need to remind that all the measurements were carried out with the same set of samples over the broadest pressure range ever studied, yet there is no pressure induced crossover in the background of  $R_{zz}$ . Instead, two characteristic backgrounds are independently observed in two different samples for all over the pressure range; the sample no. 2 conserves the type II background whereas the sample no. 3 the type I background until 11.8 kbar. This is a clear contrast to the previous reports<sup>14–16</sup> where a pressure-induced crossover from type I to type II background was mentioned.

An additional experiment confirmed the above finding. Presented in Fig. 3 are cooling curves and angular magnetoresistance of two more samples, which were arranged to measure  $R_{xx}$  (sample no. 4) and  $R_{zz}$  (sample no. 5) simultaneously in another pressure cell. There was an interval of

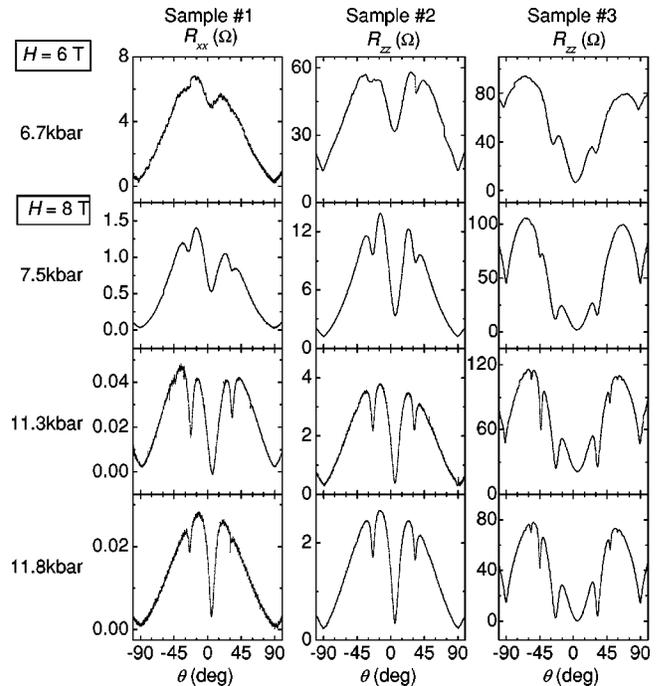


FIG. 2. Angular dependent magnetoresistance for three samples in the same pressure cell under four different pressures. The same set of samples was temperature cycled for all the four different pressure values with cooling curves displayed in Fig. 1. Notice that two  $R_{zz}$  samples show different backgrounds all over the pressure range.

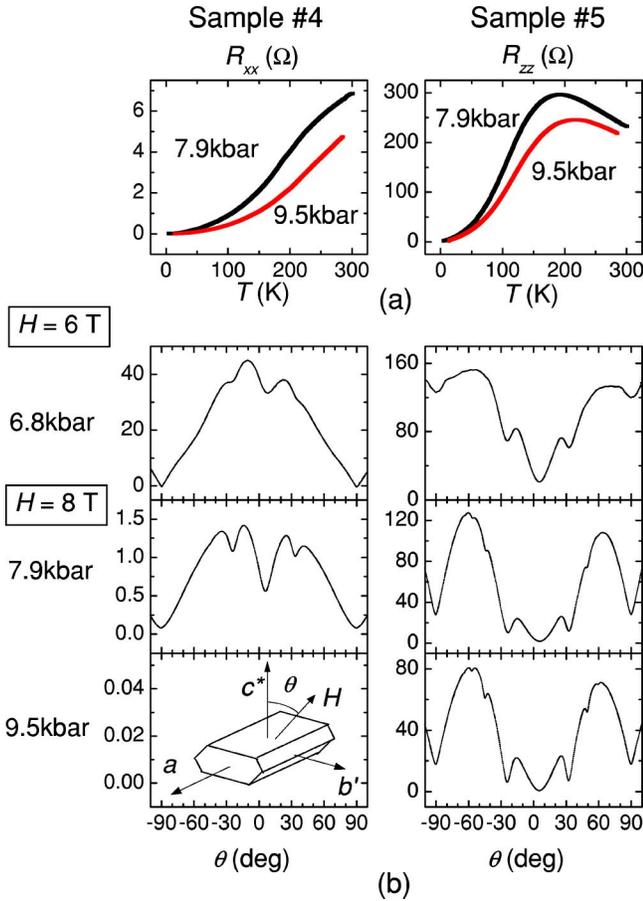


FIG. 3. (Color online) Data for two additional samples studied independently. (a) Temperature dependence of resistance. They correspond very well with the characteristics of sample no. 1 and sample no. 3, respectively. For  $R_{zz}$ ,  $R_{\max}/R_{RT} = 1.27$  and  $1.18$  at  $7.9$  and  $9.5$  kbar respectively. (b) Angular dependent magnetoresistance. There is no crossover in  $R_{zz}$  between  $6.8$  and  $9.5$  kbar, confirming the result of Fig. 2. The cartoon in the lower left box explains the symbols used in this paper.

more than a year between two experiments. Again,  $R_{xx}$  decreases monotonically with temperature while  $R_{zz}$  has a maximum around  $180$  K. The height of resistance maximum characterized by  $R_{\max}/R_{RT}$  is  $1.27$  at  $7.9$  kbar and decreases to  $1.18$  at  $9.5$  kbar, very similarly to sample no. 3 of the first experiments. In Fig. 3(b), resistances under three pressures were presented either under  $6$  T or under  $8$  T due to the same reason as in the first set of experiments. It is clearly evidenced that there is again no crossover from type I to type II background for  $R_{zz}$  between  $6.8$  and  $9.5$  kbar.

Now, it is necessary to explain the behavior of sample no. 2. First, although its electrical wires were arranged to measure  $R_{zz}$ , the angular magnetoresistance background is akin to that of  $R_{xx}$ . We also notice that the temperature dependence of  $R_{zz}$  of sample no. 2 lies between that of  $R_{xx}$  (sample no. 1) and that of  $R_{zz}$  (sample no. 3). Increase of resistance down to  $T_{\max}$  is merely  $2\%$  even at the lowest pressure of  $6.7$  kbar. Finally, overall resistance of sample no. 2 in various experiments is about one-tenth of that of sample no. 3, again in between  $R_{xx}$  and  $R_{zz}$  while all the samples studied here

have the similar size. So, resistance of the sample no. 2 is suspected to measure a kind of mixture of  $R_{xx}$  and  $R_{zz}$ . In fact, we examined the cross section of the sample no. 2 by slicing it little by little with a sharp razor blade and found that there was a small cavity hidden inside. In such a case, the current path is hardly well defined and  $R_{zz}$  is inevitably contaminated with  $R_{xx}$ . Another possibility is that the crystal is internally twinned. As the current always flows along the lowest possible resistance path, it is natural that both  $R_{zz}(T)$  and  $R_{zz}(H)$  of this sample show similar behaviors to those of  $R_{xx}$ . Calculating  $\sigma_{zz}(H, \theta)$  from semiclassical Boltzmann equation within the single-relaxation-time approximation, it is anticipated that  $\rho_{zz}(H, \theta) \propto \rho_{zz}^0 [1 + (\omega_c \tau)^2 \sin^2 \theta]$  where  $\theta$  is the angle of magnetic field with the  $c^*$  axis and  $\omega_c = eHv_{Fc}/h$  is the frequency with which an electron traverses the Brillouin zone along the  $c$  axis in a magnetic field. This equation is the simplified form of Eq. (11) of Ref. 9 by putting  $t_b/t_a \sim 0$ . It supports also the type I background for  $R_{zz}(\theta)$ , giving a simple and clean explanation.

We do not know if the data presented in Refs. 16 and 14 or Ref. 15 are obtained from the same sample or from several different samples. It is also too premature to say that two different backgrounds result only from the crystal imperfections. However, after failing to find any pressure-induced crossover of angular magnetoresistance behavior in spite of extensive search, we may exclude the possibility of type I to type II crossover of  $R_{zz}$  when the external pressure increases. Absence of crossover was also verified in another compound,  $(\text{TMTSF})_2\text{ReO}_4$ . Although some of the  $R_{zz}$  configured samples showed an  $R_{xx}$ -like behavior, pressure-induced crossover has never been observed in any samples.<sup>23</sup>

Now, it is evident that the type I background of angular magnetoresistance is intrinsic to the interlayer resistance  $R_{zz}$  even in  $(\text{TMTSF})_2\text{PF}_6$ . As a result, many of the earlier analysis based on the observation of the type II background in  $(\text{TMTSF})_2\text{PF}_6$  and its crossover needs to be revisited.<sup>13,15,19</sup>

#### IV. CONCLUSION

A careful study showed that the long-time mysterious pressure-induced crossover of angular magnetoresistance background of  $(\text{TMTSF})_2\text{PF}_6$  could not be reproduced if the same sample is temperature cycled at several different pressures. More probable is that the type II angular magnetoresistance is a characteristic of  $R_{xx}$  while the type I is that of  $R_{zz}$ . The type I nature of  $R_{zz}$  does not change with pressure contrary to the previous reports. Sometimes,  $R_{zz}$  showing  $R_{xx}$ -like behavior has been reported to occur, but it is more probable that there is some defect which modifies current path so that intraplane component is mixed to the interlayer transport. As it becomes evident that only the type I background is intrinsic property of  $R_{zz}$ , the models based on earlier data of the type II background and its crossover needs to be revisited.

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