

# On the existence of Rapid Oscillations in various phases of quasi one-dimensional $(\text{TMTSF})_2\text{PF}_6$

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We report on the studies of magnetoresistance in  $(\text{TMTSF})_2\text{PF}_6$ . We have found that Rapid Oscillations of the magnetoresistance are absent in the metallic state and are present in the spin-ordered states solely, including both the lowest- and higher-order FISDW states. The spin-ordered state, which had previously been believed to be insulating, is not totally gapped; at least, at a finite temperature, there remains a vestigial Fermi surface comprising 2D metallic “pockets”. Our data agree qualitatively with the theory that considers the coexistence of two spin-density waves with two respective nesting vectors.

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$(\text{TMTSF})_2\text{PF}_6$  represents a layered three-dimensional lattice hosting a quasi one-dimensional (Q1D) electron system. At high temperatures its Fermi surface (see Fig.1) comprises two sheets directed

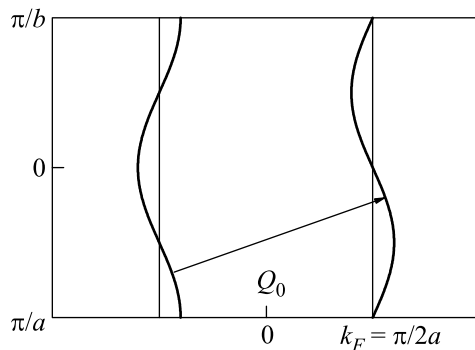


Fig.1. Schematic view of the first Brillouin zone for  $(\text{TMTSF})_2\text{PF}_6$  with a corrugated open Fermi surface (two bold lines). Arrow shows the main nesting vector  $Q_0$

perpendicular to  $a$ , which are slightly corrugated due to a small transfer integrals in  $b'$  and  $c^*$  directions (for reviews, see Refs. [1–3]). The ground state of a one-dimensional electron system is expected to be a non-Fermi-liquid. In particular, in  $(\text{TMTSF})_2\text{PF}_6$ , the electron system undergoes a phase transition to the spin-density wave (SDW) state as temperature decreases below  $T_{\text{SDW}} \approx 12$  K. In the SDW state, a nesting wavevector  $Q_0$  with its  $x$ -component  $Q_{0x} = 2k_F$  couples across the Fermi surface a large number of

degenerate states and determines direction of the antiferromagnetic ordering.

Elevated pressure  $P > 6$  kbar suppresses the onset of the SDW and stabilizes the metallic state [3–5]. Application of a sufficiently strong magnetic field in  $c^*$  direction induces a cascade of Field Induced SDW phases (FISDW) [1–3, 6, 7, 4], which are related with quantized changes in the  $x$ -component of the nesting vector  $Q_{0x}$  [1–3]

$$Q_{0x} = 2k_F - N \frac{eBb}{h}, \quad (1)$$

where  $k_F$  is the Fermi wave vector,  $b$  is the size of the elementary cell in  $y$ -direction, and  $N = i, i - 1, \dots, 0$  is an integer. The cascade terminates in the insulating  $N = 0$  phase; the latter is believed to be the same SDW phase as that for zero pressure.

One of the most puzzling features of the SDW phase in this material is the periodic in the inverse magnetic field oscillatory magnetoresistance, the so called “Rapid oscillations” (RO) [6–10]. Indeed, at first sight, for a Q1D system all electronic states are expected to be localized and hence, quantum oscillations should not occur. Secondly, the oscillation period seems to have nothing in common with the one-dimensional Fermi surface geometry. Almost all preceding studies of RO [8–13] have been performed at zero pressure and are therefore incomplete. Neither the experimental data nor corresponding theoretical suggestions [14–18] constitute an unambiguous picture of the phenomenon that can be applicable to materials with both centrosymmetric and nonsymmetric anions.

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In this paper we report experimental studies that answer the question on the potential existence of RO in various phases, including insulating FISDW, SDW and metallic states. In order to study RO in various phases in a single experiment, we applied a pressure  $P > 6$  kbar and vary magnetic field. As described above, pressure enables to suppress the onset of the SDW and to stabilize the metallic state [3]. Thus, at high pressures, by varying magnetic field we were able to explore with one and the same sample both metallic and insulating FISDW phases, including the zero-field SDW phase.

We have found and report here that RO exist only in the spin-ordered (SDW or FISDW) states and are missing in the metallic state. Beyond the lowest-order  $N = 0$  state, we also observed RO in the higher-order insulating state, such as  $N = 1$ . The oscillations in both phases have the same period in  $1/B$ , though are possibly slightly shifted in phase at the transition  $N = 1 \leftrightarrow N = 0$ . These results signify that the spin-ordered  $N = 0$  and  $N = 1$  states, which were believed to be insulating [1–3], in fact, are not totally gapped; at least at finite temperature, there remains a vestigial Fermi surface that comprises 2D metallic pockets.

Measurements were performed with  $(\text{TMTSF})_2\text{PF}_6$  sample (of a  $2 \times 0.8 \times 0.3 \text{ mm}^3$  size) grown by conventional electrochemical techniques.  $25 \mu\text{m}$  Pt-wires were attached using graphite paint to the sample on the  $a-b$  plane along the most conducting direction  $a$ . The sample and a manganin pressure gauge were inserted in a miniature spherical pressure cell [19] of 15 mm outer diameter, filled with polyethylsiloxane pressure transmitting liquid [20]. To study the dependence of the magnetotransport on magnetic field orientation under pressure, we rotated the spherical pressure cell containing the sample [19], *in situ* in the bore of a superconducting magnet. The cell was mounted at a two-axes rotation stage placed in  $\text{He}^4$  in a bore of either 17 T or 21 T superconducting magnets. The rotation system enabled rotation of the pressure cell with a sample around the two axes. Sample resistance  $R_{xx}$  was measured using a four probe ac technique with a bias current of 1 to  $4 \mu\text{A}$  at a 16 to 132 Hz frequency.

Figure 2a shows the magnetoresistance measured at fixed temperature  $T = 4.2 \text{ K}$  for various pressures. At a low pressure of 5 kbar, the sample is in the SDW state. This is illustrated by the temperature dependence of the resistance at zero magnetic field shown in the inset to Fig.2. As temperature decreases below  $\approx 6 \text{ K}$ , the resistance sharply rises (see the upper curve in the inset) signaling the transition from a metallic to an insulating state; this is in accord with the known phase diagram [3]. In this insulating SDW state, at  $T = 4.2 \text{ K}$ , as magnetic field increases, the resistance starts to oscillate above a

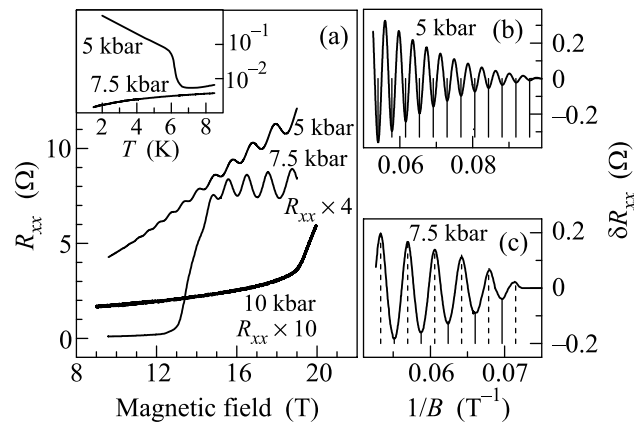


Fig.2. (a) Magnetoresistance  $R_{xx}$  measured at  $T = 4.2 \text{ K}$ ,  $B \parallel c^*$  for different pressures. Scaling factors and pressures are indicated next to each curve. Inset shows temperature dependence of  $R_{xx}$  at  $B = 0$  for  $P = 5$  and  $7.5 \text{ kbar}$ . Right panels: oscillatory part of  $R_{xx}$  at  $T = 4.2 \text{ K}$  versus  $1/B$  for  $P = 5 \text{ kbar}$  (b) and  $P = 7.5 \text{ kbar}$  (c). Vertical lines are equidistant in  $1/B$

field of approximately 10 T [see the upper curve in panel (a)]. The oscillations are periodic in  $1/B$ , as Fig.2b shows. For higher pressures  $P > 6 \text{ kbar}$ , the sample remains metallic at zero field, down to the lowest temperatures. This is demonstrated by the monotonic temperature dependence of the resistance with  $dR/dT > 0$  for  $P = 7.5 \text{ kbar}$  in the inset to Fig.2 (lower curve); for  $P = 10$  and  $15 \text{ kbar}$  (data are not shown in the figure), the temperature dependences are similar to that for  $P = 7.5 \text{ kbar}$ , and are only slightly shifted to lower resistance values.

As magnetic field rises, the sample experiences a transition from metallic state to the FISDW state. This is clearly seen on the curve for  $P = 7.5 \text{ kbar}$  as a sharp, factor of 50 increase in the resistance at  $\approx 13 \text{ T}$  (see the main panel Fig.2a). Immediately after the transition to the spin-ordered state the magnetoresistance starts to oscillate; again the oscillations are periodic in  $1/B$  (see Fig.2c). As pressure increases further, the onset of the FISDW state shifts to progressively higher fields (see curve for  $P = 10 \text{ kbar}$ ). Importantly, there are no oscillations seen at the same temperature for  $P = 10$  and  $15 \text{ kbar}$  over the whole range of magnetic fields corresponding to the metallic state.

The oscillations in the FISDW state are so large ( $\sim 20\%$  at  $B = 18 \text{ T}$  and  $P = 7.5 \text{ kbar}$ ) that can be easily extrapolated to lower fields. However, no oscillations can be seen in the metallic state at fields  $B < 14 \text{ T}$ , below the FISDW transition (see  $P = 7.5 \text{ kbar}$  curve in Fig.2). This confirms that the oscillations arise si-

multaneously with the onset of the FISDW state, in a step-like fashion with a nonzero amplitude.

An interesting question is whether RO exist only in the SDW and  $N = 0$  FISDW phases or is a more general phenomenon, intrinsic also to other ( $N \neq 0$ ) FISDW phases. In order to elucidate this issue, we present in Fig.3 the derivative of the resistance with respect

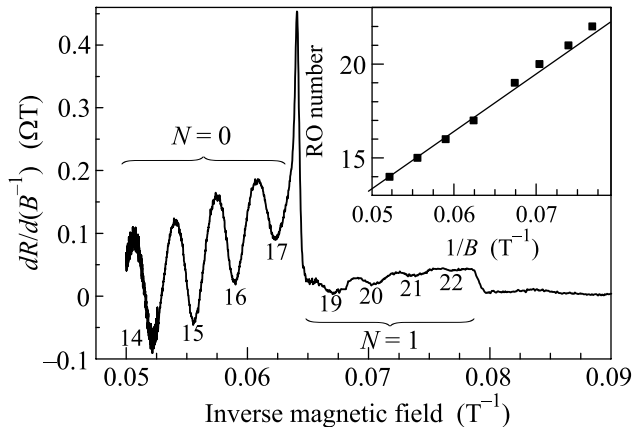


Fig.3. Derivative of the resistance  $dR_{xx}/d(1/B)$  versus  $1/B$  at  $T = 2$  K and  $P = 10$  kbar.  $N = 0$  and  $N = 1$  designate two FISDW phases; the oscillation numbers are indicated next to each minima. Insert shows RO sequential number versus inverse field

to the inverse magnetic field  $dR/d(1/B)$ . In this way, the short-period RO are magnified, as compared with the monotonic background magnetoresistance and the FISDW step-like transition  $N = 0 \leftrightarrow 1$  (see a  $\delta$ -shape peak in Fig.3). Figure 3 clearly shows that RO exist not only in the  $N = 0$  but also in the  $N = 1$  phases, though they are much weaker in the latter case. The inset to Fig.3 demonstrates that RO in the  $N = 1$  phase are an extension of those in the  $N = 0$  phase and, hence, RO in both phases have a common origin.

The totality of our experimental data taken in the ranges of temperature 1.4 – 8 K, magnetic field (up to 20 T) and pressures (0–1.5 GPa) proves that RO in  $(\text{TMTSF})_2\text{PF}_6$  are *intrinsic only to the spin-ordered state, and hence, are caused by nesting*.

The results described above fit best of all the theory by Lebed [21, 17], which considers the influence of Umklapp processes on the spin ordering [22]. Such processes can exist in a half-filled band. The Umklapp processes with a reciprocal lattice wavevector  $K_a$  result in the appearance of an auxiliary nesting vector  $Q_1$ :

$$Q_1 = Q_0 - K_a = Q_0 - 2\pi/a = Q_0 - 4k_F, \quad (2)$$

as shown in Fig.4. This theory [21] presumes a commensurability of only the  $x$ -component of the nesting vec-

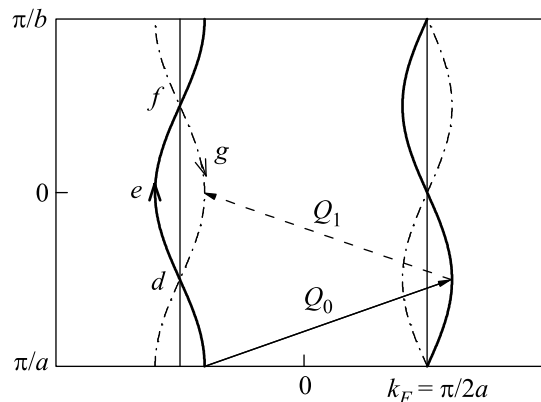


Fig.4. Schematic view of the first Brillouin zone with the open Fermi surface (two bold lines). Thick arrow shows the primary nesting vector  $Q_0$ ; dashed arrow shows the secondary nesting vector  $Q_1$ , involving the Umklapp processes. Arrows at points  $e$  and  $g$  show the direction of carrier motion in a magnetic field perpendicular to the  $a - b$  plane

tor with the lattice, in contrast to other models [9, 10]. The main spin density wave with nesting vector  $Q_0$  is responsible for the localization of the majority of the carriers, whereas the secondary spin-density wave with nesting vector  $Q_1$  is responsible for the appearance of the delocalized carriers occupying metallic “pockets” in momentum space (see Fig.4). The secondary SDW has a smaller amplitude, because it is caused by Umklapp processes.

A small share of carriers occupies novel contours of the FS (shown by the dashed lines in Fig.4), which are obtained due to the secondary SDW with  $Q_1$  wavevector Eq. (2). Correspondingly, the same small share of empty states must appear at the main contours of the FS (i.e. at the thick contours in Fig.4). The electrons belonging to the main and the secondary SDW have the same Fermi energy and move in the same direction in magnetic field (clockwise in Fig. 4). Despite a complex character of motion that involves Umklapp scattering, the motion of carriers is finite and is quantized in magnetic field. The coexistence of delocalized carriers at two branches of the FS may therefore be considered as metallic “pockets” (closed orbits  $d - e - f - g - d$  in momentum space), as illustrated in Fig.4. It is essential that, according to the theory [21], no magnetic breakdown is required for the formation of the “pockets” and, hence, for the appearance of the oscillations in  $(\text{TMTSF})_2\text{PF}_6$ ; this prediction is in agreement with our data. The closed “pockets” are two-dimensional and lie in the  $a - b$  plane, which also agrees with our measurements of the magnetoresistance anisotropy [23].

In theory [21], the size of the closed pockets in momentum space depends only on the warping of the FS (i.e. on the  $t_b$  transfer integral) and is independent of temperature and magnetic field. This is again in accord with our data. The frequency of the oscillations calculated on the basis of the model,  $4t_b/\pi e b v_F$ , equals to 286 T, where  $e$  is the elementary charge,  $b = 6.7 \text{ \AA}$ ,  $v_F = 1.11 \cdot 10^5 \text{ m/s}$  (as follows from our cyclotron resonance measurements [24]) and  $t_b$  is taken to be 200 K. The experimentally measured frequency is 275 T at  $P = 7.5 \text{ kbar}$  [23, 25], which is very close to the calculated value.

Within the framework of this theory, RO should arise only in the spin-ordered phase, as a result of the coexistence of the two spin-density waves with two nesting vectors, respectively. This prediction is in accord with our experimental data. The secondary SDW is caused by Umklapp processes and therefore has a small amplitude (corresponding to a smaller gap). This agrees with our experimental data [23], where the proportion of the delocalized states was estimated to amount to a few percent. The oscillation amplitude drastically weakens at the transition to the higher-order FISDW phase  $N = 1$ . This is consistent with the deterioration of nesting and weakening of the corresponding SDW amplitudes  $Q_0$  and  $Q_1$ . Thus, the theory explains qualitatively and, in part, quantitatively our experimental results. It remains unclear how the secondary SDW depends on temperature and whether it persists down to  $T = 0$ ; our data [23] shows weakening of the secondary SDW as  $T$  decreases and does not exclude its complete disappearance at  $T = 0$ .

*In summary*, we studied the magnetoresistance in the spin-ordered phases of  $(\text{TMTSF})_2\text{PF}_6$ . We have found the existence of delocalized states in the SDW and FISDW phases (at least, for finite temperatures), which earlier were considered to be purely insulating. The delocalized states occupy in momentum space a sort of two-dimensional "pockets" lying in the  $a - b$  plane. The corresponding closed orbits in momentum space are quantized in a perpendicular magnetic field giving rise to the Rapid Oscillations. Our data agree with the theory [21] that considers two coexisting SDW with two nesting vectors, respectively. In the theory, the second (auxiliary) spin density wave is formed due to Umklapp processes. Our results strongly support this theoretical suggestion on the coexistence of two spin density waves in quasi-one dimensional  $(\text{TMTSF})_2\text{PF}_6$ .

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