Origin of rapid oscillations in low-dimensional $(TMTSF)_2PF_6$

A. V. Kornilov,[1] V. M. Pudalov,[1] A.-K. Klehe,[2] A. Ardavan,[2] J. S. Qualls,[3] and J. Singleton[2,4]
[1]*P. N. Lebedev Physics Institute, Moscow 119991, Russia*
[2]*Clarendon Laboratory, Oxford University, Oxford OX1 3PU, United Kingdom*
[3]*Sonoma State University, Rohnert Park, California 94928, USA*
[4]*National High Magnetic Field Laboratory, Los Alamos, New Mexico 87545, USA*
(Received 10 March 2006; revised manuscript received 3 June 2007; published 17 July 2007)

We report studies of the magnetoresistance anisotropy in $(TMTSF)_2PF_6$, that shed light on the origin of the "rapid oscillations" (ROs). We have found that (i) ROs exist only in the spin-ordered state and are absent in the metallic state, (ii) decreasing temperature causes nonmonotonic variation of the RO magnitude, however, it does not affect the frequency of ROs, (iii) the spin-ordered state, which had previously been believed to be insulating, is not totally gapped (at least for finite temperatures), (iv) the RO frequency depends only on the magnetic field component that is normal to the *a-b* crystal plane. In our view, these results show that in the spin-ordered states there remains a vestigial Fermi surface comprising closed orbits in the *a-b* crystal plane. The orbits are quantized in magnetic field and give rise to the ROs. While decreasing temperature does not change the size or orientation of the orbits, it does cause a depopulation of the delocalized states (closed orbits) in favor of localized ones; this results in the disappearance of the ROs at low temperatures. Our data agree qualitatively with a theory that considers the coexistence of two spin-density waves with two respective nesting vectors. It is the coexistence of these two spin-density waves that gives rise to the closed orbits and, consequently, causes the rapid oscillations.

DOI: 10.1103/PhysRevB.76.045109 PACS number(s): 71.27.+a, 71.30.+h, 73.40.Qv

I. INTRODUCTION

The layered organic compound $(TMTSF)_2PF_6$ represents a three-dimensional lattice hosting a low-dimensional electron system. Conduction in this material is highly anisotropic, $\sigma_{xx}:\sigma_{yy}:\sigma_{zz}\sim 10^5:10^3:1$, along the a, b', and c^* crystal directions, respectively (for reviews, see Refs. 1–4). The Fermi surface (FS) comprises two sheets directed perpendicular to a, which are slightly corrugated due to a small transfer integrals in the b and c directions. Owing to its quasi-one-dimensionality (Q1D), the electron system undergoes a phase transition to a spin-density wave (SDW) state at low temperatures ($T<12$ K).

One of the most interesting features of the SDW phase in this material is the oscillatory magnetoresistance periodic in inverse magnetic field, $1/B$, the so-called "rapid oscillations" (ROs).[5–9] Their existence, by itself, is puzzling because at first sight, for a Q1D system, all electronic states are expected to be localized and, hence, quantum oscillations are not expected to occur. Another puzzle is that the ROs have a nonmonotonic temperature dependence:[5–9] as T decreases, their magnitude first grows, reaches a maximum, and then vanishes; these features are clearly in contrast with those of regular quantum oscillations (e.g., Shubnikov–de Haas oscillations).

Along with the disappearance of the ROs, in the same temperature interval a number of other anomalies are observed in studies of NMR,[10] microwave conductivity,[11] spin susceptibility,[12] specific heat,[13] etc. This set of phenomena provides evidence for an interesting transformation in the spin-density wave system with temperature; this is currently the focus of research interest and is far from being resolved. Thus, the study of the ROs may shed light on the puzzling phenomena associated with the transformation of the SDW in this would-be simplest of 1D systems.

The oscillations have been extensively studied since the early 1980s; they are thought to be related to the general properties of low-dimensional physics, because ROs are observed in almost all known Q1D organic compounds $(TMTSF)_2X$ family ($X=PF_6, AsF_6, ClO_4, NO_3$, etc.)[8,9,14–17] and in $(DMET\text{-}TSeF)_2AuCl_2$.[18] Materials with noncentrosymmetric anions (e.g., ClO_4, NO_3, etc.) undergo a doubling of the lattice periodicity due to anion ordering. The corresponding energy gap in the electron spectrum is used in various models (either in combination with or without a nesting vector) proposed for the explanation of ROs. The models are based on the formation and quantization of either closed orbits or interference orbits.[19–23] These models are evidently inapplicable to materials with centrosymmetric anions (e.g., PF_6, AsF_6), where no anion ordering occurs; for this reason, more sophisticated models have been proposed.[8,9,24,25]

Almost all preceding studies of the ROs have been performed at zero pressure. In order to verify the potential existence of the ROs in the metallic phase, we applied a pressure $P>0.6$ GPa; this is known to suppress the onset of the SDW and to stabilize the metallic state.[3] Application of a sufficiently strong magnetic field along c^* breaks down the metallic state and induces a cascade of field induced SDW (FISDW) phases, $N=i, i-1, \ldots$),[1–4] which terminates in the insulating $N=0$ phase; the latter is believed to be the same SDW phase as that for zero pressure. Thus, at high pressures, by varying the magnetic field we are able to explore both metallic and insulating FISDW phases at a single temperature. To study the dependence of the magnetotransport on magnetic field orientation under pressure, we rotated the spherical pressure cell containing the sample,[26] *in situ* in the bore of a superconducting magnet.

We have found the following:

(i) ROs exist only in the spin-ordered (SDW or FISDW) state and are absent in the metallic state (this is in contrast to those found in other TMTSF compounds with noncentrosymmetric anions).

(ii) RO magnitude changes strongly and nonmonotonically with temperature, whereas the frequency of ROs is insensitive to temperature.

(iii) The spin-ordered state, which is believed to be insulating,[1–4] is not totally gapped (at least for finite temperatures).

(iv) The ROs are determined only by magnetic field component that is normal to the *a*-*b* crystal plane.

These results, we believe, demonstrate the following:

(i) In the spin-ordered states, there remains a vestigial Fermi surface comprising closed orbits in the *a*-*b* crystal plane.

(ii) The closed orbits are quantized in magnetic field and give birth to the ROs.

(iii) Decreasing temperature does not change the size or orientation of the orbits but does cause depopulation of the delocalized states (closed orbits) in favor of the localized ones; this results in the disappearance of the ROs.

Our data agree qualitatively with a theory[25] that considers the coexistence of two SDWs with two respective nesting vectors. It is the coexistence of the two spin-density waves that gives rise to the closed orbits and, consequently, to the rapid oscillations.

II. EXPERIMENT

Two samples (of typical size $2\times0.8\times0.3$ mm^3) were grown by conventional electrochemical techniques. 25 μm 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 into a miniature spherical pressure cell with an outer diameter of 15 mm (Ref. 26) filled with Si-organic pressure transmitting liquid.[27] The pressure was created and fixed at room temperature. The pressure values quoted throughout this paper refer to the helium temperatures, which were determined as described in Ref. 28. The cell was mounted in a two-axis (θ,φ) rotation system placed in He^4 in a bore of either 17 or 21 T superconducting magnets. The rotation system enabled the rotation of the pressure cell (with a sample inside) around the main axis θ by 200° (with an accuracy of ~0.1°) and around the auxiliary axis φ by 360° (with accuracy of ~1°); this allowed us to set the sample at any desired orientation with respect to the magnetic field direction within 4π sr. The sample resistance R_{xx} was measured using a four probe ac technique with an excitation current of 1–4 μA (to avoid nonlinear effects in the SDW state) at a frequency of 16–132 Hz. In additional studies of the electric field effect on ROs, we applied currents up to 1 mA. The out-of-phase component of the measured voltage was found to be negligible in all measurements, indicating good Ohmic contacts to the sample.

A. On the existence of oscillations in various domains of the *P*-*B*-*T* phase space

Figure 1 shows the magnetoresistance measured at fixed temperature T=4.2 K for various pressures. At low pressure,

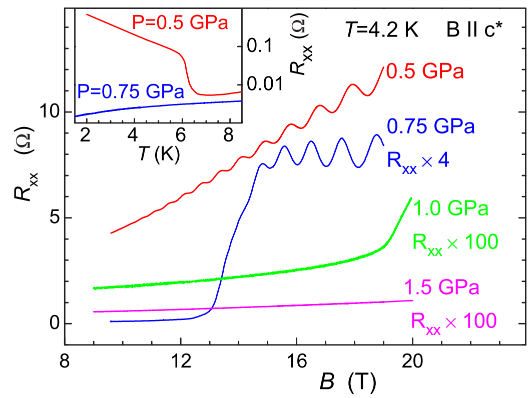

FIG. 1. (Color online) Magnetoresistance R_{xx} measured at T =4.2 K for $B\parallel c^*$ at different pressures. Scaling factors and pressure values are indicated next to each curve. The inset shows the temperature dependence of R_{xx} at B=0 for two pressures.

0.5 GPa, 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 the figure. As temperature decreases below ≈6 K, the resistance sharply rises (see the upper curve in the inset), signalling 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 K, as magnetic field increases, the resistance starts to oscillate above a field of approximately 10 T (see the upper curve in the main panel). For higher pressures $P>0.6$ GPa, 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=0.75 GPa in the inset to Fig. 1 (lower curve); for P=1 and 1.5 GPa, the temperature dependences are similar to that for P=0.75 GPa and only slightly shifted down in the resistance.

In the metallic state, as the magnetic field rises, the sample experiences a transition to the FISDW state. This is clearly seen on the curve for P=0.75 GPa as a sharp factor of 50 increase in the resistance at a field of about 13 T (see the main panel of Fig. 1). Immediately after the transition to the spin-ordered state, the magnetoresistance starts to oscillate. As the pressure increases further, the onset of the FISDW state shifts to progressively higher fields (see curves at P=1.0 and 1.5 GPa). Importantly, there are no oscillations seen at the same temperature for two lower curves (1 and 1.5 GPa) over the whole range of magnetic fields corresponding to the metallic state.

The magnetoresistance data at P=0.75 GPa are analyzed in more detail in Fig. 2. These data show well-pronounced oscillations and a clear border (at $B\approx13$ T) between the metallic and spin-ordered states; therefore, it can be used to double check whether or not the oscillations persist to the metallic state. The oscillations in the FISDW state are so large (~20% at 18 T) that can be easily extrapolated to lower fields. For this extrapolation, we used the empirical field dependence of the RO amplitude measured at P =0.5 GPa (see Fig. 1) and scaled down (using one fitting parameter) to fit the data at P=0.75 GPa in the field range of 14–19 T. The extrapolated oscillations are shown by dotted

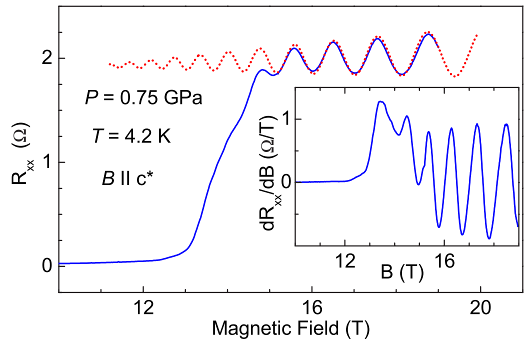

FIG. 2. (Color online) Magnetoresistance R_{xx} measured at $T = 4.2$ K for $B \| c^*$ at $P = 0.75$ GPa (continuous line). Dotted curve shows the extrapolation of the oscillatory magnetoresistance to lower fields, as explained in the text. Inset shows the derivative dR/dB that demonstrates a steplike onset of the oscillations at the FISDW transition ($B \approx 13$ T).

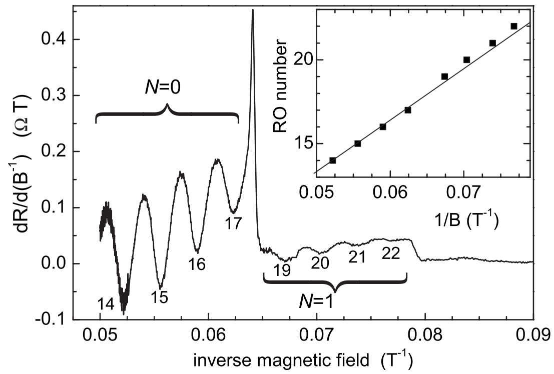

FIG. 4. Derivative of the resistance $dR_{xx}/d(1/B)$ versus inverse magnetic field at $T = 2.0$ K and $P = 1.0$ GPa. $N = 0$ and $N = 1$ designate two FISDW phases; the oscillation numbers are indicated next to the oscillation minima. The inset shows the RO number versus the inverse field.

line in Fig. 2. It appears, however, that no oscillations can be seen in the metallic state either in $R_{xx}(B)$ or in its derivative $dR_{xx}/dB(B)$ (as shown in the inset to Fig. 2). This confirms that the oscillations (i) occur along with the onset of the FISDW state and (ii) set in a steplike fashion, with a nonzero amplitude. It follows from the data presented in Figs. 1 and 2 that, at a given temperature and magnetic field, the existence of the ROs depends on whether the system is in the spin-ordered or in the metallic state.

Figure 3 shows the temperature evolution of the magnetoresistance for a fixed pressure $P = 1.0$ GPa. At high temperature $T = 4.2$ K, the system is in the metallic state and the oscillations are missing. As temperature decreases, the system undergoes transition to the FISDW state; this is concomitant with the appearance of the magnetoresistance oscillations. As above, we conclude that, for a given pressure and magnetic field, the existence of the ROs depends on whether the system is in the spin-ordered or in the metallic state.

An interesting question is whether the ROs 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. 4 the derivative of the resistance with respect to the inverse magnetic field $dR/d(1/B)$. In this way, the short-period rapid oscillations are magnified as compared with the monotonic background magnetoresistance and the FISDW transition $N = 0 \Leftrightarrow 1$ (see a δ-shape peak in Fig. 4). Figure 4 clearly demonstrates that ROs exist not only in $N = 0$ but also in $N = 1$ phases, though their magnitudes are much weaker in the latter case. The inset to Fig. 4 demonstrates that ROs in the $N = 1$ phase are an extension of those in the $N = 0$ phase and, hence, ROs in both phases have a common origin. The totality of our experimental data taken on two different samples in the ranges of temperature (1.4–8 K), magnetic field (up to 20 T), and pressures (0–1.5 GPa) proves that *the ROs in* $(TMTSF)_2PF_6$ *are intrinsic only to the spin-ordered state and hence are caused by nesting*.

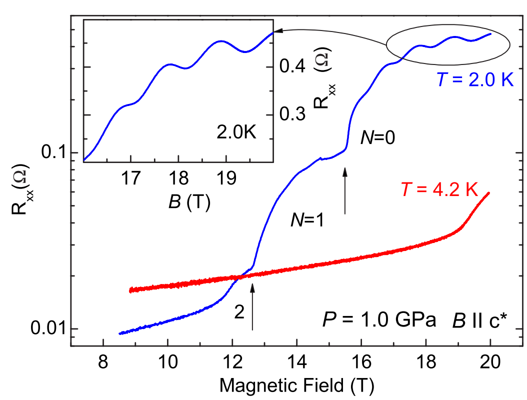

FIG. 3. (Color online) Magnetoresistance R_{xx} measured at $P = 1$ GPa for two temperatures, $T = 4.2$ and 2.0 K, with $B \| c^*$. Vertical arrows mark two FISDW phase transitions $N = 2 \leftrightarrow 1$ and $N = 1 \leftrightarrow 0$ (Refs. 3, 4, and 29). The inset blows up the oscillatory part of $R_{xx}(B)$.

B. Existence of delocalized states in the spin-ordered phase

Figure 5 shows two examples of the oscillatory part of the magnetoresistance δR_{xx} plotted versus the inverse magnetic field for two pressure values. For these two examples, as well

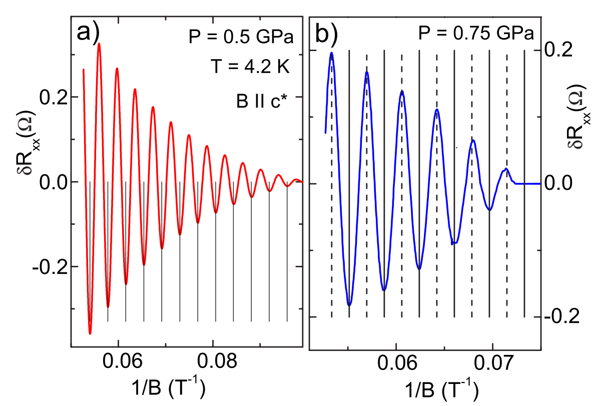

FIG. 5. (Color online) Oscillatory part of the magnetoresistance δR_{xx} versus the inverse magnetic field at $T = 4.2$ K for (a) $P = 0.5$ GPa and (b) $P = 0.75$ GPa. Vertical lines are equidistant in $1/B$.

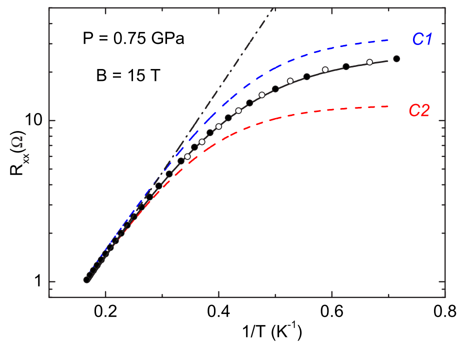

FIG. 6. (Color online) Temperature dependence of the background magnetoresistance at $P=0.75$ GPa at a magnetic field of 15 T. The bold line shows experimental data; dashed lines $C1$ and $C2$ are examples of data fitting using a two-component conductivity model [Eq. (1)] with T-independent metallic term, $C1$ and $C2$, respectively ($C1 < C2$). The dash-dotted line shows the T-activated conduction of a single component insulator with 12 K energy gap. Empty and filled circles show examples of fitting using a two-component conductivity model [Eqs. (2) and (3)] with T-dependent metallic term (described further in Sec. II D).

as for all other pressures and temperatures, studied in our experiment, the oscillations are periodic in $1/B$. Even at the FISDW transition $N=0 \Leftrightarrow 1$, the periodicity of the oscillations does not change (see Fig. 4).[30] It is therefore likely that the ROs originate from Landau quantization. This is in contrast to the quantized nesting model,[1–4] wherein the spin-ordered state should be a totally gapped insulating state. The $1/B$ periodicity of the oscillations thus strongly suggests the existence of delocalized carriers (and therefore closed orbits in the Brillouin zone).

The existence of the delocalized carriers in the spin-ordered state may be also deduced from the data shown in Fig. 6 that demonstrates the monotonic dependence of the background magnetoresistance $R_{bg}=R_{xx}-\delta R_{xx}$ on inverse temperature. For a totally gapped insulator, the temperature dependence is expected to have an activating or hopping character, $R \propto \exp(T_0/T)^p$. The experimental data indeed have an approximately activated character. However, neither activated ($p=1$) (dash-dotted line in Fig. 6) nor hopping ($p=1/2, 1/3$) temperature dependences can model the data over the whole temperature range $T=1.4-8$ K of the spin-ordered state. Taking the exponent p as a fitting parameter, one can achieve an approximate fitting only with p as low as 1/8 or 1/7, the values which have no apparent physical meaning. The lack of agreement is clearly caused by a "low-temperature" saturation of the data, which contrasts with hopping or activating dependences.

The saturation can be modeled by a straightforward addition of a "metallic conduction" component C_m to the temperature-activated (hopping) conduction of an insulator,

$$\sigma_{xx} = \sigma_{\text{ins}} + \sigma_{\text{met}} = A\exp(-T_0/T) + C_m. \tag{1}$$

The examples of fitting using Eq. (1) with two values $C_m = C1$ and $C2$ are shown by dashed lines in Fig. 6. The fit with constant $C2$ provides good agreement with the experimental data at high temperatures, whereas the fit with $C1$ provides correct slope of the $R(T)$ at low temperatures; however, neither choice of the fitting constant C_m can provide quantitative agreement with the data in the whole range of temperatures. Nevertheless, this simple model reflects the main feature of the experimental $R(T)$ dependence and qualitatively explains the crossover from the high-temperature-activated law to the low-temperature saturation of the data.

The two experimental results described above, (i) the $1/B$ periodicity of the oscillatory part of the magnetoresistance and (ii) the violation of activated temperature dependence of the background magnetoresistance, point at the *existence of delocalized states (closed orbits) in the spin-ordered phase.* These are the orbits that are quantized in a magnetic field and give rise to the rapid oscillations.

C. Magnetoresistance oscillations in a tilted field

The existence of the delocalized states in the spin-ordered phase raises the question on the geometry and orientation (in momentum space) of the corresponding Fermi surface responsible for the ROs. To address this issue, we measured the dependence of the frequency of the magnetoresistance oscillations on the magnetic field orientation. Examples of $R_{xx}(B)$ curves at $T=4.2$ K for a magnetic field tilted in the c^*-b' plane are shown in Fig. 7(a). When the field is tilted from the c^* axis ($\theta=0$), the frequency of oscillations (in inverse field) increases as $275/\cos(\theta)$ T [see the inset to Fig. 7(b)]. The same angular dependence has been obtained for tilting the field from c^* axes in other planes (the corresponding results are not shown). This angular dependence is evidence that *the closed orbits are (i) two dimensional and (ii) lie in the a-b crystal plane*.

As the temperature is changed, neither the frequency nor its angle dependence varies [see Fig. 7(b)]. This is illustrated further in the inset to Fig. 7(b), where data for $T=4.2$ and 2 K coincide with each other and follow the same angular dependence, $275/\cos(\theta)$. We conclude that *neither the size nor the orientation of the closed orbits change with temperature*.

D. Temperature dependence of the oscillations

The normalized amplitude of oscillations, $\delta R/R$, exhibits a nonmonotonic temperature dependence that is illustrated in more detail in Fig. 8. The oscillation amplitude slowly rises as T decreases from a high temperature, reaching a maximum at ≈ 3 K, and then sharply falls [see Fig. 8(b)]. The oscillation amplitude reflects the population of the delocalized states (closed orbits) and, hence, its T dependence [Fig. 8(b)] signals a temperature-dependent redistribution of the carriers between the delocalized and localized states, because the size of the closed orbits in momentum space does not change with temperature (see Sec. II C).

We propose that the measured T dependence of the oscillation amplitude is indicative of the occupation of the "metallic" two-dimensional (2D) orbits, and its decrease (as T falls) causes the disappearance of the oscillations. Hence, the metallic contribution σ_{met} to the two-component conduction model [Eq. (1)] should be taken to be temperature dependent.

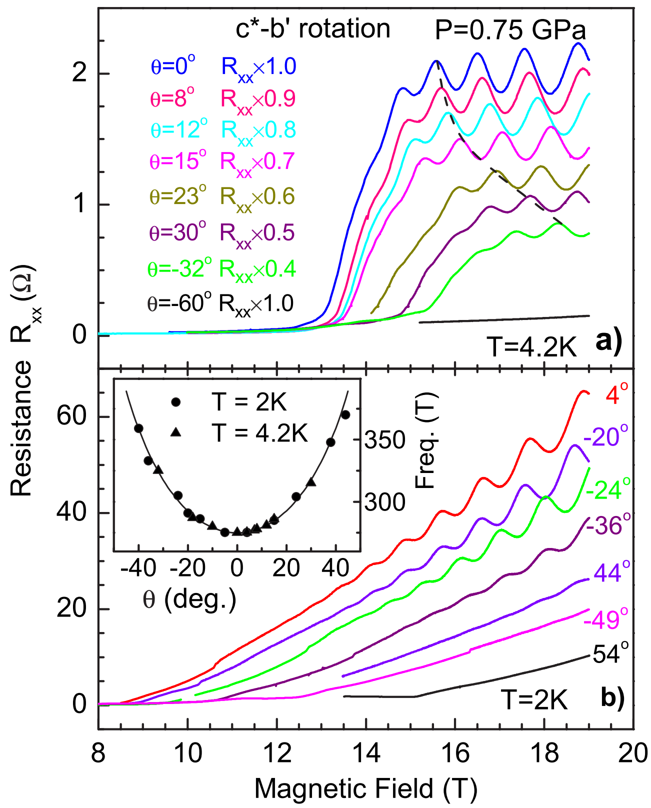

FIG. 7. (Color online) Magnetoresistance $R_{xx}(B)$ for various magnetic field orientations at $P=0.75$ GPa: (a) for $T=4.2$ K and (b) for $T=2$ K. θ designates the tilt angle in the c^*-b' plane ($\theta=0$ for field orientation along c^*). For clarity, the curves in panel (a) are scaled individually (the scaling factors are shown next to each curve). The dashed curve on the panel (a) connects the maxima of corresponding oscillations. The inset to panel (b) shows the angular dependence of the oscillation frequency for $T=4.2$ K (triangles) and 2 K (dots). Continuous curve depicts the $275/\cos(\theta)$ dependence.

It is reasonable to assume that σ_{met} is proportional to the concentration of the delocalized carriers n_m which contribute to the ROs, i.e., to the occupation of the 2D closed orbits with delocalized states. To estimate n_m versus temperature, we use an exponential function to describe the T-dependent carrier occupation,

$$n_m(T) = n_0 + n_1 \exp(-T_0/T). \tag{2}$$

This equation contains three fitting parameters: n_0 the occupation number at $T=0$, n_0+n_1 the occupation number at $T=\infty$, and T_0 the crossover temperature.

Although we do not think that the Lifshits-Kosevich formula for the 2D case[31] may describe the RO amplitude, it certainly may have some physical relevance. We therefore attempted to fit the occupation number empirically, by considering the nonmonotonic T dependence of the RO amplitude [Fig. 8(b)] as a product of (i) the growth of the carrier occupation with temperature and (ii) the decay of the amplitude of quantum oscillations with T,

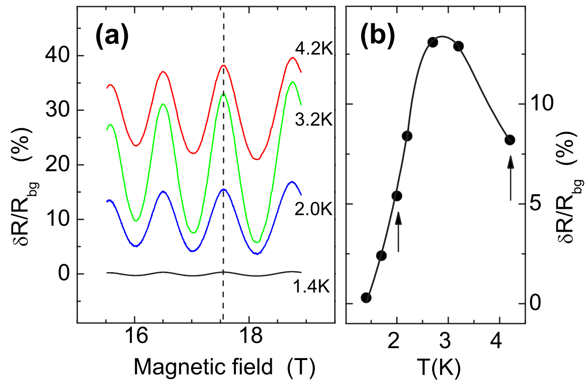

FIG. 8. (Color online) (a) Examples of the oscillatory component of the magnetoresistance $(R-R_{\mathrm{bg}})/R_{\mathrm{bg}}$ for $B \| c^*$. The curves for 4.2, 3.2, and 2.0 K are offset vertically, by 30%, 20%, and 10%, respectively. (b) Temperature dependence of the oscillation amplitude at $B=17.5$ T [marked on panel (a) with a dashed line]. Vertical arrows show the temperature values at which the angular dependence of the oscillation frequency was measured (data shown in Fig. 7). Pressure $P=0.75$ GPa for both panels.

$$n_m(T) = n_0 + n_2\left(\frac{\delta R}{R}\right)_{\mathrm{RO}}\left[\frac{2\pi^2 kT}{\hbar\omega_c \sinh(2\pi^2 kT/\hbar\omega_c)}\right]^{-1}. \tag{3}$$

In Eq. (3), $(\delta R/R)_{\mathrm{RO}}$ is the measured RO amplitude, the term in square brackets is the anticipated temperature decay of the quantum oscillations in the 2D "metal" with a fixed carrier density, the cyclotron mass is $1.55m_e$,[32] and n_0 and n_2 are the fitting parameters.

The filled and empty circles in Fig. 6 show the results of fitting with Eqs. (2) and (3), respectively. As seen, both models fit the experimental data equally well. It also follows from the above fitting that the relative proportion of the delocalized states is of the order of a few percent. Moreover, using Eq. (2), a successful fit can be achieved by using only two variable parameters, fixing $n_0=0$. Based on our experimental data and the above models, we cannot conclude whether or not the amount of delocalized states vanishes to zero at zero temperature. However, the above comparison confirms our conjecture that *the occupation of the 2D closed orbits, that provide metallic conduction, diminishes as temperature decreases and, hence, causes the disappearance of the rapid oscillations*.

III. DISCUSSION

Before comparing with theoretical models, we summarize our experimental results as follows:

(i) Rapid oscillations exist only in the spin-ordered state and are missing in the metallic state. This points at an intimate relation between the ROs and the spin ordering.

(ii) The $1/B$ periodicity of oscillations and the nonactivated (nonhopping) temperature dependence of the resistance suggest the existence (in the spin-ordered state) of the closed orbits in momentum space that provide metallic conduction at finite temperatures. These orbits are quantized in perpendicular field and give rise to the ROs.

(iii) The oscillations exist not only in the SDW and FISDW $N=0$ phases but also in the higher order FISDW

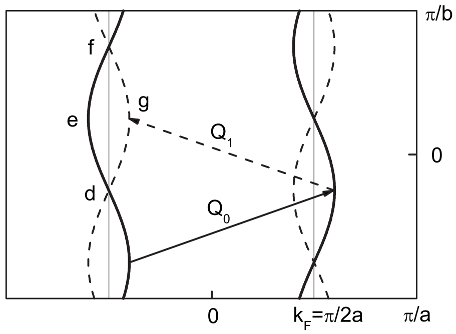

FIG. 9. Schematic view of the first Brillouin zone with a corrugated open Fermi surface (two bold lines). The thick arrow shows the primary nesting vector Q_0, and the dashed arrow shows the auxiliary nesting vector, involving the umklapp processes $Q_1 = Q_0 - 2\pi/a = Q_0 - 4k_F$ (Refs. 22 and 25).

phase $N=1$. The oscillation frequency does not change at the $N=0 \Leftrightarrow 1$ transition; however, their amplitude essentially weakens and the phase possibly changes.[30]

(iv) The oscillation frequency behaves as $1/\cos\theta$ as the magnetic field is tilted from the c^* axes in any direction. This proves that the closed orbits lie in the *a*-*b* crystal plane.

(v) Decrease in temperature causes the amplitude of the ROs to diminish; this directly shows that the occupation of the delocalized states decreases (and may even vanishes) as $T \to 0$.

(vi) Despite drastic changes of the RO amplitude, the frequency of the oscillations and its angular $1/\cos\theta$ dependence do not change with temperature. Therefore, neither areas (in momentum space) encircled by the closed orbits nor orientation of the orbits vary with temperature.

(vii) As $T \to 0$, the oscillation amplitude weakens, whereas the oscillation frequency does not change. This indicates that the temperature drop causes redistribution of the carriers from a delocalized to a localized subsystem.

The experimental results described above fit best to theories which consider the influence of umklapp processes on spin ordering.[24,25] Such processes can exist in a half-filled band with an energy spectrum consisting of two phase-shifted warped Fermi contours.[33] In the Yamaji model,[24] the umklapp processes lead to minigaps in the energy spectrum. For ideal crystal with k_F exactly equal to $\pi/2a$, the minigaps would fall in Landau gaps and would be invisible in transport. Due to imperfections, such as impurities, nonstoichiometry, dislocations, etc., k_F deviates from the ideal $\pi/2a$ value and minigaps split off the Landau gaps. Oscillations of the minigap parameters with field is the origin of the oscillations in resistivity in the Yamaji model.

In the Lebed model,[25] the umklapp processes result in the appearance of an auxiliary nesting vector Q_1 beyond the primary nesting vector Q_0: $Q_1 = Q_0 - 2\pi/a = Q_0 - 4k_F$, as shown in Fig. 9. Correspondingly, in addition to the main gap Δ_0 caused by Q_0, the second gap Δ_1 appears in the energy spectrum. The calculations in Ref. 25 are performed for $T \gg \Delta_0 \gg \Delta_1$. This theory[25] presumes a commensurability of only the *x* component of the nesting vector with the lattice, in contrast to other models.[8,9] The main spin-density wave with nesting vector Q_0 is responsible for the localization of the majority of the carriers, whereas the auxiliary spin-density wave with nesting vector Q_1 is responsible for the appearance of the delocalized carriers occupying closed orbits in momentum space. The auxiliary SDW has a smaller amplitude because it is caused by umklapp processes. The coexistence of the two SDWs is the origin of the oscillations.

Despite both theories[24,25] give correct value of the RO frequency (see below), they differ essentially in several other predictions.

(1) Within the framework of the Lebed theory, the ROs 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. In the Yamaji model, the oscillations may be expected to exist both in metallic and spin-ordered states. Our experiment firmly demonstrates that ROs exist only in the spin-ordered phase and are absent in the metallic state.

(2) In the Yamaji model, ROs arise due to crystal imperfections. In contrast, in the Lebed theory, crystal imperfections would cause broadening of the energy contours and decaying of the oscillations. Experimentally, the amplitude of ROs is the largest for the highest quality samples; it diminishes as the sample quality deteriorates.

(3) In the Yamaji model, transport occurs via tunneling through the minigaps and, hence, the RO magnitude should depend on electric field E as $\exp(-E_0/E)$, with $E_0 \approx 40$ mV/cm.[24] In contrast, there is no electric field dependence of the RO amplitude in the Lebed theory. In our experiments, by increasing current through the sample we observed that electric field up to $E=20$ mV/cm did not affect the RO amplitude within $\approx 1\%$ accuracy.

The above experimental results are in agreement with theory[25] and seems to disagree with Ref. 24; therefore, we perform further more detailed comparison only with the former theory.

The auxiliary SDW is caused by umklapp processes and therefore has a small amplitude. This agrees with our fitting [Eqs. (2) and (3)], where the proportion of the delocalized states was estimated to amount to a few percent. The oscillation amplitude drastically weakens at the transition to higher order FISDW phase $N=1$. This is consistent with deterioration of nesting and weakening of the corresponding SDW amplitudes Q_0 and Q_1.

The model explains the existence of closed orbits (*d*-*e*-*f*-*g*-*d*) in momentum space as illustrated in Fig. 9.[34] Moreover, according to the theory,[25] no magnetic breakdown is required for the formation of the closed orbits and, hence, for the appearance of the oscillations in $(TMTSF)_2PF_6$. The orbits are two dimensional and lie in the *a*-*b* plane, which is also in agreement with our data. It is the magnetic field quantization of the closed orbits in momentum space that gives rise to the rapid oscillations. In the theory, the size of the closed orbits 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, $b=6.7$ Å, $v_F = 1.11 \times 10^5$ m/s (as follows from the cyclotron resonance measurements[32]), and t_b is taken to be 200 K. The experimentally measured frequency at $P=0.75$ GPa is 275 T (see the inset to Fig. 7) that is very close to the calculated value.

Thus, the theory explains qualitatively and, in part, quantitatively almost all of our experimental results, except for the diminishing of the oscillation amplitude with decreasing temperature. In order to explain this experimental fact and complete the picture, the theory should be amended to account for the temperature dependence of the effectiveness of umklapp processes.

In summary, we studied the magnetoresistance in the spin-ordered phase of $(TMTSF)_2PF_6$. We have found that in this supposedly purely insulating phase, there exist delocalized carriers (at least, at finite temperature). These carriers occupy two-dimensional orbits in momentum space, lying in the *a*-*b* plane. The closed orbits in momentum space are quantized in a perpendicular magnetic field giving rise to the rapid oscillations. Temperature decrease does not change the size or orientation of the closed orbits, but it does cause a redistribution of carriers from the delocalized to the localized states and, hence, the disappearance of the ROs. Our data agree qualitatively with a theory that considers two coexisting SDWs with two nesting vectors, respectively. In the theory, the second (auxiliary) spin density wave is formed due to umklapp processes; in order to fit all our experimental data, its amplitude should weaken as temperature decreases. Our results thus shed light on the origin of the "rapid oscillations" in $(TMTSF)_2PF_6$.

ACKNOWLEDGMENTS

The authors are grateful to S. Brazovzkii and A. G. Lebed for fruitful discussions. The work was partially supported by the Programs of the Russian Academy of Sciences, RFBR, Russian Ministry of Education and Science, EPSRC, and the Royal Society.

[1] L. P. Gor'kov and A. G. Lebed, J. Phys. (Paris), Lett. **45**, L–433 (1984).
[2] M. Heritier, G. Montambaux, and P. Lederer, J. Phys. (Paris), Lett. **45**, L–943 (1984).
[3] P. M. Chaikin, J. Phys. I **6**, 1875 (1996).
[4] T. Ishiguro, K. Yamaji, and G. Saito, *Organic Superconductors*, 2nd ed. (Springer-Verlag, Heidelberg, 1998).
[5] J. P. Ulmet, P. Auban, A. Khmou, and S. Askenazy, J. Phys. (Paris), Lett. **46**, L–545 (1985).
[6] J. R. Cooper, W. Kang, P. Auban, G. Montambaux, D. Jérome, and K. Bechgaard, Phys. Rev. Lett. **63**, 1984 (1989).
[7] S. T. Hannahs, J. S. Brooks, W. Kang, L. Y. Chiang, and P. M. Chaikin, Phys. Rev. Lett. **63**, 1988 (1989).
[8] S. Uji, J. S. Brooks, M. Chaparala, S. Takasaki, J. Yamada, and H. Anzai, Phys. Rev. B **55**, 12446 (1997).
[9] J. S. Brooks, J. O'Brien, R. P. Starrett, R. G. Clark, R. H. McKenzie, S.-Y. Han, J. S. Qualls, S. Takasaki, J. Yamada, H. Anzai, C. H. Mielke, and L. K. Montgomery, Phys. Rev. B **59**, 2604 (1999).
[10] W. G. Clark, M. E. Hanson, W. H. Wong, and B. Alavi, J. Phys. IV **3**, 235 (1993).
[11] J. L. Musfeldt, M. Poirier, P. Batail, and C. Lenoir, Phys. Rev. B **51**, 8347 (1995).
[12] T. Takahashi, Y. Maniwa, H. Kawamura, and G. Saito, J. Phys. Soc. Jpn. **55**, 1364 (1986).
[13] J. C. Lasjaunias, K. Biljacović, F. Nad, P. Monceau, and K. Bechgaard, Phys. Rev. Lett. **72**, 1283 (1994); N. A. Fortune, J. S. Brooks, M. J. Graf, G. Montambaux, L. Y. Chiang, Jos A. A. J. Perenboom, and D. Althof, *ibid.* **64**, 2054 (1990).
[14] J.-P. Ulmet, A. Narjis, M. J. Naughton, and J. M. Fabre, Phys. Rev. B **55**, 3024 (1997).
[15] S. Uji, S. Yasuzuka, T. Konoike, K. Enomoto, J. Yamada, E. S. Choi, D. Graf, and J. S. Brooks, Phys. Rev. Lett. **94**, 077206 (2005).
[16] J. S. Qualls, C. H. Mielke, J. S. Brooks, L. K. Montgometry, D. G. Rickel, N. Harrison, and S. Y. Han, Phys. Rev. B **62**, 12680 (2000).
[17] N. Biskup, L. Balicas, S. Tomić, D. Jérome, and J. M. Fabre, Phys. Rev. B **50**, 12721 (1994).
[18] N. Biskup, J. S. Brooks, R. Kato, and K. Oshima, Phys. Rev. B **60**, R15005 (1999).
[19] S. A. Brazovskii and V. M. Yakovenko, JETP Lett. **43**, 134 (1986).
[20] A. G. Lebed and Per Bak, Phys. Rev. B **40**, R11433 (1989).
[21] V. M. Yakovenko, Phys. Rev. Lett. **68**, 3607 (1992).
[22] A. G. Lebed, Phys. Rev. Lett. **74**, 4903 (1995).
[23] K. Kishigi and K. Machida, Phys. Rev. B **53**, 5461 (1996).
[24] K. Yamaji, J. Phys. Soc. Jpn. **56**, 1101 (1987).
[25] A. G. Lebed, Phys. Scr., T **T39**, 386 (1991).
[26] A. V. Kornilov and V. M. Pudalov, Instrum. Exp. Tech. **42**, 127 (1999).
[27] A. S. Kirichenko, A. V. Kornilov, and V. M. Pudalov, Instrum. Exp. Tech. **48**, 813 (2005).
[28] A. V. Kornilov, V. M. Pudalov, Y. Kitaoka, K. Ishida, G.-q. Zheng, T. Mito, and J. S. Qualls, Phys. Rev. B **69**, 224404 (2004).
[29] A. V. Kornilov, V. M. Pudalov, Y. Kitaoka, K. Ishida, T. Mito, J. S. Brooks, J. S. Qualls, J. A. A. J. Perenboom, N. Tateiwa, and T. C. Kobayashi, Phys. Rev. B **65**, 060404(R) (2002).
[30] Whereas the oscillation period does not change at the FISDW transition, our data suggest that the phase of the oscillations is rather likely to change (see inset to Fig. 4). The accuracy of our experimental data is, however, insufficient to quantify the RO phase shift through the FISDW transition.
[31] A. Isihara and L. Smrčka, J. Phys. C **19**, 6777 (1986).
[32] A. V. Kornilov, P. J. M. van Bentum, J. S. Brooks, J. S. Qualls, J. Perenboom, and V. M. Pudalov, Synth. Met. **103**, 2246 (1999).
[33] For $(TMTSF)_2PF_6$, according to the NMR data (Ref. 12), the two warped Fermi contours are shifted by $\pi/2b$ along the b direction.
[34] Since umklapp processes are elastic, the quasiparticles on dashed and thick lines in Fig. 9 have the same energy and the contour *d*-*e*-*f*-*g*-*d* is equipotential. This contour looks similar to the ordinary 2D "metallic pocket." However, this is not a real 2D

metallic pocket because there is no 2D parabolic energy spectrum underneath the states shown by dashed lines in Fig. 9 (in contrast to the states shown by thick lines). These states can be considered similar to the known resonant states in semiconductors. The occupation number of these resonant states is determined by the intensity of the umklapp processes; it is not related with the area of the *d*-*e*-*f*-*g*-*d* contour (as it would be in case of a real metallic pocket). The geometry of the dashed contour and, hence, the *d*-*e*-*f*-*g*-*d* area are firmly determined by the Q_1 vector.