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Existence of two-dimensional closed orbits and delocalized carriers in the spin-ordered state of quasi one-dimensional $(TMTSF)_2PF_6$

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We report studies of the magnetoresistance in the spin-ordered state of the quasi one-dimensional $(TMTSF)_2PF_6$. We have found that both frequency and anisotropy of the magnetoresistance oscillations are independent of temperature; this proves temperature independence of the geometry of two-dimensional closed contours at the Fermi level. On the other hand, the oscillation amplitude decay as temperature decreases, the latter result indicates depopulation of the closed contours in the $T = 0$ limit. We propose an explanation for the above seemingly controversial results.

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The bulk organic crystal $(TMTSF)_2PF_6$ has a strongly anisotropic quasi one-dimensional (Q1D) carrier system. In the 1D case, the Fermi-liquid (FL) state is unstable, and, in particular, in $(TMTSF)_2PF_6$ the carrier system undergoes a phase transition to an antiferromagnetic spin-density wave (SDW) state below $T_{\mathrm{SDW}} \approx 12\,\mathrm{K}$ (at ambient pressure) [1 – 3]. As pressure increases, T_{SDW} decreases and vanishes at $P = 6\,\mathrm{kbar}$ [1 – 3], restoring the "metallic" Fermi liquid state. Application of the magnetic field B along the least conducting direction z (crystal axis c^*) [2, 3] restores the insulating state; this takes place via a cascade of the field induced spin density wave states with a quantized nesting wave vector [2 – 6]. Thus, by varying pressure or magnetic field one can drive the electronic system in this material from FL to non-FL states.

At first sight, the SDW state may be considered as an ordinary gapped insulating state. However, a number of anomalies were noticed in the SDW state in studies of NMR [7], microwave conductivity [8], spin susceptibility [9], specific heat [10] etc. Beyond that, an oscillatory magnetoresistance, the so called "Rapid oscillations" (RO) is observed in the SDW state [4, 5, 11 – 13]. These results point at a non-trivial character of the spin-ordered state. Indeed, it is well known that for a 1D system all electron states should be localized and hence, quantum oscillations are not expected to occur. Secondly, the oscillation period seems to have nothing in common with the one-dimensional Fermi surface geometry. Thus, the study of the RO may shed light on the puzzling origin of the spin-ordered state in the Q1D systems.

Earlier, we studied RO in $(TMTSF)_2PF_6$ and found them to exist not only in the generic SDW state but also in other magnetic field induced SDW phases [14]. We have also found that RO never exist in the metallic state; they arise in a step-like fashion, along with the onset of the spin-ordered state [14, 15]. These results signify that the spin-ordered state, which was believed to be purely insulating [16, 17], in fact, is not totally gapped; at least at finite temperature, its Fermi surface comprises closed 2D contours which mimic the metallic-like behavior.

The anomalous properties of the spin-ordered state are explained in the framework of the theoretical model [18, 19] that considers the influence of Umklapp processes on spin ordering [20]. However, the theory [18] is a consequence of the Landau theory of the 2nd order phase transitions and is valid in the vicinity of phase transition only, i.e. at high temperatures $T \geq T_{\mathrm{SDW}}$. The theory is inapplicable at low temperatures where the spin-ordered state demonstrates most puzzling features.

In the current paper we investigate properties of the spin-ordered state over a wide temperature range, by measuring the amplitude and frequency of the magnetoresistance oscillations. We report the following results: (i) Angular dependence of the RO frequency and its in-

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dependence of temperature prove that the closed contours are two-dimensional, lie in the $(a-b)$ crystal plane, and have a temperature-independent area and geometry. (ii) The disappearance of RO with lowering temperature points at diminishing population of the closed contours as temperature decreases.

At first sight, the two above results seem to be inconsistent with each other. We suggest here a physical picture that naturally incorporates the known features of RO and explains how the 2D closed contours with temperature-independent dimensions can have a T-dependent carrier population.

Measurements were performed with $(TMTSF)_2PF_6$ sample ($2 \times 0.8 \times 0.3\,\text{mm}^3$ size) grown by conventional electrochemical techniques. 25μm 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 [21] of 15 mm outer diameter, filled with polyethylsiloxane pressure transmitting liquid [22]. To study the dependence of the magnetotransport on magnetic field orientation under pressure, we rotated the spherical pressure cell containing the sample, *in situ*. The cell was mounted at a two-axes rotation stage placed in He^4 in a bore of either 16 T or 21 T superconducting magnets. The rotation system enabled rotation the pressure cell (with a sample inside) around two axes (with accuracy $\sim 1°$) within 4π steradian. The sample resistance R_{xx} was measured using a four probe ac technique with an excitation current of 1 to 4 μA (to avoid nonlinear effects in the SDW state) at a frequency of 16 to 132 Hz.

Figure 1a shows the magnetoresistance measured at four fixed temperatures. For the given applied pressure $P = 5\,$kbar, in accord with the known phase diagram [2, 3, 23], the sample is in the insulating SDW state. This is illustrated by the negative derivative dR/dT at zero magnetic field (Fig.1b), and also by the exponential field dependence of the magnetoresistance $\delta R \propto \exp[-(\Delta_0 + \alpha B)/T]$ (see Fig.1b) that is characteristic of an insulator. In the SDW state, as magnetic field increases, the resistance starts to oscillate above a field of approximately 13 T (see the inset to Fig.1a).

The oscillatory magnetoresistance data at $P = 7.5\,$kbar are shown in more detail in Fig.2a. The periodicity of oscillations in $1/B$ points at the existence of closed contours in the Brillouin zone of the Q1D system.

In order to clarify the geometry and orientation of the closed contours in momentum space we measured frequency of the magnetoresistance oscillations versus magnetic field orientation. Examples of the $R_{xx}(B)$-

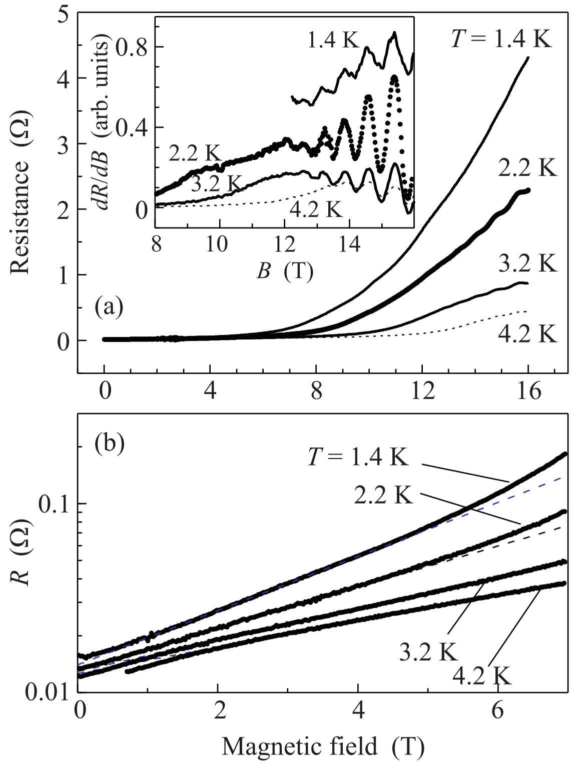

Fig.1. (a) Resistance R_{xx} versus magnetic field $B \parallel c^*$ measured at four different temperatures in the SDW phase, at $P = 5\,$kbar. The inset shows derivative dR/dB for the four curves from the main panel. (b) Semilogarithmic plot of the same $R(B)$ data. Dashed lines show the exponential $R(B)$ dependences

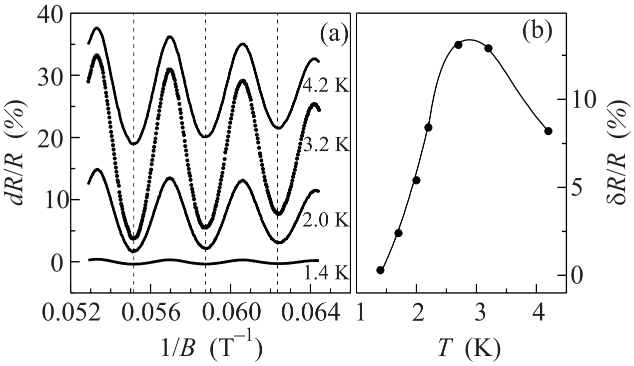

Fig.2. (a) Oscillatory part of the resistance $\delta R_{xx}/R$ versus inverse magnetic field at four temperatures and $P = 7.5\,$kbar. Vertical dashed lines are equidistant in $1/B$. Three upper curves are offset shifted. (b) Temperature dependence of the oscillation magnitude measured at $B = 17.5$T

curves at $T = 2\,$K for a magnetic field tilted in the $c^* - b'$ plane are shown in Fig.3. When the field is tilted from the c^*-axis ($\theta = 0$), the oscillation frequency increases as $275/\cos(\theta)$ T (see the inset to Fig.3). The same angular

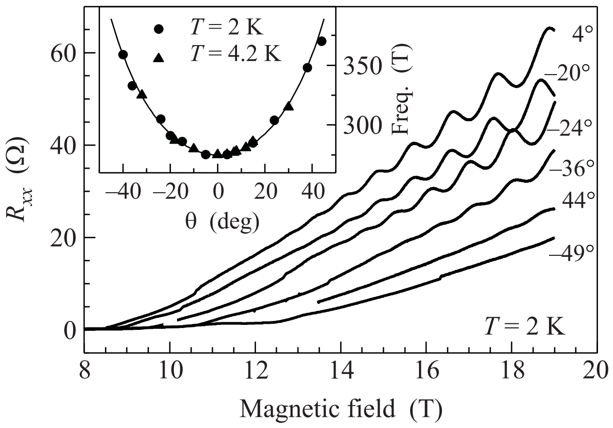

Fig.3. Magnetoresistance $R_{xx}(B)$ for various field orientations measured at $P = 7.5$ kbar, and $T = 2$K. θ designates the tilt angle in the $c^* - b'$ plane ($\theta = 0$ for $B \parallel c^*$). The inset shows angular dependence of the oscillation frequency for $T = 4.2$ and 2K. Continuous line depicts the fitting dependence, $275/\cos\theta$

dependence has been obtained for tilting field in the perpendicular plane. This angular dependence proves that the closed contours are (i) two-dimensional, and (ii) lie in the $a - b$ crystal plane.

Importantly, as temperature is changed, neither the frequency nor its angle dependence varies. This is illustrated in the inset to Fig.3, where the data for $T = 4.2$ and 2 K coincide with each other. We conclude that neither the area nor the orientation of the closed contours change with temperature.

The normalized amplitude of oscillations, $\delta R/R$, has a nonmonotonic temperature dependence (Fig.2b). The oscillation amplitude slowly rises as T decreases from high temperatures, reaching a maximum at ≈ 3 K, and then sharply falls. Although we don't think that the Lifshits-Kosevich (LK) formula for 2D case may describe the RO amplitude, it certainly have some physical relevance. Correspondingly, the T-dependent amplitude of oscillations in the range $T > 3$K may be due to the broadening of the Landau levels, as described by the LK formula. Exploiting this analogy further, we conjecture that the oscillation amplitude reflects the population of the delocalized states (closed contours), and that vanishing of the RO amplitude at $T \to 0$ (Fig.2b) signals a temperature-dependent depopulation of the closed contours.

The two above results, temperature independent area of the closed orbits and decay of the oscillation amplitude at $T \to 0$ seem to be in a contradiction with each other. Indeed, for a 2D energy spectrum, the number of carriers is firmly related with the k_F size and, hence, with the period of quantum oscillations. The measured RO period does not vary with temperature indicating constant concentration of the delocalized states, whereas the RO amplitude vanishes as T decreases; the latter indicates vanishing occupation of the delocalized states.

Despite the theoretical model [18] is valid only for high temperatures, we use it for qualitative analysis of our data taken in a wide temperature range. We show below that with an additional natural assumption, the model explains the above controversy. This model [18] considers, beyond the main SDW with the primary nesting vector Q_0, also a secondary SDW with Q_1 nesting vector:

$$Q_1 = Q_0 - K_a = Q_0 - 2\pi/a = Q_0 - 4k_F, \qquad (1)$$

where K_a is the reciprocal lattice vector (see Fig.4a). Consequently, the energy spectrum consists of two

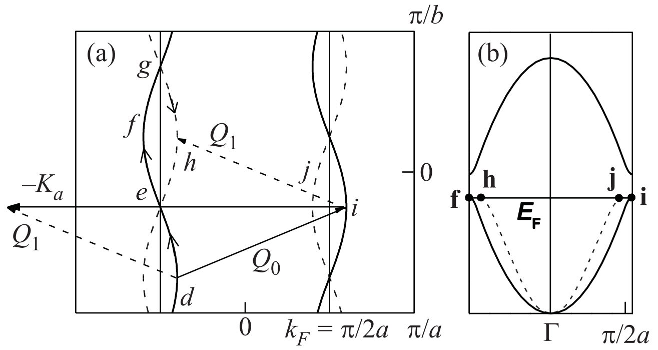

Fig.4. (a) The first Brillouin zone with a corrugated open FS (two bold lines). Solid arrow Q_0 shows the primary nesting vector, dashed arrow Q_1 – the secondary nesting vector, horizontal arrow – the reciprocal lattice vector $-K_a$; other details are explained in the text. (b) Schematic view of the energy spectrum $E(k)$ in the Q_1 direction, from i to f. Solid and dashed parabolic lines represent the primary and the secondary branches of the energy spectrum, respectively. Bold dots j and h represent the filled resonant states at the Fermi level (solid horizontal line)

branches: the primary branch related with Q_0 and the secondary branch related with Q_1 (bold and dashed parabolic lines in Fig.4b, respectively).

The main SDW is responsible for the localization of the majority of carriers. As a result of the Umklapp scattering, a small share of carriers is transferred to novel contours of the FS (shown by the dashed lines in Fig.4a), obtained due to the secondary SDW with Q_1 wave vector, Eq. (1). Correspondingly, the same small share of empty states must appear at the main branches of the FS (thick contours in Fig.4a). The electrons belonging to the main SDW branch (e.g., $d - e - f - g$), and the secondary branch ($g - h - e$) of the FS have the same energy E_F and move in the same direction in magnetic field (clockwise in Fig.4a). The coexistence of delocalized carriers at two branches of the FS may be considered as closed orbits $e - f - g - h - e$ in momentum space, as illustrated in Fig.4a. Despite a complex character of motion that involves Umklapp scattering, the motion of carriers is finite and therefore is quantized in magnetic field.

Due to the ensemble averaging, the Umklapp scattering takes place only at the Fermi energy E_F. Correspondingly, we assume that the carriers scattered in this way (dashed contours in Fig.4a) are transferred only at the Fermi level; they occupy discrete resonance states in the energy spectrum (depicted by the dots **j** and **h** in Fig.4b). Importantly, the closed contours are not equivalent to ordinary 2D closed pockets, because the states below E_F at the secondary branch are empty (dashed parabolic line in Fig.4b); this is in contrast to the filled states at the main branch (bold parabolic line). This is the reason why the T-dependent density of carriers occupying the closed contours appears to be irrelevant to the T-independent size of the closed orbits (the latter is determined by the main FS geometry solely).

In summary, we studied the oscillatory magnetoresistance in the spin-ordered phase of $(TMTSF)_2PF_6$. We have found that the size and orientation of the closed 2D orbits (responsible for the oscillations) does not change with temperature. On the other hand, the nonmonotonic temperature dependence of the oscillation amplitude provides an evidence for the temperature dependent population of the closed contours. This apparent contradiction finds a natural explanation within the theory model [18] supplemented with an assumption that the Umkplapp-scattered carriers occupy resonant states at the Fermi level.

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