Coexistence or Separation of the Superconducting, Antiferromagnetic, and Paramagnetic Phases in Quasi One-Dimensional (TMTSF)$_2$PF$_6$?

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Abstract

We report on experimental studies of the phase state and the character of phase transitions in the quasi-one-dimensional organic compound (TMTSF)$_2$PF$_6$ in the close vicinity of the borders between the paramagnetic metal PM, antiferromagnetic insulator AF, and superconducting SC states. In order to drive the system precisely through the phase border $P_0(T_0)$, the sample was maintained at fixed temperature $T$ and pressure $P$, whereas the critical pressure $P_0$ was tuned by applying the magnetic field $B$. In this approach, the magnetic field was used (i) for smooth and precise tuning $\delta P = P - P_0$ (thanks to a monotonic $P_0(B)$ dependence) and (ii) for identifying the phase composition (due to qualitatively different magnetoresistance behavior in different phases). Experimentally, we measured magnetoresistance $R(B)$ and its temperature dependence $R(B, T)$ in the pressure range $(0 - 1)$ GPa. Our studies focus on the features of the magnetoresistance at the phase transitions.
between the PM and AF phases and in the close vicinity to the superconducting transition at $T \approx 1\text{K}$. We found pronounced history effects arising when the AF/PM phase border is crossed by sweeping the magnetic field: the resistance depends on a trajectory which the system arrives at a given point of the $P - B - T$ phase space. In the transition from the PM to AF phase, driven by increasing magnetic field, the features of the PM phase extends well into the AF phase. At the opposite transition from the AF to PM phase, the features of the AF phase are observed in the PM phase. These results evidence for a macroscopically inhomogeneous state, which contains macroscopic inclusions of the minority phase, spatially separated from the majority phase. When the system is driven away from the transition, the homogeneous state is restored; upon a return motion to the phase boundary, no signatures of the minority phase are observed up to the very phase boundary.
I. INTRODUCTION

The interplay (co-existence, segregation, or competition) of the magnetic spin ordering and the superconducting pairing of electrons is in the focus of the broad research interest. These effects are of the key importance for understanding the rich physics of high $T_c$ superconductors, heavy fermion compounds and also organic conductors [1–4]. Indeed, for these materials, having low-dimensional electron systems, the phase diagrams are surprisingly similar on the plane “pressure” $P$ - temperature $T$ (here by “pressure” we mean either externally applied pressure or internal “chemical pressure”, i.e. dopant concentration)- see Fig. 1a. The phase diagrams for these materials include a magnetically ordered phase, metallic, and superconducting phases [1–4].

The origin of the superconducting phase in (TMTSF)$_2$PF$_6$ remains puzzling; there are experimental and theoretical results pointing at a triplet mechanism of electron pairing [5]. Therefore, the issue of the character of the phase border and the origin of the transitions between magnetic, superconducting, and paramagnetic phases (caused, e.g. by pressure changes) become of the fundamental importance [3,6]. The most general approach to the problem was suggested by a symmetry theory, which incorporates descriptions of the magnetic and superconducting phases by introducing a superspin, whose three components correspond to magnetic- and two others to superconducting- order parameters [7]. In the frameworks of this SO(5) symmetry theory, at a certain intermediate pressure there might exist a state in which all components of the superspin (magnetic and superconducting) are nonzero. Such a “microscopically” mixed state possesses magnetic ordering in the superconducting state. There are indeed experimental indications for the existence of a local magnetic order in the high temperature superconductors [8]. Recently, the microscopically mixed state was experimentally found in the heavy fermion compound CeRhIn$_5$ [9].

On a different footing, the microscopically mixed state has been suggested in Ref. [10] to occur in (TMTSF)$_2$PF$_6$ due to lacking of the complete nesting over the entire Fermi surface. On the $P - T$ phase diagram, the mixed state should occupy a narrow strip of pressures just
below the critical $P_{SDW}$ value. At temperature $T = 1.4$ K, this region should be about 4% wide on the pressure scale.

One should admit an alternative possibility, where in the vicinity of the phase border, a macroscopically inhomogeneous state may arise; this state incorporates inclusions of the minority phase embedded in the majority phase. As an example, it is well known, that the two-dimensional Mott type insulators tend to the formation of phase-inhomogeneous states [1,11]. It is also known that martensitic transformations [12] concomitant with phase segregated states take place in such materials, where the free energy of electron system (including the magnetic energy of spin ordering) depends linearly on lattice deformation. If the inhomogeneous state with spatially separated phases emerged on the border of the magnetic and superconducting states (see Fig. 1a), it would have demonstrated simultaneously magnetic and superconducting properties, similarly to those of the heterophase mixed state.

The mixed state, as well as the state with spatial phase-separation, are expected to exhibit similar purely superconducting or purely magnetic properties far away from the phase boundary, so that their behaviors are indistinguishable. However, in the close vicinity of the phase boundary ($T_0, P_0$), properties of these two types of states are different. In principle, one may distinguish these two possibilities, if the system is forced to cross the phase border by varying pressure at constant temperature, along the vertical trajectory in Fig. 1a. In particular, for the inhomogeneous state with inclusions of the minority phase, one might expects such effects as pre-history and hysteresis: the properties of the system at a given point of the $P – T$ phase diagram may depend on the pathway which the system has arrived at this point, due to a path-dependent concentration of segregated phases. In contrast, there is no reason to expect history effects for the mixed state.

Straightforward performing such experiment represents a hard technical task. Nevertheless, such measurements of $R(T)$ at fixed pressure values have been described recently in Ref. [13], where the $R(T)$-dependence was studied for a number of pressure values $P_i$ in the vicinity of $P_0$. The authors of Ref. [13] observed hysteresis effects within the AF phase and fitted the set of the measured $R(T, P_i)$ curves using a simple percolation model (which
modeled the inclusions of one phase into another one). It was concluded that the observed hysteresis in the $R(T)$-dependences reflects a macroscopically inhomogeneous state, i.e. a mixture of the two phases. We note, however, that the identification of the AF and metallic phases is not trivial; at zero magnetic field the signature of the AF state is the onset of the $R(T)$ rise with cooling. Under circumstances when the system may contain a new phase with apriory unknown conduction, such procedure may be potentially ambiguous; therefore, the conclusions made in Ref. [13] require additional verification.

In the current paper, we applied a different experimental approach. Using the magnetic field dependence of the $T - P$ phase border for this compound, we swept the magnetic field at a number of fixed pressure values in the vicinity of $(T_0, P_0)$; the magnetic field caused changes of the phase boundary and the corresponding phase transitions between the AF and PM states. Thus, the magnetic field was used in our experiment for both, driving the system through the phase transition (instead of pressure), and for reliable identifying the phase content.

We observed strong prehistory effects in the resistivity (in the presence of magnetic field), similar to those reported in Ref. [13] for the $B = 0$ case. Besides, we found prehistory effects also in the character of the magnetic field dependence $R(B)$, which occur when the system crosses the phase boundary. These results evidence for the macroscopically inhomogeneous heterophase state in the vicinity of the AF-PM border. Depending on the direction of the magnetic field sweeping, the minority phase extended across the phase border, deep into the majority phase. Observation of the hysteresis and prehistory effects is not consistent with the model of the microscopically mixed (coexisting) antiferromagnetic and paramagnetic states. Thereby, in the current paper we have unambiguously determined that the transition from the AF insulating to PM metallic phase takes place through emergence of a macroscopically inhomogeneous state with spatially separated phases. Thus, our results obtained by independent technique, are in agreement with the preceding data by Vuletić et al. [13].
II. THE IDEA OF THE EXPERIMENT

For the quasi-one dimensional compound \((\text{TMTSF})_2\text{PF}_6\) at zero magnetic field, there is a narrow pressure range in the vicinity of \(P \approx 0.6\ \text{GPa}\), where the two electronic phase transitions take place as temperature decreases. Firstly, there is a transition from the paramagnetic PM metallic phase \([14]\) to the insulating AF phase (spin density wave state), and further, from the AF state to the superconducting (SC) state. Figure 1a shows the corresponding phase diagram \([4,13,15]\), which incorporates the domains of the AF, PM, and SC phases. A vertical trajectory \(P = 0.54\ \text{GPa}\) on the phase diagram corresponds to the temperature dependence of resistance \(R(T)\), shown in the inset to Fig. 1a. Crossings of the phase boundaries are marked with dots on the vertical trajectory (Fig. 1a) and on the measured \(R(T)\) dependence.

A. Traditional approach: varying \(P\) and \(T\)

In order to explore the character of the transition, one has to be able to unambiguously identify the phase character and component content in the vicinity of the phase boundary. Observation of the absolute resistivity solely at zero field can hardly provide the required information, for the resistance changes smoothly and insignificantly in the vicinity of the second order transition. To identify the pure homogeneous PM and AF states, one could, in principle, make use of the temperature dependence of conduction, which has an activated character in the AF phase and diffusive character with the “metallic” sign \(d\sigma/dT < 0\) in the PM phase. In practice, however, this would require \(R(T)\) measurements over a broad temperature range, which is inaccessible in the AF phase. Indeed, for the most interesting regime in the vicinity of the contiguity of the three phases, \(T_0 = 1.3\ \text{K}\) and \(P_0 = 0.61\ \text{GPa}\), the temperature range of the existence of the AF phase is limited both, from the high and low-side (see Fig. 1a). Besides, direct studies of such transition by changing the pressure \(in situ\) (i.e. along horizontal trajectories in Fig. 1a) at low temperatures represent a very
B. Alternative approach: varying $P_0$ and $T_0$

According to our measurements in magnetic field and the earlier results (see, e.g., Ref. [16], the AF/PM border $T(P)$ shifts to higher temperatures as magnetic field grows. Figure 1b shows schematically the changes of the border with magnetic field. Due to the smooth and monotonic dependence of $T_0$ on magnetic field, this dependence may be used for varying $T_0$. Thus, the system may be forced to cross the border by varying the magnetic field at fixed values of pressure and temperature. Figure 1b shows that when the initial $P$ and $T$ values (at zero field) are chosen in the vicinity but bigger than $P_0$, $T_0$, the phase trajectory of the system (trajectory 2) will cross the phase border with magnetic field growth. Thus, crossing the border occurs due to the changes in $T_0(B)$ and $P_0(B)$ at fixed $T$ and $P$ values. Besides, crossing the border in the presence of magnetic field causes qualitative changes in the behavior of magnetoresistance, which are used in the current work for identifying the phase state and phase content of the system.

III. EXPERIMENTAL

Two samples - (TMTSF)$_2$PF$_6$ single crystals have been grown using a conventional electrochemical technique; the typical sample sizes were $2 \times 0.8 \times 0.3$ mm$^3$, along the crystal directions $a$, $b$, and $c$, correspondingly. The two nominally equivalent samples showed qualitatively similar behavior and had slightly different resistivity value at low temperature. Measurements were made by four probe ac lock-in technique at 32 Hz frequency. For electrical contacts, four 25$\mu$m Pt-wires were attached by a graphite conductive paint to the sample along the most conducting direction $a$ at the $a$-$c$ plane. The sample and a manganin pressure gauge were placed inside a nonmagnetic pressure cell [17] filled with Si-organic (polyethilenesiloxane) PES-1 pressure transmitting liquid; a required pressure was created at room temperature. The Ohmic character of the contacts to the sample was confirmed.
by the negligibly small out-of phase component of the measured voltage drop between the contacts.

The pressure cell was mounted in a cryostat in the bore of a 16 T superconducting magnet. Measurements at temperatures $T \geq 1.4 \text{K}$ were done in the cryostat with $^4\text{He}$ pumping. For all measurements, the magnetic field was applied along the least conducting direction, $c$, of the crystal and current was applied along $a$. Temperature of the pressure cell was measured using the RuO$_2$ resistance thermometer, and heat contact of the sample to the liquid helium (or mixture) bath was provided with Pt-wires. In order to implement the idea of measurements with crossing the phase border for the account of magnetic field changes, the pressure value has to be set in the interval $0.62 - 0.64 \text{GPa}$ at $T \approx 1.4 \text{K}$.

IV. THE RESULTS OF MEASUREMENTS

The magnetic field dependence of the resistance is qualitatively different for three different trajectories (1, 2, 3) in the $B - P - T$ phase space, depicted on Fig. 1 b.

A. Trajectory 1

Figure 2 a shows a typical example of the magnetic field dependence of the resistivity, corresponding to the trajectory 1 on Fig. 1 b. For such trajectory, which entirely belongs to the AF domain, changes in the resistivity are not accompanied with hysteresis. Resistance increases monotonically with magnetic field; in strong field, the so called “Rapid oscillations” (RO) appear on the background of the monotonic $R(B)$ growth [18]; such $R(B)$ dependence is typical for the AF phase [19]. The oscillations can be more clearly seen in the derivative $dR(B)/dB$, shown in the inset to Fig. 2 a. As pressure increases (but still remains less than the critical $P_0(B)$ value), the resistance magnitude decreases, whereas $R(B)$ dependence does not change qualitatively.
B. Trajectory 3

When the initial $T, P$ values are chosen essentially larger than $T_0, P_0$, the trajectory 3 of the system (Fig. 1 b) lies entirely in the PM domain over all range of the magnetic field changes. Figure 2 b shows that the magnetoresistance in this case has a character qualitatively different from that discussed above for the AF phase. As magnetic field increases, the smooth growth of $R(B)$ transforms into step-like changes, which are related to the developing cascade of transitions between the states with different nesting vector [4,20–22]. In strong fields and at low temperatures, the transitions between the states with different nesting vector have a character of the 1st order phase transitions [22,23]. Correspondingly, the step-like changes of $R(B)$ in strong fields are accompanied with hysteresis in $R(B)$ [22]. Such hysteresis may be noticed in $R(B)$ traces in strong fields, presented in Fig. 2 b.

The inset to Fig. 2 b shows on the $B − T$ plane the corresponding phase diagram, which includes different sub-phases of the field-induced spin density waves [4,20–22]. On Figure 2 b, the $R(B)$ curve at $T = Const$ corresponds to the isothermal trajectory on the $B − T$ phase diagram (shown in the inset to this figure) which sequentially crosses different subphases. The corresponding jumps in $R(B)$ are periodic in $1/B$ [21,20] and well correspond to the phase boundaries on the known $B − T$ phase diagram of the FISDW-regime (see inset to Fig. 2 b) [22].

Each individual subphase has its own nesting vector and the quantized Hall resistance value [20,21,24]. Indices $N$ for different subphases in Fig. 2 b correspond to the number of filled Landau levels in the quantum Hall effect, and, simultaneously, determine quantized changes of the nesting vector [23]. When pressure $P$ decreases (but still remains bigger than the critical $P_0$ value), the resistance magnitude smoothly increases whereas $R(B)$ dependence does not change qualitatively. The resistance jumps related to the phase transitions persist and monotonically shift to lower fields, thus indicating the shift of the phase boundaries [22].
When the initial $P, T$ values are chosen in the vicinity but slightly bigger than $P_0, T_0$, one can expect the phase trajectory 2 to cross the border with increasing magnetic field, as discussed above. The inset to Fig. 3a shows the $R(T)$ dependence measured at $B = 0$; it evidences for the true metallic initial state of the sample at $P = 0.64$ GPa (which is close to the critical value $P_0 \approx 0.61$ GPa). When magnetic field increases, the resistance changes insignificantly up to $B \approx 7$ T (see Fig. 3a). Upon further increase of $B$ up to 16 T, the resistance sharply grows by 3 orders of magnitude. This growth indicates the transition from the metallic PM to insulating AF phase. On the background of the growing monotonic resistivity component, one can note the appearance of non-monotonic periodic variations of resistance (starting from $B \approx 8$ T), which are absolutely untypical for the AF phase.

As magnetic field is swept down (i.e. decreases from 16 T to 7 T), a strong hysteresis ($\sim 20\%$) is revealed in the resistance (Fig. 3a), whereas the non-monotonic component of $R$ practically disappears. The $R(B)$ hysteresis and the appearance and disappearance of the non-monotonic component of resistance depend only on the absolute magnetic field value and do not depend on its direction (compare $R(B)$ and $R(-B)$ in Fig. 3a). The magnitude of hysteresis grows with magnetic field. Upon repeated magnetic field sweeps from 0 to 16 T, the described above $R(B)$ dependence reproduces fully. The non-monotonic component of the resistance is more clearly seen in the $dR/dB$ derivative shown in Fig. 3b. It is worthy of note that the non-monotonic component is observed only when magnetic field is increased and is practically invisible when magnetic field is decreased from 16 T.

Vertical arrows in Fig. 3b depict the locations of the FISDW phase boundaries, which were experimentally determined in Ref. [22] from the jumps in $R(B)$ versus field in the FISDW area of the pure PM phase. The location of peaks in $dR/dB$ in Fig. 3b coincides well with the arrows (i.e. with the anticipated borders of the FISDW phases). The inset to Fig. 3b demonstrates that these peaks are equidistant in $1/B$. For these two reasons, we may identify the observed peaks in $dR/dB$ with crossing the borders between the FISDW
phases with $N = 6 \leftrightarrow 5$, $5 \leftrightarrow 4$, $4 \leftrightarrow 3$, and $3 \leftrightarrow 2$, correspondingly, on the phase diagram Fig. 2b upon isothermal sweeping the field.

We wish to stress once more that the existence of the peaks and their periodicity in $1/B$ would be quite natural for the PM state, but is absolutely unexpected for the AF state. On the $dR/dB$ dependence, the next peak ($N = 2 \leftrightarrow 1$) at $B \sim 14$ T is not seen, despite the peak amplitude is known to strengthen with decreasing $N$ [22]. The absence of this peak indicates almost complete disappearance of the PM phase and the onset of the homogeneous AF state at the field of $B = 14$ T. As an additional confirmation of this conclusion, we note that in stronger fields $B > 14$ T rapid oscillations may be seen in Fig. 3b; these RO oscillations are characteristic for the AF phase [18].

Magnetic field dependences of the derivative $dR/dB$ are shown in Fig. 4 for different temperatures. One can see that the hysteresis of $R(B)$ in fields sweeping up and down disappears as temperature increases. Note that at low temperature, the hysteresis reveals itself not only in the magnitude of $R(B)$ (and $dR/dB$) but also in the qualitatively different character of the $R(B)$ dependence. When the field is swept up, the $R(B)$ dependence exhibits jumps (marked by arrows in fields $B = 8 − 12$ T). The jumps are characteristic for the FISDW phase transitions in the PM phase, whereas the system passed transformation to the AF phase starting from field $B \approx 6$ T; the strong growth of resistance in Fig. 3a and the appearance of RO evidence for this transformation. When the magnetic field is swept down, these anomalous jumps are almost invisible and one can see only the anticipated RO oscillations [18].

V. DISCUSSION OF THE RESULTS

The most essential results of our studies are as follows:
1) As expected, when the field increases (decreases) along the trajectory 2 (see Fig. 1b), the system exhibits the phase transition. The steep, a factor of $10^3$, raise (fall) of the resistance at $B \approx 6$ T evidence for this phase transformation.
2) At the transition from the PM to AF phase, rather far away from the phase border, the magnetoresistance continues to exhibit residual signatures of the metallic (minority) phase. When the field grows, the signatures of the minority phase almost disappear and are not restored when the system approaches back to the same phase border (i.e. as field decreases). In other words, at such transition, a strong hysteresis is observed both, in the magnitude of $R$ and in the character of $R(B)$ dependence.

3) Upon return transformation from the AF to PM phase (with decreasing field), a hysteresis in the magnitude of $R$ is observed: at the transition, the resistance is noticeably higher than that for the pure metallic PM phase (or than the resistance value measured as the field grows from $B = 0$). The “true” value of $R$ is restored only when the field is decreased to zero.

In view of the complicated character of the magnetoresistance behavior, which exhibits signatures of both phases, the experimental determination of the AF and PM phases becomes of the principle importance. According to the existing theory [26], the SDW-PM transition is expected to be either of the second, or weak first order. Experimental data are in agreement with this conclusion [25]. In the vicinity of the critical pressure the SDW gap $\Delta$, in general, might be small as compared to the antinesting parameter $t'_b$ [26]. In this case, the pseudo-activated $R(T)$ dependence in the SDW phase would have a semi-metallic character and would be indistinguishable from the “metallic” $R(T)$ dependence, thus making the identification of the two phases difficult. However, for the specific 2D tight binding case in (TMTSF)$_2$PF$_6$, $\Delta$ does not depend on pressure in the vicinity of the critical pressure and is not small at the transition [26]. This agrees with experimental observations [27], where $R(T)$ was shown to have a pseudo-activated character with rather big gap in the vicinity of the critical pressure. We use therefore, the sharp growth of $R(T)$ (starting from $B \approx 7$T in Fig. 3 a) as a firm indication of the onset of the SDW phase.
A. Inhomogeneous state: phase separation or phase mixing?

Manifestly, our experimental results do not fit the behavior, anticipated for a microscopically mixed state made of the two coexisting phases. For such a state, the hysteresis effects and the dependence of the phase content on the prehistory, should not occur. The behavior described above is also not typical for a homogeneously “overheated” or “overcooled” phases at the first order phase transitions, for the minority phase disappears smoothly with no sharp jumps in $R$. Besides, for the 2nd order transition in a homogeneous system, neither hysteresis nor “overheating/overcooling” should take place. In the domain of the phase space, where only PM or AF phase should exist, clear signatures of the opposite phase are observed beside the “correct” phase. Therefore, the appearance of the hysteresis and the distinct signatures of the presence of both phases in the same domain of the phase space, both evidence that the phase content of the system becomes inhomogeneous. From a theoretical viewpoint, the cascade of transitions could also exist in the AF phase (accompanied with a corresponding jumps in $R(B)$) [26]. However, such cascade has never been observed experimentally in the AF phase. Furthermore, even if the cascade of transitions occurs, as a homogeneous state, it would not give raise to pre-history effects such as observed in our experiment.

The phase-inhomogeneous state is not a consequence of the inhomogeneity of the sample or of the external pressure. The experimental results which prove this are as follows:

1. The phase-inhomogeneous state was observed on two different samples; the hysteresis and prehistory effects were qualitatively similar in both samples (compare Figs. 3 b and 4).
2. The existence of the prehistory in the appearance of the phase-inhomogeneous state contradicts the assumption of the inhomogeneity of the sample or external pressure. Indeed, if such inhomogeneities exist, they would manifest always, and would not disappear at the field sweeping through the border of the 2nd order transition; therefore, the prehistory effects would not take place.
3. Hysteresis in the character of $R(B)$ dependence arises only at pressure and temperature
values in the vicinity of the phase border \((T_0, P_0)\). No history effects are observed when the system is moved away from the phase boundary in either pressure or temperature axes. This may be seen, e.g. in Fig. 4, where in strong fields \(B > 12\) T, rapid oscillations \([18]\) have the same magnitude and phase for the field sweeping up and down.

4. Low residual value of the resistivity, \(60 \times 10^{-6}\) Ohm-cm (see Fig. 3 a), evidences for high quality of the samples.

One could assume that the phase-inhomogeneous state arises due to a positive surface energy at the border between AF and PM phases, and therefore, the two phases are spatially separated. On the contrary, the microscopic coexistence of the two phases would require a negative surface energy. However, the existence of a noticeable surface energy would mean that this transition is of the well-pronounced 1st order; such assumption seems to disagree with theoretical predictions for the SDW phase transition \([26]\) and experimental data \([25]\).  

**B. Prehistory effects**

The prehistory effect is the most unexpected among the results obtained, even more unexpected than the hysteresis. This phenomenon is illustrated on Fig. 5 b, where four different dependences \(dR(B)/dB\) are compared; these dependences correspond to 4 trajectories \((AB, BC, CD,\) and \(DE)\) shown in Fig. 5 a. When the system crosses the PM-AF phase border (at \(B \approx 6\) T) and moves deep into the AF domain along the trajectory \(AB\), the derivative \(dR(B)/dB\) exhibits peaks (marked with vertical arrows). These peaks correspond well to the resistance jumps observed in the PM phase at crossing the borders between the FISDW sub-phases \(4 \leftrightarrow 3\), and \(3 \leftrightarrow 2\) \([22]\); correspondingly, the peaks have nothing in common with the AF phase in which the system is supposed to be for the given \(P, B, T\) values. The existence of these peaks evidence that, at least, a part of the sample did not transform into the insulating AF phase and remains in the metallic PM phase. In the field \(B \approx 15\) T, instead of the next anticipated peak (which would correspond to the FISDW transition \(2 \leftrightarrow 1\)), one can see only weak oscillations reminiscent of the RO oscillations in
the AF phase. This points out at almost complete disappearance of the PM phase and the onset of the homogeneous AF state.

C. Phase separation at zero magnetic field

In the described above experiments, the presence of the magnetic field was not of a principle importance. The role of the magnetic field was to produce a qualitative difference between the $R(B)$ dependences in the AF and PM phases; this is necessary to identify crossing the border and to reveal the phase content of the inhomogeneous state. In our view, the phase-inhomogeneous state with inclusions of minority phase imbedded into the majority phase must also arise in the transition from PM to AF phase with decreasing temperature (see the phase diagram in Fig. 1a). In this case, however, the resistance changes are anticipated to be weak and of a quantitative rather than qualitative character. Such measurements have been already undertaken in Ref. [13], and our task was to test or confirm these results for same samples in which we have determined the character of the transition in non-zero magnetic field.

For the experiment we have chosen the pressure $P = 0.5$ GPa, which is less than the critical value. The results are represented in Fig. 6. For this pressure and $B = 0$, the system transforms from the metallic to AF state as temperature decreases below $T = 7$ K [4,13]. At the transition, the resistance sharply raises and further grows with decreasing the temperature; this behavior corresponds to the onset of the insulating state (spin density wave). The variations of the resistance with temperature along the trajectory $AB$ are shown in Fig. 6 a. The final resistance value at point $B$ corresponds to the minimal temperature 4.2 K in this experiment.

According to the above assumption, at point $B$ the system has a spatially inhomogeneous phase content: beyond the majority insulating AF phase, it contains also inclusions of the minority metallic PM phase. The following experiment was done in order to check this assumption: the magnetic field was increased from 0 to 16 T; according to the above results,
such strong field should destroy completely the inclusions of the minority phase. Figure 6b shows resistance changes with increase (trajectory $BC$) and subsequent decrease (trajectory $CD$) of the magnetic field. After magnetic field is decreased to zero, the system returns to a state (point $D$) similar to the initial (point $B$). However, magnifying the data in the inset to Fig. 6b reveals a small ($\sim 5\%$) increase in the resistance at point $D$ as compared to that at the initial point $B$. This minor difference evidences for decreasing the share of the well-conducting metallic phase. The observed hysteresis is weak, therefore its interpretation could hardly be done without preliminary studies of much stronger hysteresis effects in magnetic field. The hysteresis in our $R(T, B = 0)$ measurements is essentially weaker than that in Ref. [13]. The reasons for this might be related to a somewhat smaller deviation of the pressure from the critical $P_0$ value in Ref. [13] than in our studies. Indeed, the lower transition temperature $T_{SDW} = 2.5\,\text{K}$ in Ref. [13] as compare to $T_{SDW} = 7\,\text{K}$ in our studies (see Fig. 6a) indirectly indicates for such difference.

As the field varies repeatedly from $D$ to $C$ and back, no irreversible changes in the resistance are observed; we conclude the inclusions of the metallic phase disappeared. Upon further increase of temperature at zero field, the resistance varies along the trajectory $DA$. Repeated coolings reproduce the trajectory $AB$ within $1\%$ accuracy, the result which evidences for restoring the phase-inhomogeneous state.

VI. CONCLUSION

The experiments described above reveal hysteresis in the magnitude of the resistance and the character of its variation with magnetic field, which develop at the transition from metallic to antiferromagnetic insulator state. Furthermore, we found that the behavior of the resistance with magnetic field becomes prehistory dependent. These results evidence unambiguously for the occurrence of the inhomogeneous state in the vicinity of the phase boundary between the PM and AF phases; we conclude that this state consists of inclusions of the minority phase imbedded into the majority phase. No data are available yet on
the spatial arrangement of the two-phase state. The conclusions we drawn, do not depend on any model assumptions about the spatial arrangement of the two-phase state, because for identifying the phase content we used qualitative difference in the magnetoresistance behavior in AF and PM states. Our results are in a good agreement with previous data [13], obtained in a different way.

The observed phenomena of the phase separation at the second order transitions are similar to those typically seen in martensitic transformations. If this analogy is not accidental, it suggests that the free energy of the electron system depends linearly on the charge or SDW distortion. This conclusion requires experimental verification.

The minority phase inclusions can be eliminated completely as the system moves far away from the boundary, deep in the majority phase domain. The hysteresis in the magnitude and in the field dependence of the resistance does not depend on time; it is a stationary and well reproducible effect. The most striking evidence for the heterophase content was obtained from experiments in finite magnetic fields. However, similar phase-inhomogeneous state has been also observed in zero field, for the transition from metallic to antiferromagnetic insulator (SDW)-state. These observations confirm that the magnetic field does not play an essential role in the occurrence of the heterophase state. Extending this analogy to the superconducting transition, we note an interesting possibility that the transition from the antiferromagnetic insulator to superconductor state might also occur via superconducting transition in inclusions of the minority metallic phase, rather than between the two homogeneous AF and SC states. This suggestion also requires an additional experimental verification.

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Figure captions

Fig. 1. a) The $P-T$ phase diagram for (TMTSF)$_2$PF$_6$ in the absence of magnetic field. Difference phases are designated as follows: PM - paramagnetic metal, AF - antiferromagnetic insulator, SC - superconductor. Vertical line shows the isobaric trajectory $P = 0.54$ GPa, which crosses the PM - AF and AF - SC phase borders. The inset shows the temperature dependence of the resistance at $P = 0.54$ GPa, which corresponds to the vertical trajectory at the main panel. Two bold dots at the $R(T)$ curve and at the trajectory mark two corresponding phase transitions, from the metallic PM to the insulating AF state, and from AF to the superconducting SC state. b) Schematic $P-B-T$ phase diagram for the nonsuperconducting phase space ($T \geq 1.14$ K).

Fig. 2. a) Variation of the resistance with magnetic field in the AF state, which corresponds to the motion along the trajectory 1 in Fig. 1 b. In the inset, on the derivative $dR/dB$, one can see the RO oscillations, which are typical for the AF phase. b) Variation of the resistance with magnetic field in the PM state, (trajectory 3 in Fig. 1 b). In high fields, one can see jumps in $R(B)$, which are typical for the PM state and correspond to crossing the boundaries between the FISDW phases with different nesting vector. The inset shows the corresponding phase diagram for the FISDW region, experimentally determined in Ref. [22].

Fig. 3. a) Variation of the resistance with magnetic field for the case of crossing the PM-AF boundary (trajectory 2 in Fig. 1 b). The inset shows $R(T)$ changes in the initial state at $B = 0$, characteristic for the PM metal ($dR/dT > 0$). The sharp growth of $R(B)$ at $B \approx 7$ T corresponds to the PM $\rightarrow$ AF transition. b) Magnetic field dependence of the derivative $dR/dB$, which corresponds to the $R(B)$ curves in Fig. 3 a upon increasing and decreasing field. The vertical arrows show the borders between the FISDW phases, depicted from the experimentally determined phase diagram (see [22] and the inset to Fig. 2 b). The inset demonstrates the periodicity of the $dR/dB$ peaks in $1/B$. Sample #1.
Fig. 4. Magnetic field dependence of the derivative $dR/dB$ for four temperatures. The curves show rapidly disappearing hysteresis as temperature grows, and a characteristic non-monotonic temperature dependence of the rapid oscillations. Sample #2.

Fig. 5. History effects developing in $R(B)$ (panel a) and in $dR/dB$ (panel b) as magnetic field varies along the trajectories $AB$, $BC$, $CD$, and $DE$. Two arrows mark the peaks in $dR/dB$ corresponding to the FISDW transitions in the PM phase; these peaks are missing as field decreases.

Fig. 6. a) History effects in the temperature dependence $R(T)$ at zero field when the trajectory crosses the PM-AF phase boundary; b) Re-establishing the homogeneous state by sweeping the magnetic field to 16 T and back to zero. Pressure is $P = 0.5$ GPa.
FIG. 1.

FIGURES

a) Temperature (K) vs. Pressure (kbar) showing the transitions between AF, PM, and SC phases.

b) 3D plot showing the relationship between Temperature (K), Pressure (GPa), and Magnetic Field (T).
FIG. 2.
\[ T = 1.4\text{K} \]
\[ P = 0.64\text{GPa} \]

**FIG. 3.**

a) 

b) 

\[ \frac{dR}{dB} (\Omega/T) \]

\[ \frac{dR}{dB} (\Omega/T) \]

\[ T = 1.4\text{K} \]
\[ P = 0.64\text{GPa} \]
FIG. 4.
