Evidence for fast oscillations vanishing at the spin-density-wave–metal transition in Bechgaard salts

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We have studied two Bechgaard salts $(TMTSF)_2X$ with different anion geometry, namely octahedral ($X=PF_6$) and triangular ($X=NO_3$), in pulsed magnetic fields up to 37 T at ambient pressure in the temperature range 3–13 K. The temperature and magnetic-field dependence of the amplitude of the magnetoresistance oscillations have been studied. For both salts, the results unambiguously demonstrate that fast oscillations are limited to the spin-density-wave state. In addition, a recent model based on Fermi-surface reconstruction of the $(TMTSF)_2PF_6$ salt cannot account for the magnetic-field dependence of the fast oscillation amplitude.

I. INTRODUCTION

Bechgaard salts $(TMTSF)_2X$, where TMTSF stands for tetramethyltetraselena-fulvalene and X is an inorganic anion (such as PF_6, AsF_6, NO_3, ClO_4…), are well known quasi-one-dimensional conductors. By varying the applied pressure or the magnetic field, their low-temperature state can be either spin-density wave (SDW), superconducting, metallic or field-induced SDW. In some of these salts, Shubnikov–de Haas-like (SdH)[1-3] conductivity oscillations were observed at low temperature and high magnetic field. De Haas–van Alphen (dHvA) magnetization oscillations were also observed in the relaxed $(TMTSF)_2ClO_4$ salt, only.[4] These oscillations, usually called fast oscillations (FO), have a frequency of around 200 T.

In Bechgaard salts for which the anion possesses an octahedral geometry, only one series of FO is observed at low temperature. Their frequency exhibits an anion volume dependence [from 230 T for PF_6 down to 170 T for SbF_6 (Ref. 5)]. Due to the triangular geometry of the NO_3 anion, the $(TMTSF)_2NO_3$ salt undergoes an anion ordering (AO) transition from a metallic to a semimetallic state at 41 K. In this salt, two series of oscillations have been observed in the magnetoresistance.[3] One of them, which rises up at ~16 T with a frequency of 245 T, corresponds to the FO. The other one, with a frequency of 64 T, can be observed at lower field and has been associated with residual pockets resulting from the imperfect nesting of the Fermi surface at the SDW transition.

At present, there is no clear interpretation of the FO phenomenon, since the conventional Landau quantization model for the SdH and dHvA effects cannot account for the observed frequencies. Recently, fermiological approaches based on Fermi-surface (FS) reconstruction and taking into account magnetic breakdown (MB) and Bragg reflection phenomena have been proposed in order to interpret the data recorded on $(TMTSF)_2PF_6$,[6,7] $(TMTSF)_2ClO_4$,[8] and $(TMTSF)_2NO_3$.[9] In $(TMTSF)_2PF_6$, the FS reconstruction model predicts that the oscillations should appear only in the SDW state. In $(TMTSF)_2NO_3$, FO have been associated with bona fide closed orbits, induced by the anion ordering, which can be reconstructed by MB in the SDW state.[9] As a consequence, these oscillations should also be visible in the metallic state, as suggested in Ref. 9.

In $(TMTSF)_2PF_6$ and $(TMTSF)_2NO_3$, FO were observed at low temperature (<8 K) within the SDW state. Up to now, no additional data are available to decide if these oscillations are connected with the SDW state, or if they could be visible in the metallic state [as it is the case of $(TMTSF)_2ClO_4$ (Ref. 2)].

In order to solve this problem, we have thoroughly studied the temperature dependence of the FO in the Bechgaard salts $(TMTSF)_2PF_6$ and $(TMTSF)_2NO_3$ up to 37 T, at ambient pressure, in the range from 2 to 14 K, i.e., in both the SDW and metallic states. We *definitely show* that, for both salts, *fast oscillations of the magnetoresistance are limited to the SDW state*. In addition, the FS reconstruction model is discussed through the temperature and magnetic-field dependence of the FO amplitude.

II. EXPERIMENT

The experiments were carried out in the Toulouse pulsed magnetic-field facility. The pulses are characterized by a long decrease time (~1 s) and a maximum field of 37 T. Contacts on the sample were done on pre-evaporated gold contacts with graphite paint using 17 μm gold wires. Resistance was measured using the standard four-contacts technique, with a lock-in amplifier at a frequency of 20 kHz. The current, of low enough amplitude to avoid non-Ohmic effects, was injected through the needle along the *a* axis. A rotating sample holder allowed us to rotate the magnetic field

in the plane perpendicular to *a*. The magnetic field was oriented parallel to b' (the projection of *b* onto the plane perpendicular to *a*) using the anisotropy of the magnetoresistance at liquid-helium temperature, which exhibits a deep minimum for this field direction. A 90° rotation allowed the c^* axis to be aligned along the magnetic field.

III. RESULTS

As already reported, a single FO series is observed for the $(TMTSF)_2PF_6$ salt, while two series arise, respectively, at low and high (~16 T) field for $(TMTSF)_2NO_3$. For $(TMTSF)_2PF_6$, Fourier transforms show a very small second harmonic (less than 5%), which allows us to directly measure the peak-to-peak FO amplitude. For $(TMTSF)_2NO_3$, a direct measurement of the FO amplitude is not possible due to mixing of the two series at high field. In this latter case, the oscillation amplitude has been measured using Fourier transform.

The temperature and field dependences of FO amplitudes have been analyzed in the framework of the conventional Lifshitz-Kosevich (LK) model:

$$\frac{R(B)-R_{\text{background}}}{R_{\text{background}}}=\sum_n (-1)^n A\cos\left(2\pi n\left(\frac{F}{B}-\gamma\right)\right), \quad (1)$$

where

$$A=A_0\frac{u_0 T m_c (n/B)^{1/2}}{sh(u_0 T m_c n/B)}\exp\left(-\frac{u_0 T_D n m_c}{B}\right).$$

A_0 is a constant, $u_0=2\pi^2 k_B m_0/\hbar e=14.694$, m_c is the effective cyclotron mass in m_0 units, T_D is the Dingle temperature, *n* is the harmonic order, and γ is the Onsager phase factor. Figures 1 and 2 display the temperature dependence of the FO amplitude for $(TMTSF)_2PF_6$ and $(TMTSF)_2NO_3$, respectively. In these figures, conventional plots of $\ln(A/T)$ versus temperature are presented. In Fig. 1, data for the oscillation at $B=32.2$ T are considered. In Fig. 2, data from three different samples are presented, with a window field covering three oscillations centered at 26.6 T. In addition, the lower inset to Fig. 1 shows oscillatory magnetoresistance for the PF_6 salt, where the FO can be seen to vanish at high temperature.

Full lines in Figs. 1 and 2 are best fits to Eq. (1). A good agreement between fit and experimental data is obtained in the temperature range 4–9 K (11 K) for $(TMTSF)_2NO_3$ [$(TMTSF)_2PF_6$]. The effective cyclotron mass is field independent in the range 22–32.2 T for both salts, namely, $m_c=0.44\pm0.05$ for the NO_3 salt and 1.35 ± 0.05 for the PF_6 salt. It can be remarked that, contrary to FO, the measured effective cyclotron mass of the slow oscillations in the NO_3 salt is field dependent (from 0.45 to 0.75 in the range 8.3 T to 13.7 T).[10] A downward deviation from the LK model can be observed both in the lower and the higher temperature ranges. The decrease of the FO amplitude below 4 K, which is a quite common characteristic of the FO,[11] could be due to an additional phase transition at low temperature, as suggested by specific heat[12] and ^{77}Se NMR (Ref. 13) measurements. The steep decrease of the FO amplitude in the temperature range close to the SDW transition temperature cannot be a consequence of the thermal damping, and has to

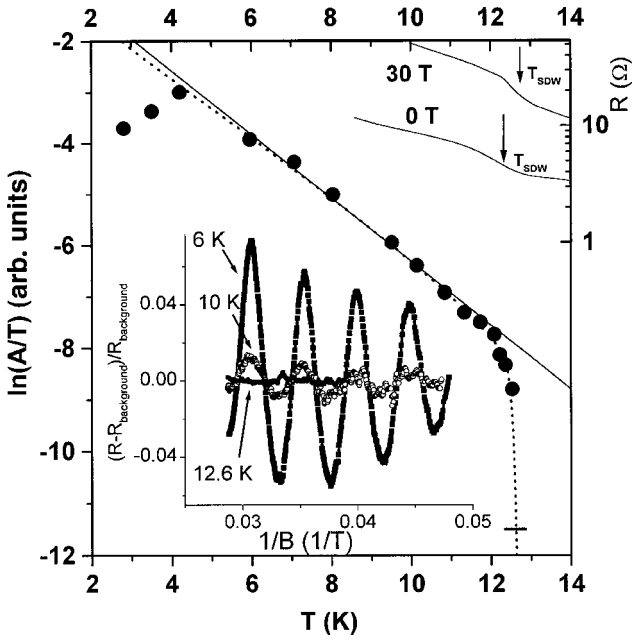

FIG. 1. Temperature dependence of the fast oscillation amplitude at 32.2 T for the $(TMTSF)_2PF_6$ salt. Solid and dotted lines are best fits to Eqs. (1) and (2), respectively. The horizontal dash stands for the noise level for the measurement at $T=12.6$ K. Upper inset: temperature dependence of the resistance in zero field and at 30 T; lower inset: fast oscillations vs inverse field at the temperatures indicated.

be explained by the transition from the SDW state into the metallic one. Indeed, the insets of Figs. 1 and 2 show the temperature dependence of the resistance at 0 and 30 T. Such data allow us to derive the field dependence of the transition temperature (T_{SDW}).[14,15] At 30 T data yield $T_{\text{SDW}}=12.5$ K and 10.6 K for $(TMTSF)_2PF_6$ and $(TMTSF)_2NO_3$, respec-

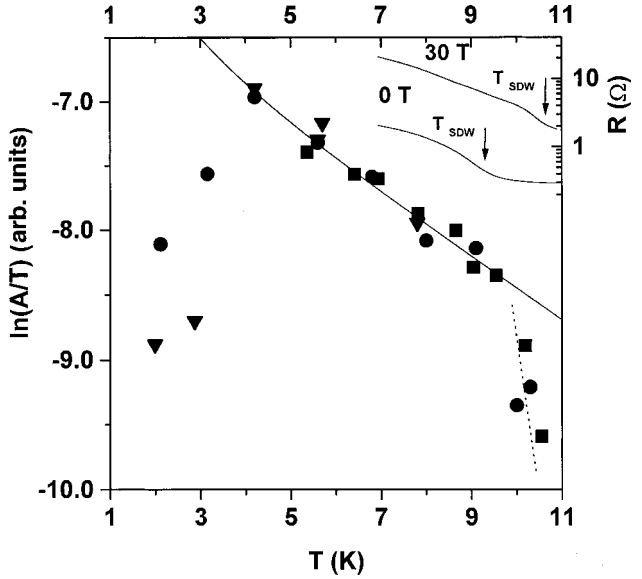

FIG. 2. Temperature dependence of the fast oscillation amplitude, deduced from the Fourier transform, centered at 26.6 T (see text) for the $(TMTSF)_2NO_3$ salt. Data collected on three samples, labeled by three different symbols, is displayed. Solid line is best fit to Eq. (1). The inset displays the temperature dependence of the resistance in zero field and at 30 T.

tively. T_{SDW} lies in the range where the FO vanish, which demonstrates that the FO are restricted to the SDW state in these Bechgaard salts. As a consequence, the FO cannot be linked to the 2D orbits induced by anion ordering in $(TMTSF)_2NO_3$ unless the transition into the metallic state induces a dramatic decrease of the carrier mobility. In that respect, it has been suggested that umklapp electron-electron scattering strongly decreases the quasiparticle mean free path of the metallic state of the $(TMTSF)_2PF_6$ salt.[16] It should be noted that Kang *et al.*[17] reported quantum oscillations with a frequency of 190 T in the pressure-induced metallic state of the $(TMTSF)_2NO_3$ salt. Keeping in mind that the FO frequency is 246 T these oscillations cannot be accounted for by the fermiological model of Kishigi and Machida.[9] Indeed, the pressure effect is to increase both the first Brillouin-zone area and the amplitude of the warping, thus leading to an increase of the area of the two-dimensional (2D) tubes induced by anion ordering. In our opinion, the frequency observed in Ref. 16 might correspond either to the slow frequency ($F=64$ T at ambient pressure) or to an additional series.

Uji *et al.* have proposed an interpretation of the FO in $(TMTSF)_2PF_6$ based on a fermiological approach, taking into account a Fermi-surface reconstruction due to the SDW transition.[6] This model allows to calculate the evolution of the FO versus temperature and magnetic field [as it has been done recently by Brooks *et al.* up to 9 K (Ref. 7)] using an LK formula modified by the probability of four magnetic breakdowns and two Bragg reflections:

$$A=A_0\frac{u_0Tm_c(n/B)^{1/2}}{sh(nu_0Tm_c/B)}\exp\left(-\frac{u_0T_Dm_c}{B}\right)P^4(1-P)^2. \quad (2)$$

Here, the probability of a single magnetic breakdown is expresses as $P=\exp(-B_{MB}/B)$. The LK model can be further modified by taking into account the temperature dependence of the magnetic breakdown field due to the gap Δ opening at the SDW transition. This is done by using $\Delta(T)$ as determined in μsr measurements by Le *et al.*,[18] and the fact that B_{MB} is proportional to Δ^2.[19] The dotted line in Fig. 1 is the best fit of Eq. (2) to our data, using $m_c=1.2$ and $T_{SDW}=12.5$ K. It should be mentioned that, due to the lack of sensitivity of the fit to the value of B_{MB}, a meaningful value for this latter parameter cannot be deduced unambiguously from the fit. At this point, we can only say that the good agreement between this model and the experimental data in the temperature range higher than 4 K suggests that this model *could* be compatible with the temperature dependence of the FO amplitude, in particular at the SDW transition temperature. As mentioned above, an increase of the scattering rate at the SDW transition may also account for the oscillation amplitude damping.

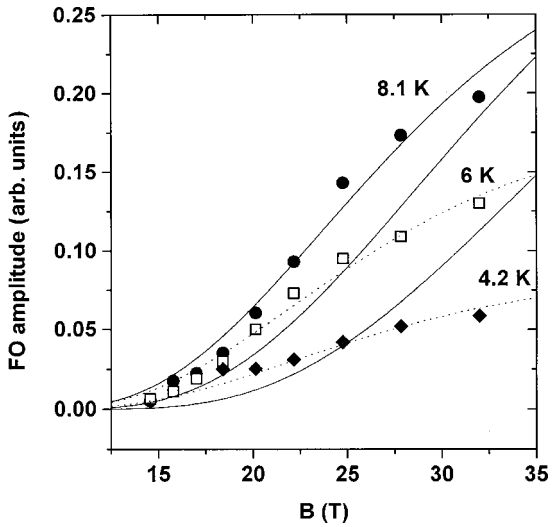

FIG. 3. Magnetic-field dependence of the fast oscillation amplitude at different temperatures for the $(TMTSF)_2PF_6$ salt. The solid lines are obtained keeping m_c, T_D, and B_{MB} fixed, while for the dotted lines, only B_{MB} varies.

If the Fermi surface reconstruction model is correct, it should also work for the field dependence of the FO amplitudes, which is shown at various temperatures in Fig. 3. Data analysis has been made in two ways: (1) keeping m_c, T_D, and B_{MB} fixed at values for 4.2 K and (2) keeping m_c and T_D fixed, and letting B_{MB} vary. The solid (dotted) lines in Fig. 3 are best fits to Eq. (2) for case 1 (case 2). Extremely poor reproduction of data is found between Eq. (2) and data for case 1, and only rough agreement is achieved for case 2. As it was the case for the temperature dependence of the FO reported above, the Eq. (2) parameters involved, in either case, cannot be determined unambiguously. In principle, case 2 should be considered the more physically meaningful of the two, since the magnetic breakdown field is expected to change with T through the gap variation [i.e., $B_{MB}\sim\Delta^2$, and $\Delta=\Delta(T)$]. However, the B_{MB} value used in case 2 is found to change by almost a factor of 1000 from 4.2 to 8.1 K. This huge variation cannot be accounted for by the T dependence of the SDW gap, which should lead to a change of the order of 10% in B_{MB} over this temperature range. This excludes the FS reconstruction model[6] as a consistent explanation of the field-dependent data.

In summary, the main point of this paper is that the fast oscillation phenomenon in the quasi-1D $(TMTSF)_2X$ materials $X=PF_6$ and NO_3 is a property of the SDW state only, with the FO vanishing at T_{SDW}, where the transition to the un-nested metallic state occurs. We also analyzed the field dependence of the FO amplitudes in the SDW state, and found that a proposed Fermi-surface reconstruction model cannot explain the data.

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