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High-field magnetoresistance of the Bechgaard salt $(\mathrm{TMTSF})_2\mathrm{AsF}_6$: Fast oscillations and spin-density-wave transition

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We report a detailed study of the transverse magnetoresistance of the Bechgaard salt $(\mathrm{TMTSF})_2\mathrm{AsF}_6$ at ambient pressure, in fields up to 37 T, in the temperature range 1.8–30 K. Fast oscillations (FO's), already evidenced by our group two years ago, have been studied versus the tilt angle of B with respect to the least conducting (c^*,b') plane at 4.2 K. Only one series, having a fundamental frequency of 208 T for $B \| c^*$, has been observed in a temperature range which seems limited to the spin-density-wave (SDW) state domain ($T<11$ K). As with other Bechgaard salts, the oscillation amplitude exhibits a low-temperature maximum located here around 3 K. We have also studied the behavior of the mean magnetoresistance which exhibits, in addition to a local maximum near 3 K, a second one at the metal-SDW transition temperature T_{SDW}. A shift of this later towards higher values has been observed when the field is increased. A comparison with other TMTSF salts exhibiting the FO phenomenon is presented. [S0163-1829(97)00705-4]

The so-called Bechgaard salts, $(\mathrm{TMTSF})_2X$, where TMTSF stands for tetramethyltetraselenafulvalene and X is an inorganic anion (PF_6, ClO_4,NO_3, AsF_6,...) have been extensively studied during the past 15 years, and are now well known to exhibit very rich physics linked to their quasi-one-dimensional behavior.[1] Among these molecular metals, the compound with anion $X=\mathrm{AsF}_6$ has been less studied than others, especially in the high magnetic-field regime. This salt and $(\mathrm{TMTSF})_2\mathrm{PF}_6$ are believed to be rather similar: very close crystalline and band parameters,[2] no anion ordering (both PF_6 and AsF_6 are centrosymmetric anions) and also a metal to spin-density-wave (SDW) transition occurring around 11.5 K (Ref. 3). The search and discovery[4] of a single series of fast oscillations (FO's) in the high-field magnetoresistance (MR) of AsF_6 was a strong motivation to obtain additional high-field data in order to compare it with the ''brother'' PF_6 and other materials exhibiting FO's.

The name FO's was coined to distinguish them from the lower frequency field-induced SDW transitions in the same materials. To date, FO's have been seen on the five Bechgaard salts $X=\mathrm{ClO}_4$,[5,6] PF_6,[7] $R\mathrm{O}_4$,[8,9] NO_3,[10,11] and AsF_6 (Ref. 4) but presently one can say that no general theory is able to explain all the different features of these oscillations, despite the large data base now available on this phenomenon and the numerous models developed by theorists. Simply stated, it remains difficult to explain their origin since there should be *no closed orbits* on the quasi-one-dimensional Fermi surface. The present work investigates the correlation between the temperature and magnetic-field dependences of these FO's and the ambient magnetoresistance. We have considered the mean MR and its behavior mainly around the metal-SDW transition, where a maximum is observed. On the other hand, a shift of the transition temperature T_{SDW} due to the magnetic field is evidenced on the resistivity. These two effects are reminiscent of previous observations on NO_3.[12,13]

The experiments were carried out in the pulsed magnetic field facility at Toulouse, which is characterized by a long decrease time close to 1 s. The standard four-contact technique (gold paste and 17 μm gold wires) was used for the mounting of samples. The resistivity and the transverse $\mathrm{MR}(B,T)\equiv[R(B,T)-R(0,T)]/R(0,T)$ was measured using a 20 kHz ac current of low enough amplitude to avoid heating or non-Ohmic effects, flowing along the best conduction needle axis (a axis). A rotating sample holder allows a 360° variation of the (B,c^*) angle where B is tilted in the plane (c^*,b') for the study of the transverse magnetoresistance angular dependence. As usual, to get fewer cracks (we have noticed that AsF_6 compound seems more sensitive to cracks than other Bechgaard salts), the samples were slowly cooled down to 4.2 K (cooling rate not exceeding 0.1–0.2 K min^{-1}) so that the expected behavior of the resistivity was obtained, metallic down to about 11.5 K followed by a sharp increase when the SDW phase is installed. The temperature T_{SDW} of the metal to SDW transition (with or without applied B) was conventionally estimated from the extrema of $d(\ln R)/d(1/T)$ as for the NO_3 salt.[14] For the present series of experiments, we have worked on two different samples from the same batch: one allowed us to study the MR in a wide range of temperatures from 1.8 to 30 K, B being aligned along the c^* direction, while on the other we have explored the angular behavior of the oscillations at a fixed temperature of 4.2 K

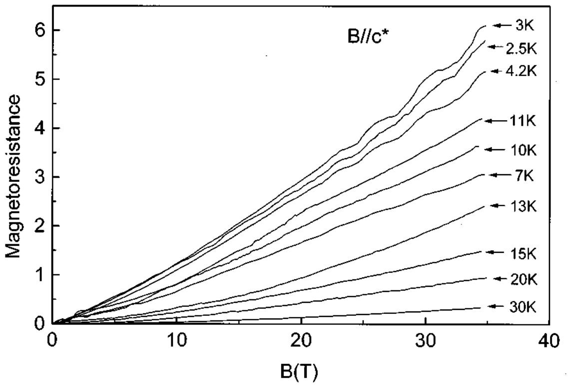

FIG. 1. Transverse magnetoresistance of $(TMTSF)_2AsF_6$ recorded at different temperatures for $B \| c^*$ (sample 1).

[the crystal orientation was achieved in the usual way for TMTSF salts by assigning the b' direction, the projection of the b axis onto a plane perpendicular to a, to the field orientation which gives the lowest MR (Ref. 15)].

The two studied samples exhibit rather similar behavior. However, the absolute magnetoresistance is sample dependent, certainly linked to the amount of defects created during the cooling process or to the intrinsic quality of each sample. In Fig. 1 we give a selection of different MR curves covering a part (2.5 to 30 K) of the experimental temperature range explored. The mean MR has a smooth B dependence and no evolution towards a saturation is visible even at low temperature. The magnitude of the MR is moderate, reaching a value close to 6 at (3 K, 35 T). However some samples have exhibited a much larger MR.

A more comprehensible way of seeing the MR is displayed in Fig. 2 where we have plotted the transverse MR $(B \| c^*, I \| a)$ against temperature for different magnetic fields up to 30 T. The maximum observed around T_{SDW} is reminiscent of what is observed on the NO_3 salt.[12] This large effect could be accounted for by some enhancement of thermal fluctuations of the order parameter[16] together with a field-induced increase of the transition temperature T_{SDW}. To evidence this shift we have plotted the resistance (Fig. 3) and its logarithmic derivative $d(\ln R)/d(1/T)$ (Fig. 4) against the temperature for different B values up to 30 T. The final results are given in Fig. 5 where T_{SDW}, taken at the different maxima in Fig. 4, exhibits a field dependence in good qualitative agreement with Bjelis and Maki calculations[17] which applied very well to the case of NO_3.[13] However, the shift seems to be smaller in AsF_6 ($\Delta T_{\mathrm{SDW}}/T_{\mathrm{SDW}} \approx 9\%$ at 30 T) than in NO_3 ($\Delta T_{\mathrm{SDW}}/T_{\mathrm{SDW}} \approx 14\%$ at 30 T). For perfect nesting, the parameter $\epsilon_0 = t_b^2/\sqrt{2}t_a$ defined by Yamaji[18] should be equal to zero and yield no shift effect. In PF_6 and AsF_6, the nesting is believed to be better than in NO_3, where pockets are left by the anion ordering along the a axis, and thus a smaller shift effect is expected. This is in agreement with the lower shift indeed observed for T_{SDW} in the AsF_6 salt.

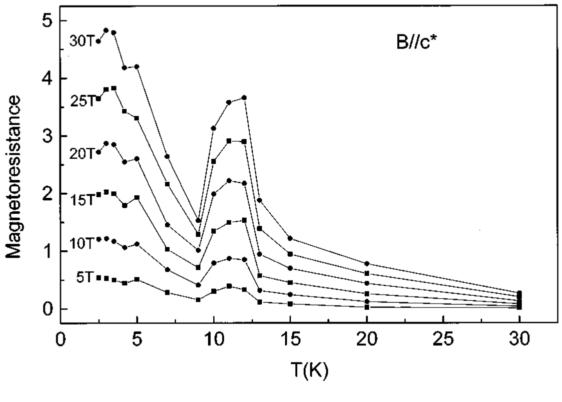

FIG. 2. Temperature dependence of the mean magnetoresistance for different magnetic fields, showing pronounced peak near T_{SDW} (sample 1).

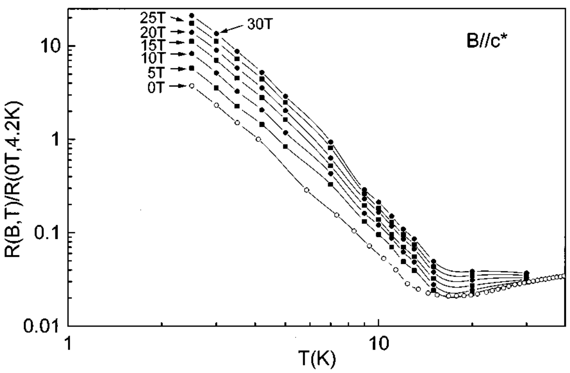

FIG. 3. Temperature dependence of the normalized resistance for different magnetic fields (sample 1).

For fields higher than 20 T, oscillations are directly visible on the MR in the range 1.8–9 K and can be extracted using a compensation of the background by a polynomial function (see Fig. 6). It is worth noticing that, like in the PF_6 salt, only one series is observed, which seems to hold for ‘‘simple bands.’’ On the contrary, compounds affected by a band reshaping coming from an anion ordering exhibit a much more complicated structure, with two series having the same frequency [for ClO_4 (Ref. 5)] or different frequencies [for NO_3 (Ref. 11)]. For $B \| c^*$, in good agreement with the first determination,[4] the frequency of this single series has been found to have a mean value of $B_F = (208 \pm 4)$ T either from the Fourier transforms or from the Onsager relation originally used for Shubnikov–de Haas oscillations:

$$1/B_n = (n + \gamma)/B_F, \tag{1}$$

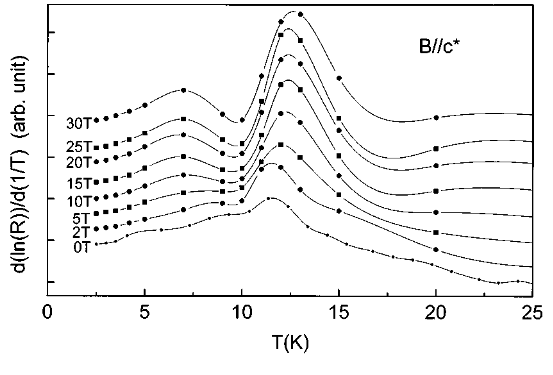

FIG. 4. Temperature dependence of $d(\ln R)/d(1/T)$ for different magnetic fields, showing field dependence of the SDW transition temperature (sample 1).

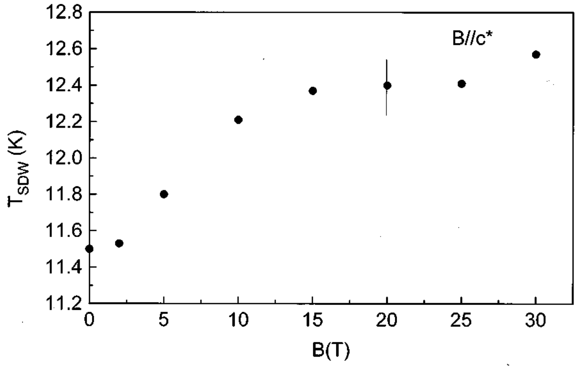

FIG. 5. Magnetic-field dependence of T_{SDW} derived from Fig. 4.

where B_n is the field value at the maximum of the nth oscillation (n are integers chosen to have a γ value lying between 0 and 1). As for FO's in other Bechgaard salts, a good agreement with relation (1) is obtained indicating true periodicity in $1/B$. The value of B_F has to be compared to that of the PF_6 salt, which is very close to 230 T under comparable conditions.[7] The difference is not very large ($\approx$10% lower) but impossible to explain without a general and detailed theory of FO's in Bechgaards. The amplitude of the four main oscillations (labeled as $n=6,7,8,9$ from the Onsager plot) obtained from the MR has been estimated for different temperatures. A rather sharp maximum at 3 K is obtained (see Fig. 7). The decrease of the amplitude towards the low-temperature side after a maximum is typical of the FO's and has been verified in all the salts where they exist.[7,19,11] This behavior completely distinguishes the FO's from conventional Shubnikov–de Haas oscillations, which never weaken when temperature is lowered, and its origin is certainly linked to the intrinsic nature of the FO's. A recent and interesting paper from Kishigi and Machida[20] brings in additional matter to explain this strange behavior in salts undergoing an anion ordering. If the FO's are due to some closed orbits arising from a magnetic breakdown linked to the SDW gap, the opening of this later gap when lowering the temperature strongly decreases the magnetic breakdown probability and leads to the vanishing of FO's. The competition of this phenomenon with the normal thermal increase of the oscillations towards the low-temperature side could explain the maximum generally seen. The same observation done on PF_6 and AsF_6 which present no anion ordering is a good clue that some SDW gap-dependent magnetic breakdown exists. The problem of the vanishing of the FO's around T_{SDW} is not completely solved: we cannot be sure whether it comes from the normal thermal (Dingle) damping like for the Shubnikov–de Haas oscillations or from an effect directly linked to the nesting destruction when the compound recovers its metallic state. In NO_3, the analysis of the data seems to support the later hypothesis[21] but no argument allows presently a definitive choice as theoretical works consider also the possibility of having magnetic oscillations even in the metallic state.[22,23] We must not exclude the possibility of both thermal damping and the disappearance of the SDW state as being responsible for the FO's vanishing on the high-temperature side.

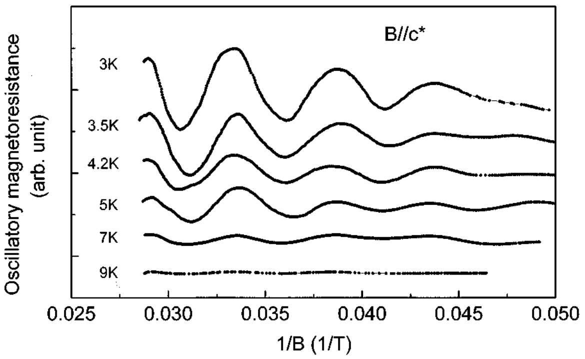

FIG. 6. Oscillatory part of the magnetoresistance at different temperatures extracted from Fig. 1.

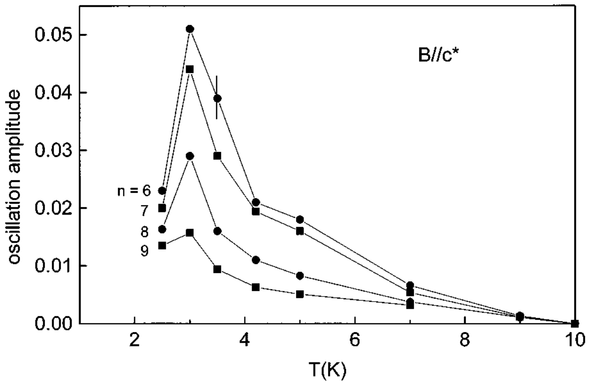

FIG. 7. Amplitudes of the four largest oscillations $n=6,7,8,9$ as functions of temperature, showing rapid decrease below 3 K (sample 1).

Within experimental error, the angular dependence of the oscillations, checked at helium temperature, seems to indicate that only the component of B along c^* is involved, giving the usual cosine law for the variation of $B_F(\theta)$ with $\theta=(B,c^*)$:

$$1/B_F(\theta)=\cos\theta/B_F(0). \tag{2}$$

This latter is plotted in Fig. 8. The angular behavior of the amplitude has also been studied and is given in Fig. 9 for a range of ±50° apart the c^* direction. A rather good symmetry about $B\|c^*$ is obtained and as for the frequency, there is

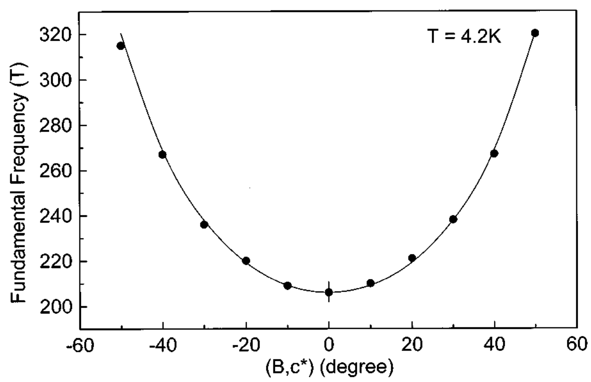

FIG. 8. Angular dependence of the fundamental frequency $B_F(\theta)$ at 4.2 K (sample 2). The full line represents an inverse cosine law.

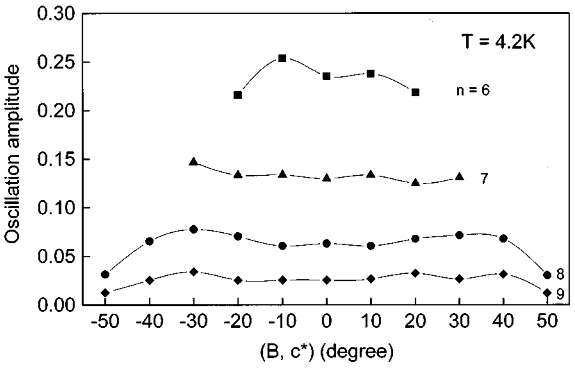

FIG. 9. Angular dependence of the amplitude of FO's $n=6,7,8,9$ at 4.2 K (sample 2).

no obvious departure from a simple two-dimensional (2D) behavior. This is very much different from NO_3 salt where a clear asymmetry for positive and negative angles[12] is certainly connected to the 3D character of the Fermi surface. Also, note that in PF_6 a slight deviation from a true cosine law has been reported[7] but very close to the experimental error. We have also plotted the mean MR angular dependence (not displayed in this paper) which presents also a symmetrical behavior for the same angle away from the c^* direction. No magic-angle effect is seen but also the rather large angular steps (10°) of our measurements prevent us from seeing any detailed angular structure.

In conclusion, we have explored the high-field, ambient pressure MR of $(TMTSF)_2AsF_6$ and shown that FO's share many similarities with PF_6: only one series is observed, with rather close frequencies, and also a maximum of amplitude when the T dependence is considered. The problem of the high-temperature vanishing of the FO's is not completely clear but seems linked to the recovery of the metallic state. Certainly it would be worthwhile to do additional and more complete experiments on $(TMTSF)_2PF_6$ to allow an accurate comparison between the two salts concerning the temperature and angular behavior. Moreover, the shift effect on T_{SDW}, not studied on PF_6, should be measured and quantified, including the use of magnetic probes. If the FO can qualitatively be explained in materials undergoing an anion ordering (ClO_4 and NO_3), either with bona fide closed orbits or Stark quantum interference via magnetic breakdown orbits,[24,25] it is clear that the case of PF_6 and AsF_6 remains unexplained.

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