PHYSICAL REVIEW B **71**, 020404(R) (2005)

Field-induced spin-density wave in $(TMTSF)_2NO_3$

David Vignolles, Alain Audouard,* Marc Nardone, and Luc Brossard
Laboratoire National des Champs Magnétiques Pulsés (UMR CNRS-UPS-INSA 5147), 143 avenue de Rangueil, 31432 Toulouse cedex 4, France

Sabrina Bouguessa and Jean-Marc Fabre
École Nationale Supérieure de Chimie de Montpellier, 8 rue de l'École Normale, 34296 Montpellier cedex 5, France

(Received 30 September 2004; published 19 January 2005)

Interlayer magnetoresistance of the Bechgaard salt $(TMTSF)_2NO_3$ is investigated up to 50 T under pressures of a few kilobars. This compound, the Fermi surface of which is quasi-two-dimensional at low temperature, is a semimetal under pressure. Nevertheless, a field-induced spin-density wave is evidenced at 8.5 kbars above ~20 T. This state is characterized by a drastically different spectrum of the quantum oscillations compared to the low-pressure spin-density wave state.

DOI: 10.1103/PhysRevB.71.020404   PACS number(s): 75.30.Fv, 71.18.+y, 72.15.Gd

Bechgaard salts $(TMTSF)_2X$, where TMTSF stands for tetramethyltetraselenafulvalene and X is an inorganic anion, have been widely studied over the past 20 years for their very complex (pressure, magnetic field, temperature) phase diagrams that involve quasi-one-dimensional (q-1D) metallic, spin-density wave (SDW), superconducting and field-induced spin-density wave (FISDW) states (for a review, see Ref. 1). Nevertheless, FISDW phenomenon is still attracting experimental[2] and theoretical[3] studies. In the framework of the quantized nesting model[4] and its latest refinements,[3] orbital effects stabilize SDW states in compounds with q-1D Fermi surface (FS) at low temperature through the increase of the 1D character of the electronic movement. Furthermore, it has also been stated that a FISDW can only occur provided the ground state is superconducting.[5] In line with both of these predictions, a FISDW has so far only been observed in Bechgaard salts with both a superconducting ground state and a q-1D FS, e.g., in slowly cooled $(TMTSF)_2ClO_4$ at ambient pressure or $(TMTSF)_2PF_6$ under pressures of a few kilobars. In contrast, a FISDW has also been reported in $(DMET\text{-}TSF)_2AuCl_2$ which remains a q-1D metal at least down to 42 mK.[6] It should also be mentioned that a field-induced insulating phase, essentially driven by Pauli effects, has been evidenced in τ-phase organic metals with q-2D FS.[7] The aim of this paper is to argue that, at variance with the above theoretical statements, a FISDW state can be observed in a q-2D semimetal, namely, $(TMTSF)_2NO_3$ under pressure.

Among the members of the Bechgaard salts family, $(TMTSF)_2NO_3$ exhibits numerous peculiar features. As the temperature decreases, an anion ordering (AO) transition with wave vector (1/2, 0, 0) is observed at ambient pressure at a temperature $T_{AO} \simeq 45$ K,[8,9] which remains independent of the magnetic field up to at least 36 T.[10] This leads to a q-2D FS with compensated electron and hole tubes.[11] At lower temperature, a SDW state with incommensurate wave vector and imperfect nesting is stabilized ($T_{SDW}=9.1$ K at ambient pressure).[12] Under pressure higher than ~8 kbars, the AO transition is shifted towards slightly higher temperatures[13] and a metallic state is stabilized at low temperature. Asymmetrical warping of the FS along the less conducting direction c^* has been inferred from the absence of Yamaji features in the angular dependence of the magnetoresistance both in the SDW (Ref. 14) and metallic[15] states. Unlike other Bechgaard salts, no superconducting transition has been observed at temperatures down to 0.5 K.[13,15] As it was expected for a q-2D metal, no sign of a FISDW has been evidenced in $(TMTSF)_2NO_3$ for magnetic fields up to 30 T.[13,15] The absence of a FISDW in this salt has also been interpreted on the basis of a nested 2D excitonic phase.[16]

As it is the case for other Bechgaard salts, $(TMTSF)_2NO_3$ exhibits fast oscillations of the magnetoresistance in the SDW state at ambient pressure with frequency $F_H=(248\pm5)$ T. Unlike the other Bechgaard salts, a second oscillation series with frequency $F_L=(63\pm2)$ T is observed. This frequency is very close to the frequency of the magnetoresistance anomalies linked to the FISDW cascade in other salts. E.g., frequencies of 76 T and 60 T have been reported for $(TMTSF)_2PF_6$ at 6.9 kbars (Ref. 17) and 8 kbars,[18] respectively. However, the oscillations observed in $(TMTSF)_2NO_3$ have a clear sinusoidal shape,[19,20] which is not the case of the magnetoresistance anomalies linked to the FISDW cascade. F_H and F_L have been attributed to Shubnikov–de Haas orbits linked to AO- and SDW-induced compensated electron and hole tubes,[21] respectively, although this interpretation is still under debate.[22] In the metallic state at 8 kbars, only one oscillation series with frequency $F=190$ T has been observed at 0.5 K in magnetic fields lower than 19 T.[15] This latter frequency, which behaves as expected for a q-2D orbit, has been regarded as arising from F_H although F_H, as well as F_L, is thought to increase under pressure due to the increase of both the first Brillouin zone area and the warping of the FS sheets.

In the following, it is demonstrated that (i) a field-induced phase transition is observed above ~20 T in $(TMTSF)_2NO_3$, starting from the q-2D metallic state obtained under a pressure of 8.5 kbars, and (ii) the field-induced phase is characterized by a spectrum of the oscillatory magnetoresistance strongly different from that observed in the low-pressure

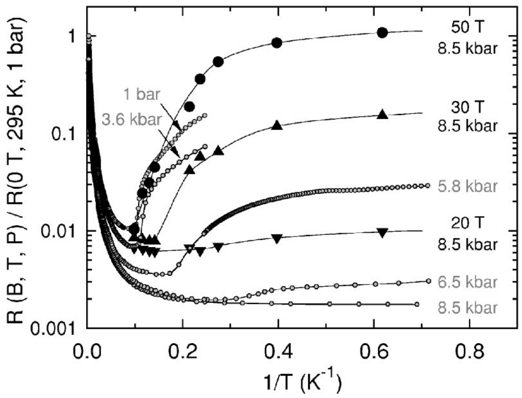

FIG. 1. Temperature dependence of the normalized resistance of $(TMTSF)_2NO_3$ at different pressures in zero-field (small symbols) and in finite magnetic field at 8.5 kbars (large symbols). Solid lines are guides to the eye.

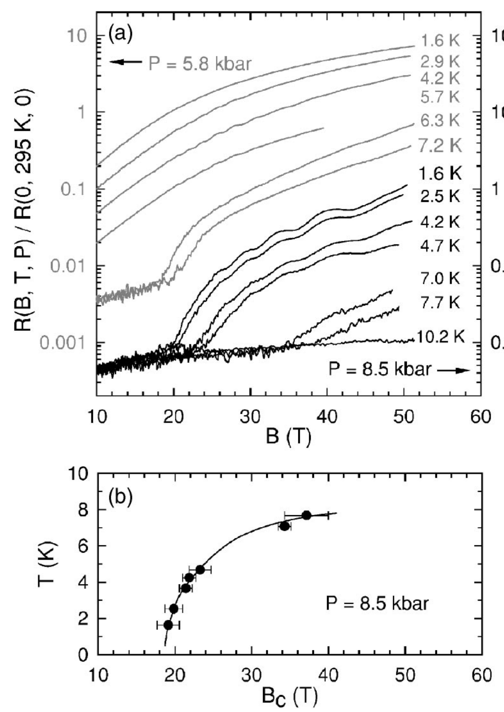

FIG. 2. (a) Magnetic-field-dependent resistance of $(TMTSF)_2NO_3$ at different temperatures for $P=5.8$ kbar (gray lines) and $P=8.5$ kbars (black lines) and (b) semimetal-FISDW phase transition deduced from the data at $P=8.5$ kbars. The solid line is a guide to the eye.

SDW state. Since the high-pressure oscillatory spectrum and background magnetoresistance are very similar to that of $(TMTSF)_2PF_6$ under high pressure, it is inferred that this phase transition is a FISDW.

Magnetoresistance experiments were performed in pulsed magnetic field up to 50 T (pulse decay duration of 0.18 s). Electrical contacts were made to the crystal using annealed gold wires of 10 μm in diameter glued with graphite paste. Alternating current (2 μA, 20 kHz) was injected parallel to the c^* direction (interlayer configuration). A lock-in amplifier with a time constant of 100 μs was used to detect the signal across the potential leads. As reported in Ref. 23, and contrary to other Bechgaard salts, the largest faces of most of the crystals are not perpendicular to the c^* direction. For this reason, the ambient pressure transverse magnetoresistance was first measured with the current injected along the most conductive direction (*a* axis) at a temperature of 4.2 K, using a rotating sample holder in order to determine the direction of c^*. Subsequent measurements of the interlayer magnetoresistance were performed in the temperature range from 1.6 K to 11 K, under pressures up to 8.5 kbars in an anvil cell.[24] Prior to these latter experiments, the pressure dependence of the interlayer resistance was determined in a pressure clamp at room temperature $[d(\ln(R))/dP=-0.11 \text{ kbar}^{-1}]$ with a manganin piezoresistive sensor. This parameter was used to determine the pressures achieved in the anvil cell. As reported in Ref. 24, it is expected that pressure variation during cooling is very small, owing to the cell geometry.

The temperature dependence of the normalized interlayer resistance is displayed in Fig. 1. As previously observed for in-plane measurements,[25] the zero-field resistance at ambient pressure exhibits a steep rise below 9.5 K related to the onset of the SDW transition. In the lower temperature range, the resistance tends to saturate due to the imperfect nesting. As the pressure increases, the resistance rise is less and less pronounced and shifts towards low temperatures. Finally, at 8.5 kbars, the resistance displays a metallic behavior down to the lowest temperature explored. The AO transition is preserved under pressure, in agreement with findings of Ref. 13. Indeed, T_{AO} deduced from these data increases from 43 K at ambient pressure up to 50 K at 8.5 kbars. The absence of superconductivity has been confirmed in a different run performed in a dilution refrigerator down to 64 mK.

Magnetoresistance data collected at different temperatures are displayed in Fig. 2(a). For $P=8.5$ kbars, a sudden resistance rise is observed at temperatures below 10 K at a threshold field B_c that increases as the temperature increases [see Fig. 2(b)]. This behavior is typical of the resistance rise due to FISDW transition to the $N=0$ state as it is observed in $(TMTSF)_2PF_6^2$ and $(TMTSF)_2ClO_4$.[26,27] In that respect, it should be noticed that no FISDW cascade can be detected in the temperature range explored. The temperature dependence of the resistance at 8.5 kbars and in magnetic fields above 20 T (see solid symbols in Fig. 1) is also consistent with a FISDW at low temperature. Similar behavior of the background magnetoresistance is observed at 5.8 kbars but at 6.3 K and 7.2 K only, i.e., at temperatures sufficiently above the zero-field SDW transition ($T_c=5.3$ K). Nevertheless, as reported hereafter, the spectrum of the oscillatory magnetoresistance is strongly different from the high-pressure state.

Fourier analysis of the magnetoresistance data at various pressures is displayed in Fig. 3. As expected, F_H increases as the pressure increases. Namely, $d(\ln(F_H))/dP\simeq 0.05 \text{ kbar}^{-1}$, which is very close to the data for the fast oscillations in the SDW state of $(TMTSF)_2PF_6$ (Ref. 28) (see the inset of Fig. 4). F_L also increases with pressure although with a lower rate $[d(\ln(F_L))/dP\simeq 0.02 \text{ kbar}^{-1}$, see Fig. 4]. At $P=8.5$ kbars, for which a metallic ground state is achieved, a drastically dif-

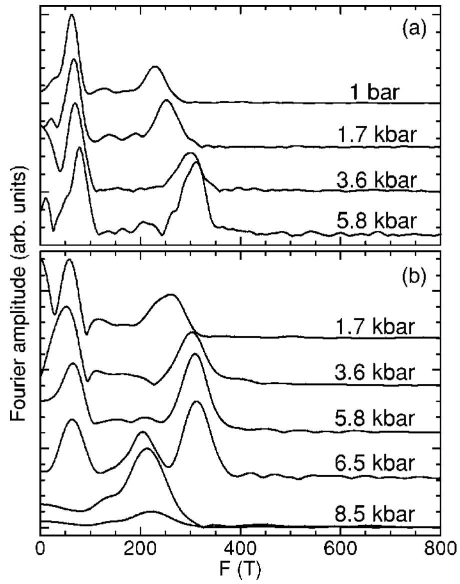

FIG. 3. Fourier analysis of the oscillatory magnetoresistance at different pressures. The black and gray lines correspond to data at 4.2 K and 1.6 K, respectively. The magnetic-field range is 10–36 T in (a). In (b), the magnetic-field range is 15–50 T for pressures up to 6.5 kbar. For $P=8.5$ kbar, the magnetic-field range is 22–50 T (since no oscillation can be detected below 22 T).

ferent behavior is observed since only one frequency $F_c=(214\pm 5)$ T is observed in the Fourier spectrum above B_c. This feature is a strong indication that a phase different from the low-pressure SDW phase is induced above B_c.

It should be noted that the observed pressure dependence of the oscillatory spectrum is at variance with the behavior of $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ for which only one frequency is observed whatever the pressure is. In the former salt, the oscillation frequency increases monotonously as the pressure increases so that no significant disruption between SDW and FISDW states is observed (see the inset of Fig. 4). In the latter salt, depending on the considered FISDW subphase, either one or two out-of-phase series with the same frequency are observed in the FISDW state[26,27] while only one series is observed in the metallic state.[29] In short, whereas the oscillatory spectrum of $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ does not evidence abrupt changes in the different (metallic, SDW, or FISDW) phases, a drastic pressure-induced modification of the spectrum is observed in $(TMTSF)_2NO_3$. Besides, the oscillation amplitude (A) at 8.5 kbars continuously increases as the temperature decreases and follow the LK behavior. This is at variance with the behavior observed in the FISDW state of $(TMTSF)_2ClO_4$ (Refs. 26, 27, 29, and 30) and in the SDW state of $(TMTSF)_2NO_3$ for which A goes to a maximum at a temperature of a few kelvins. The present study does not display any oscillation in the metallic state down to 1.6 K

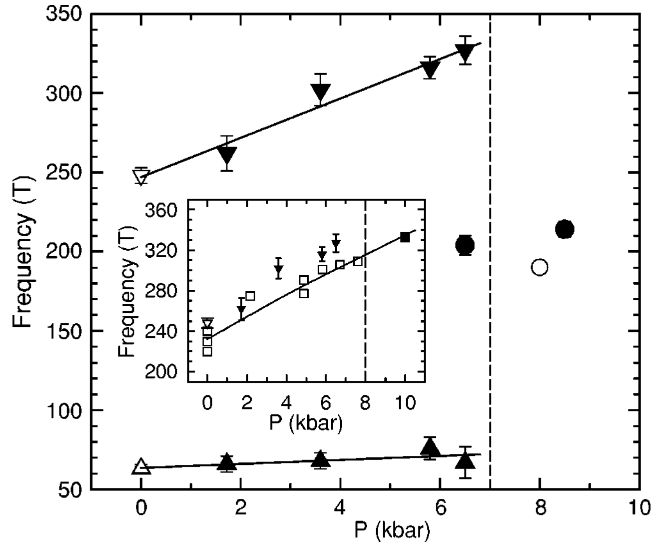

FIG. 4. Pressure dependence of the frequencies F_H (down triangles), F_L (up triangles), and F_c (circles) discussed in the text. Solid symbols are deduced from Fig. 3. Open triangles and circle are from Refs. 19 (ambient pressure SDW state) and 15 (metallic state), respectively. The inset compares the pressure dependence of the frequency F_H to the pressure dependence of the fast oscillations in $(TMTSF)_2PF_6$. Open squares (SDW state) and solid square (FISDW state) are from Refs. 28 and 2, respectively. The dotted lines mark the transition between SDW and metallic (in the main frame) or superconducting (in the inset) ground states. Solid lines are guides to the eye.

and below B_c. Nevertheless, it can be remarked that F_c is close to the frequency ($F=190$ T) that has been reported in the metallic state below 20 T, at a pressure of 8 kbars but at a lower temperature ($T=0.5$ K).[15] Finally, it is interesting to note that, at the pressure of 6.5 kbars, F_L, F_H, and F_c are observed simultaneously in the oscillatory spectrum (see Fig. 3). This may be the signature of some precursor effects, rather than phase mixing induced by some pressure inhomogeneity.

In conclusion, at variance with the idea that a FISDW state can only be observed in q-1D superconductors, we have evidenced a FISDW state in a q-2D semimetal. As demonstrated by the measured pressure dependence of the oscillatory spectrum, the (presumably $N=0$) FISDW state is strongly different from the ambient pressure SDW state. A new model, explaining FISDW in q-2D metals is clearly needed.

We are indebted to Hélène Bouchiat, Sophie Guéron, and Richard Deblock for having made available their dilution refrigerator and for their assistance. We wish to acknowledge Geert Rikken and Denis Jérome for stimulating discussions.

*Email address: audouard@lncmp.org

[1] *Low Dimensional Conductors and Superconductors*, Vol. 155 of NATO Advanced Studies Institute, Series B: Physics, edited by D. Jérome and L. G. Caron (Plenum, New York, 1987); T. Ishiguro, K. Yamaji, and G. Saito, *Organic Superconductors*, 2nd ed. (Springer Verlag, Heidelberg, 1998).
[2] N. Matsunaga, K. Yamashita, T. Oota, K. Nomura, T. Sasaki, T. Hanajiri, J. Yamada, and S. Nakatsuji, Physica B **329-333**, 1154 (2003).
[3] K. Sengupta and N. Dupuis, Phys. Rev. B **68**, 094 431 (2003).
[4] L. P. Gorkov and A. G. Lebed, J. Phys. (France) Lett. **45**, L-433 (1984); M. Héritier, G. Montambaux, and P. Lederer, *ibid.* **45**, L-943 (1984).
[5] V. M. Yakovenko, Zh. Eksp. Teor. Fiz. **93**, 627 (1987) [Sov. Phys. JETP **66**, 355 (1987)]; V. M. Yakovenko, Fizika (Zagreb) **21**, 44 (1989).
[6] N. Biškup, J. S. Brooks, R. Kato, and K. Oshima, Phys. Rev. B **60**, R15 005 (1999).
[7] J. S. Brooks, D. Graf, E. S. Choi, L. Balicas, K. Storr, C. H. Mielke, and G. C. Papavassiliou, Phys. Rev. B **67**, 153 104 (2003).
[8] J. P. Pouget, R. Moret, R. Comes, and K. Bechgaard, J. Phys. (France) Lett. **42**, 543 (1981).
[9] Y. Barrans, J. Gaultier, S. Brachetti, P. Guionneau, D. Chasseau, and J. M. Fabre, Synth. Met. **103**, 2042 (1999).
[10] A. Audouard and S. Askénazy, Phys. Rev. B **52**, R700 (1995).
[11] P. M. Grant, Phys. Rev. Lett. **50**, 1005 (1983); S. Hébrard-Brachetti, Ph.D. thesis, Bordeaux, 1996.
[12] L. P. Le, A. Keren, G. M. Luke, B. J. Sternlieb, W. D. Wu, Y. J. Uemura, J. H. Brewer, T. M. Riseman, R. V. Upasani, L. Y. Chiang, W. Kang, P. M. Chaikin, T. Csiba, and G. Gruner, Phys. Rev. B **48**, 7284 (1993).
[13] W. Kang, S. T. Hannahs, L. Y. Chiang, R. Upasani, and P. M. Chaikin, Phys. Rev. Lett. **65**, 2812 (1990).
[14] H. I. Ha, W. Kang, and J. M. Fabre, Synth. Met. **103**, 2117 (1999).
[15] W. Kang, K. Behnia, D. Jérome, L. Balicas, E. Canadell, M. Ribault, and J. M. Fabre, Europhys. Lett. **29**, 635 (1995).
[16] D. Jérome, C. R. Acad. Sci., Ser. II: Mec., Phys., Chim., Sci. Terre Univers **317**, 569 (1993).
[17] J. F. Kwak, J. E. Schirber, R. L. Greene, and E. M. Engler, Phys. Rev. Lett. **46**, 1296 (1981).
[18] J. R. Cooper, W. Kang, P. Auban, G. Montambaux, D. Jérome, and K. Bechgaard, Phys. Rev. Lett. **63**, 1984 (1989).
[19] A. Audouard, F. Goze, S. Dubois, J. P. Ulmet, L. Brossard, S. Askénazy, S. Tomić, and J. M. Fabre, Europhys. Lett. **25**, 363 (1994); A. Audouard, F. Goze, J. P. Ulmet, L. Brossard, S. Askénazy, and J. M. Fabre, Phys. Rev. B **50**, 12 726 (1994).
[20] N. Biškup, L. Balicas, S. Tomić, D. Jérome, and J. M. Fabre, Phys. Rev. B **50**, 12 721 (1994).
[21] K. Kishigi and K. Machida, Phys. Rev. B **53**, 5461 (1996); J. Phys.: Condens. Matter **9**, 2211 (1997).
[22] D. Vignolles, J. P. Ulmet, A. Audouard, M. Naughton, and J. M. Fabre, Phys. Rev. B **61**, 8913 (2000).
[23] N. Biškup, M. Basletić, S. Tomić, B. Korin-Hamzić, K.Maki, K. Bechgaard, and J. M. Fabre, Phys. Rev. B **47**, 8289 (1993).
[24] M. Nardone, A. Audouard, D. Vignolles, and L. Brossard, Cryogenics **41**, 175 (2001).
[25] S. Tomić, J. R. Cooper, D. Jérome, and K. Bechgaard, Phys. Rev. Lett. **62**, 462 (1989); S. Tomic, J. R. Cooper, W. Kang, D. Jérome, and K. Maki, J. Phys. I **1**, 1603 (1991).
[26] Ok-Hee Chung, W. Kang, D. L. Kim, and C. H. Choi, Phys. Rev. B **61**, 11 649 (2000).
[27] S. Uji, C. Terakura, T. Terashima, J. S. Brooks, S. Takasaki, S. Maki, S. Nakatsuji, and J. Yamada, Curr. Appl. Phys. **1**, 77 (2001).
[28] N. Matsunaga, K. Yamashita, H. Kotani, K. Nomura, T. Sasaki, T. Hanajiri, J. Yamada, S. Nakatsuji and H. Anzai, Phys. Status Solidi B **223**, 545 (2001).
[29] S. Uji, J. S. Brooks, S. Takasaki, J. Yamada, and H. Anzai, Solid State Commun. **103**, 387 (1997).
[30] J. S. Qualls, C. H. Mielke, J. S. Brooks, L. K. Montgomery, D. G Rickel, N. Harrison, and S. Y. Han, Phys. Rev. B **62**, 12 680 (2000).