

Quantum oscillations in quasi-one-dimensional metals with spin-density-wave ground states

J. S. Brooks*

Department of Physics, Florida State University, Tallahassee, Florida 32310

J. O'Brien, R. P. Starrett, R. G. Clark, and R. H. McKenzie

National Pulsed Magnet Laboratory, School of Physics, University of New South Wales, Sydney 2052, Australia

S.-Y. Han and J. S. Qualls

Department of Physics, Florida State University, Tallahassee, Florida 32310

S. Takasaki, J. Yamada, and H. Anzai

Himeji Institute of Technology, Akahogun, Hyogo 678-12, Japan

C. H. Mielke

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

L. K. Montgomery

Chemistry Department, Indiana University, Bloomington, Indiana 47405

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We consider the magnetoresistance oscillation phenomena in the Bechgaard salts $(\text{TMTSF})_2X$, where $X = \text{ClO}_4$, PF_6 , and AsF_6 in pulsed magnetic fields to 51 T. Of particular importance is the observation of a new magnetoresistance oscillation for $X = \text{ClO}_4$ in its quenched state. In the absence of any Fermi-surface reconstruction due to anion order at low temperatures, all three materials exhibit nonmonotonic temperature dependence of the oscillation amplitude in the spin-density-wave (SDW) state. We discuss a model where, below a characteristic temperature T^* within the SDW state, a magnetic breakdown gap opens.

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A subject of continued interest and controversy is the observation of oscillations in the magnetoresistance of quasi-one-dimensional organic conductors, specifically the Bechgaard salts.¹ The nature of the oscillations is shown in Figs. 1–4. Here, riding on a sometimes very large magnetoresistance are oscillations commonly called rapid oscillations or ‘RO.’ At the center of the problem is the expectation, from tight-binding band-structure calculations,² that the metallic Fermi surface of the Bechgaard salts $(\text{TMTSF})_2X$ consists of two warped, open-orbit sheets in the first Brillouin zone, as is shown in Fig. 5(b). Clearly in such a topology there are no extremal-closed orbits to allow conventional oscillations in the magnetoresistance, i.e., the Shubnikov–de Haas (SdH) effect. Nevertheless, a preponderance of evidence suggests that these materials exhibit SdH-like oscillations in applied magnetic fields below their respective spin-density-wave (SDW) transitions, and that these oscillations are periodic in inverse field. Furthermore, angular-dependent studies³ indicate a quasi-two-dimensional character of the observed SdH frequency. The two questions that naturally arise in considering these oscillations are: how do closed orbits arise from open sheets; and if closed orbits do exist, why are their amplitudes nonmonotonic with decreasing temperature? In most cases, below the SDW ordering temperature T_{SDW} (≈ 5 – 12 K), the RO amplitude peaks at T^* (≈ 2 – 5 K), then rapidly diminishes.⁴

We argue in this paper that the ‘RO’ are a result of a semiclassical treatment of the reconstructed and/or nested

quasi-one-dimensional Fermi-surface effects. For high-symmetry anions such as $X = \text{AsF}_6$ or PF_6 , there is no anion ordering, but below T_{SDW} the Fermi surfaces are nested with wave vector \mathbf{Q} . Following the arguments of Uji *et al.*^{5,6} (for $X = \text{PF}_6$ and ClO_4) and Kishigi and Machida⁷ (for $X = \text{NO}_3$ and ClO_4), the new periodicity introduces a new Brillouin zone and creates a magnetic breakdown network of electron and hole pockets [see Fig. 5(b)]. For low-symmetry anions such as $X = \text{ClO}_4$, and NO_3 , anion ordering (at T_{AO}) takes place, and the Fermi surface reconstructs. Here, further nesting may result, either induced by lower temperatures (NO_3) or by application of high-magnetic fields (ClO_4). Small changes in the nesting condition may change the size of the small pockets and density of occupied states, but the area associated with the larger magnetic breakdown paths remains essentially constant, as depicted in Fig. 5(b).

The nature of the SDW state has been most extensively studied for $X = \text{PF}_6$. These include proton⁸ and ⁷⁷Se NMR studies,⁹ muon spin studies,¹⁰ and theoretical treatments by Yamaji,¹¹ Montambaux,¹² and Bjelis and Maki.¹³ We take the view, based on the Korringa-like behavior of ⁷⁷Se NMR spin relaxation, the characteristic increase in T_{SDW} with increasing field¹⁴ and the existence of SdH-like oscillations, that the nesting is incomplete below T_{SDW} . Furthermore, at T^*1/T_1 in ⁷⁷Se NMR takes on an activated form⁹ and the magnetization shows a small bump.¹⁵ These experiments indicate that at T^* there is a transition to a new electronic state, but still, below T^* only oscillations associated with the large breakdown areas are observed.

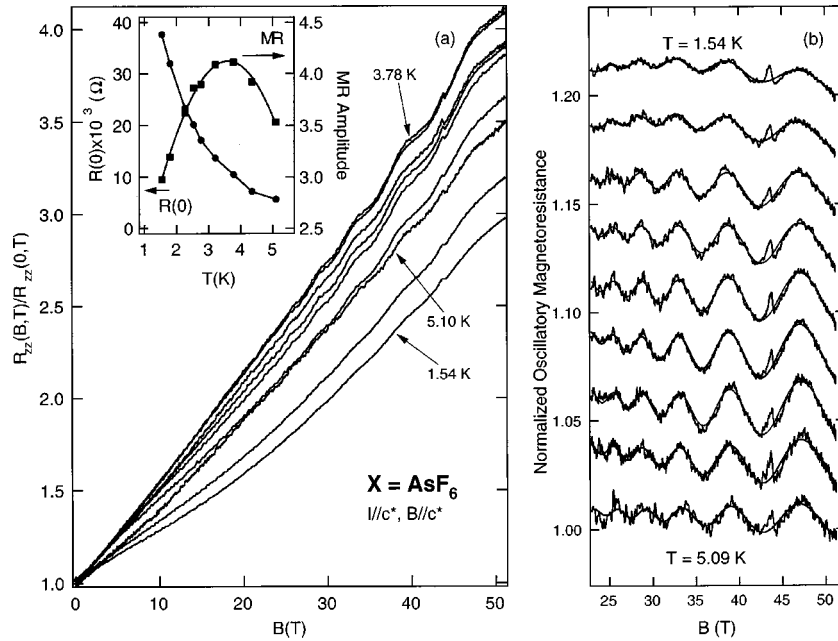


FIG. 1. $X = \text{AsF}_6$. (a) Temperature-dependent MR. Inset: zero-field resistance and MR at 50 T vs temperature. Intermediate temperatures are as shown in the inset. (b) Temperature dependence of the RO amplitudes. Solid lines-LK fits. Curves are offset.

The present study is a concurrent investigation of the c^* axis (R_{ZZ}) magnetoresistance of three of the Bechgaard salts, $(\text{TMTSF})_2X$, where $X = \text{AsF}_6$, PF_6 , and ClO_4 , in pulsed fields to 51 T at the Australian National Pulsed Magnet Laboratory and also at the US pulsed field facility at Los Alamos. Contact to samples was made with Au paint and thin ($10 \mu\text{m}$) Au wires. Transport measurements in the high-field region was via planar transmission lines—a technology developed for microwave measurements^{16,17} in explosively generated fields >500 T. The lines are $9 \mu\text{m}$ thick and $68 \mu\text{m}$ wide, and the dB/dt pickup to the signal is generally negligible. Measurements were made with B and I parallel to the c^* (lowest conductivity) axis. dc currents of $1 \mu\text{A}$ (PF_6 and AsF_6) and $50 \mu\text{A}$ (ClO_4) were used to avoid sample heating. The samples were cooled *very quickly* (~ 1 s) from liquid-nitrogen to liquid-helium temperatures. This is essential to suppress anion ordering in $X = \text{ClO}_4$ in order to stabilize the SDW (quenched) state.

Our results are summarized in Figs. 1–3 in terms of the normalized magnetoresistance $R_{ZZ}(B,T)/R_{ZZ}(0,T)$ and the oscillatory magnetoresistance $R_{\text{OSC}}(B,T)$. $R_{\text{OSC}}(B,T)$ from c^* axis measurements, not seen in some reports,⁴ is clearly observed in all materials we studied. $R_{\text{OSC}}(B,T)$ is obtained by dividing $R_{ZZ}(B,T)$ by a low-order polynomial fit to $R(B,T)$. This allows comparison with the conventional description of SdH oscillations in terms of the Lifshitz-Kosevich (LK) formulation,¹⁸ which we have used to *directly* fit the oscillatory data. The parameters effective mass m^* , Dingle temperature, and oscillation frequency were obtained from the fit. Values were generally in the range of $0.5m_0$ to $1.5m_0$ for m^* , and 2 K to 10 K for T_D . Details of this technique will be reported elsewhere.¹⁹

We now discuss our results. (a) The material $(\text{TMTSF})_2\text{AsF}_6$ (Figs. 1 and 4) exhibited a monotonically increasing zero-field resistance with lower temperatures, but

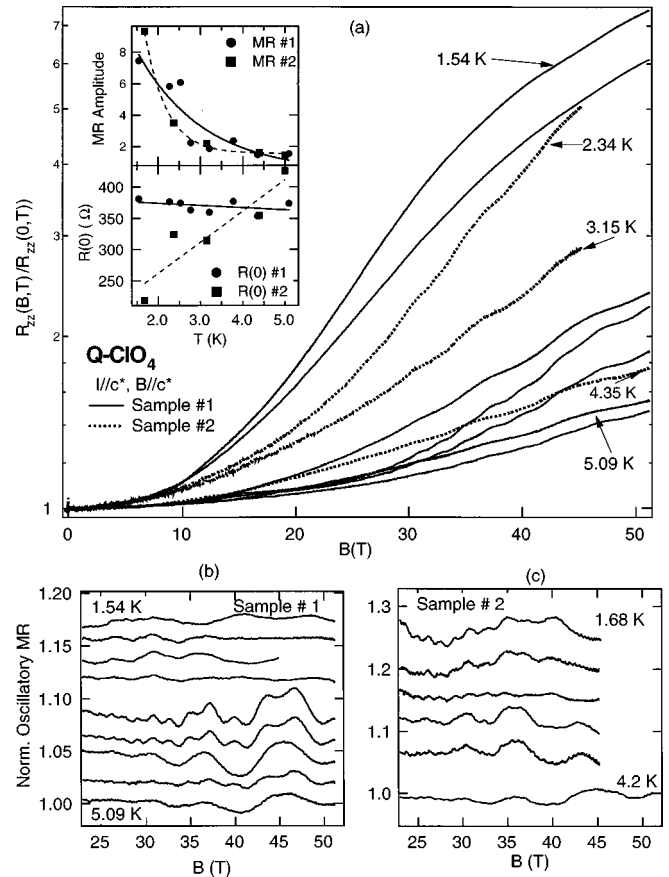


FIG. 2. $X = \text{ClO}_4$ (Q state) (two samples). (a) Temperature-dependent MR. Inset: zero resistance and high-field MR at 50 T vs temperature. (b) Temperature dependence of the RO amplitudes. Curves are offset. Sample 1: $T(\text{K}) = 5.09, 4.30, 3.78, 3.21, 2.76, 2.53, 2.26, 2.25$ K; Sample 2: $T(\text{K}) = 4.20, 3.15, 2.37, 2.08, 1.96, 1.68$ K.

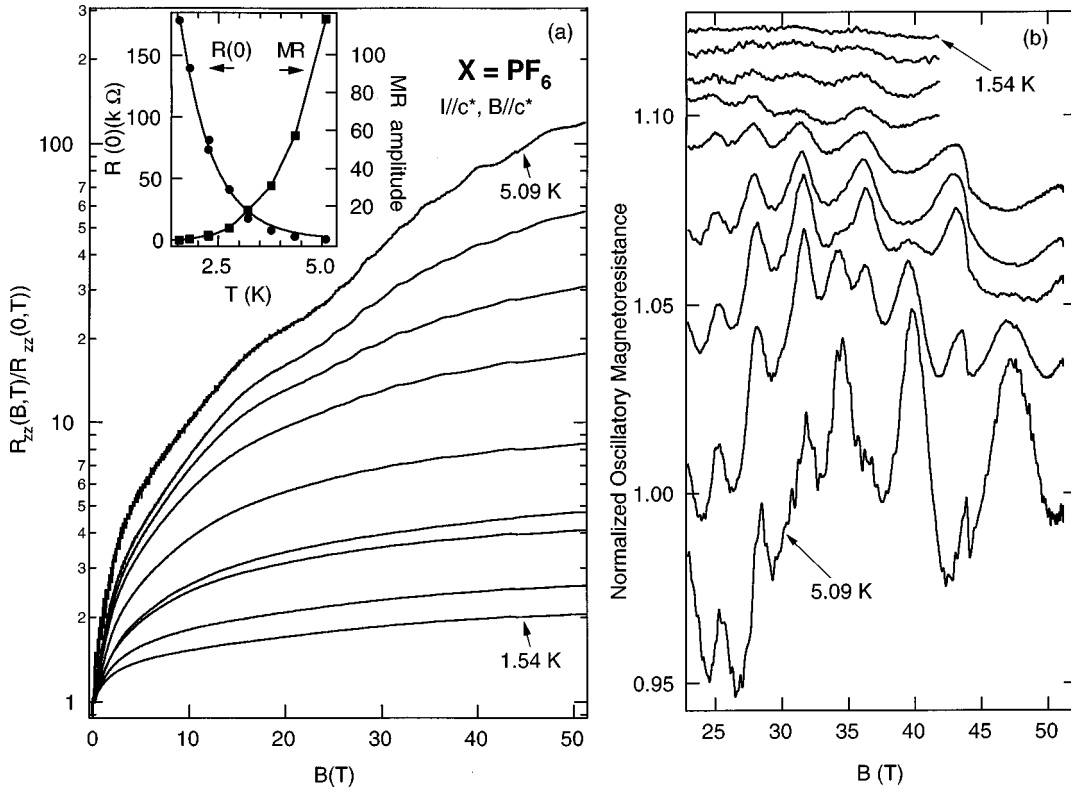


FIG. 3. $X = \text{PF}_6$. (a) Temperature-dependent MR. Inset: zero resistance and high field MR at 50 T vs temperature. Intermediate temperatures are as shown in the inset. (b) Temperature dependence of the RO amplitudes. Curves are offset.

a nonmonotonic dependence of the magnetoresistance (MR) is apparent with a maximum around 3.8 K. Quantum oscillations with a single frequency $F_1 = 222$ T are observed in R_{ZZ} , also with a nonmonotonic temperature dependence of the oscillation amplitude, in reasonable agreement with other

reports.⁴ (The small spike near 43 T, which appears in high-resistance samples is due to the pulsed-magnet crowbar.) (b) For the quenched version $Q-(\text{TMTSF})_2\text{ClO}_4$, we observed entirely new behavior (Figs. 2 and 4), which has not been previously reported in any detail. Here a number of independent measurements in this work and other preliminary^{17,20} and complementary²¹ work of this state of the material, where the anion ordering is suppressed, clearly show an orbital frequency at $Q - F_1 = 190$ T. This frequency is the result of the nesting of the original Fermi surface by the SDW wave vector $Q(\frac{1}{2}, \frac{1}{4})$ [similar to PF_6 as determined from NMR (Ref. 22)], in contrast to the $A0 - F_1 = 265$ T frequency, which appears with anion ordering for $Q(0, \frac{1}{2}, 0)$. Although the $Q - F_1$ frequency is dominant in highly quenched samples, the $A0 - F_2 = 530$ T oscillation, which is a remnant of the ordered state, appears in at least one measurement. The $Q - F_1 = 190$ T oscillation amplitude in all cases is nonmonotonic with a maximum around 3.5 K. We find also a temperature-dependent phase shifted first harmonic. $A0 - F_2$ also shows a similar dependence, in agreement with detailed studies of the ordered state.⁶ (c) The behavior for $X = \text{PF}_6$ is shown in Figs. 3 and 4. We find an oscillation frequency $F_1 = 240$ T along with a temperature-dependent phase shifted first harmonic. The zero-field resistance is activated, and the MR decreases monotonically with lower temperature. We note structure in the MR above 3 K near 30 T. The RO amplitude near 50 T for R_{zz} as shown in Fig. 4 decreases monotonically from the highest temperature. The estimated T^* is at higher temperature (5 vs 3.5 K) than that reported⁴ up to 30 T for R_{xx} . Sample heating or errors in temperature have been ruled out as the origin of this differ-

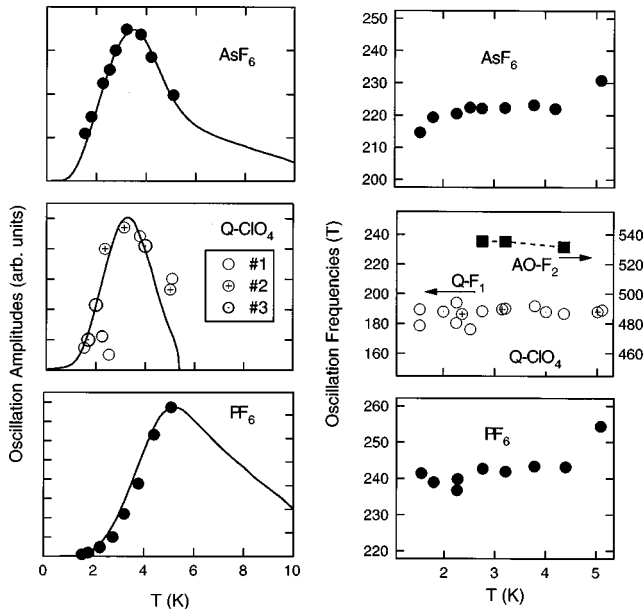


FIG. 4. Results of the modified Lifshitz-Kosevich analysis. Left panels, oscillation amplitudes for the three compounds. Right panels, oscillation frequencies. Note that T_{SDW} for ClO_4 is ~ 5 K at zero field, but may be higher at 50 T based on the finite RO amplitude at 5 K.

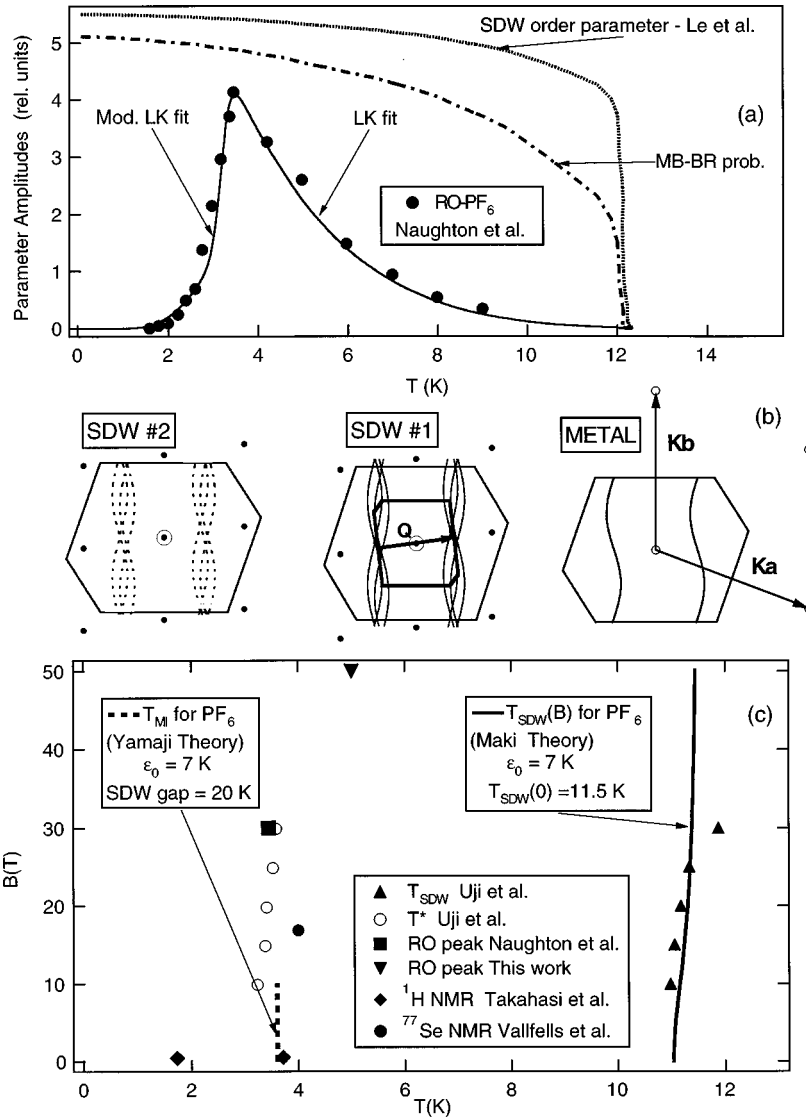


FIG. 5. (a) Parameters and test of modified-LK model. For the $X=PF_6$ data fitted, $T^*=3.2$ K with a width of 0.1 K, $T_D=10$ K, and a breakdown gap energy of 12 K. Note that the order parameter alone, and its contribution to magnetic breakdown, are nearly constant at T^* , and *cannot* influence the RO amplitude in this range. (b) Progression of Fermi-surface topology with temperature. (c) Magnetic field dependence of T_{SDW} , T_{MI} , and T^* from theory and experiment.

ence. Factors such as the high fields (50 T vs 30 T or less), and the component of the resistivity (R_{ZZ}) studied, may account for some differences between this work and previous studies.

Although all three systems share a common Fermi-surface topology, and SDW ground state, there is no clear relationship between the magnetoresistance, the zero-field resistance, and the RO amplitudes, as shown in the insets of Figs. 1–3. For $Q\text{-ClO}_4$ this may be due to residual anion order, as is evidenced by the presence of the F_2 orbit, and also by the weakly metallic (weakly insulating) nature of $R(0)$. But for $X=\text{AsF}_6$ and PF_6 , there is a difference in the temperature dependence of the MR, even though $R(0)$ rises rapidly at low temperatures. The relationship of the various transport parameters remains an open question.

Theoretical treatments of the possible transitions in the SDW ground state, as illustrated in Fig. 5(c), originate from Yamaji's treatment of the formation of the SDW state^{1,11} in terms longitudinal and transverse hopping t_a and t_b , respec-

tively. The influence of t_b is characterized by the parameter $\varepsilon_0=t_b^2/t_a \times 1.4$, which gauges the degree of imperfect nesting. Maki²³ has extended the treatment of the T_{SDW} formation to include the improvement of the nesting condition with high-magnetic fields, and Audouard and Askenazy²⁴ have successfully applied this treatment to the case of $X=\text{NO}_3$. Importantly, Yamaji's model predicts *two* transitions, T_{SDW} into a semimetallic SDW, and a second transition at lower temperature (T_{MI}) into an insulating SDW. How close T_{MI} is to T_{SDW} depends crucially on t_b . For $t_b=0.012$ eV, T_{MI} and T_{SDW} coincide [this is the case of AO-ClO_4 (Ref. 25)], but for t_b twice this value, (as preliminarily reported for non-AO states²⁵), T_{MI} lies well below T_{SDW} . Indeed, for $\varepsilon_0=7$ K, T_{MI} and T^* would coincide. However, the anomalous behavior of transport properties below T^* cannot simply be explained in terms of a completely gapped Fermi-surface (FS). We therefore consider a *modified*-LK description of the RO amplitudes. In our analysis, we follow rigorously the LK treatment of Uji *et al.*¹⁵

above T^* . However, below T^* , the amplitude seems to be exponentially dependent on temperature. This suggests that a magnetic breakdown gap opens. We consider, therefore, a simple, heuristic model closely following the behavior of the ^{77}Se NMR results, which show a rapid (activated) decrease in relaxation rate⁹ below T^* . This model simply introduces an activated prefactor to the LK formula below T^* , assuming a finite transition width of order 0.1 K. As shown in Figs. 4 and 5, this model provides an excellent description of the temperature dependence of R_{OSC} for the present and previous results.

To summarize, we have examined the anomalous temperature dependence of the rapid oscillations in the SDW state of $(\text{TMTSF})_2X$ compounds. By thermally quenching the $X=\text{ClO}_4$ compound, anion ordering is suppressed and the resulting reconstructed Fermi surface is similar in topology to that for $X=\text{PF}_6$ and AsF_6 . The observation of a rapid oscillation frequency at 190 T for $Q\text{-ClO}_4$, which is reproducible in independent experiments with samples from inde-

pendent synthesis labs, supports this interpretation. The temperature dependence of the $Q\text{-ClO}_4$ oscillation amplitude is nonmonotonic, as in other $(\text{TMTSF})_2X$ materials where an ambient SDW phase exists in the absence of pressure or magnetic field. In reference to Fig. 5(a), we find that this temperature dependence below T_{SDW} is well described by a Lifshitz-Kosevich description with magnetic breakdown and Bragg reflection points on the reconstructed Fermi surface. Below the second transition T^* , a model where a magnetic breakdown gap opens up gives an accurate account of the observed temperature dependence. We note that the connection of T^* and T_{MI} is suggestive, and further theoretical work is needed.

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*Author to whom correspondence should be addressed. Present address: NHMFL/Department of Physics, Florida State University, Tallahassee FL 32310. FAX: 1-850-644-5038. Electronic address: brooks@magnet.fsu.edu

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