The effects of high magnetic fields on the quasi-one- and two-dimensional electronic structure of organic conductors is a rich area of investigation. In Bechgaard and related salts which remain metallic at low temperatures, a magnetic field applied parallel to the least conducting direction (perpendicular to the conducting chains) produces a field-induced spin density wave (FISDW) ground state. A simple description of this effect is that the magnetic field decreases the amplitude of the lateral motion of the carriers as they move along the conducting chains, thereby making the electronic structure increasingly more one-dimensional. Hence eventually 1D instabilities become favorable. In reference to Fig. 1, a nested quasi-1D Fermi surface is induced at a second-order phase boundary where a FISDW gap opens above a threshold field $B_{\text{th}}$. Due to quantization of the orbital nature of the FISDW subphases, in the quantum limit, the optimum nesting vector (where $N=0$) yields the final FISDW state in the case of $(\text{TMTSF})_2\text{ClO}_4$ which has a single quasi-one-dimensional Fermi surface ($Q1D$ FS).

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PACS numbers: 74.70.Kn, 75.30.Gw, 76.60.-k

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has been described in the Bechgaard salts by NMR studies. The characteristic NMR lineshape in the SDW phase involves multiple peaks due to the antiferromagnetic nature of the local fields, and the peak separations vary systematically with field orientation due to changes in the dipolar coupling with respect to the (TMTSF)$_2$ClO$_4$ donor axes.

NMR pulse optimization involved $\pi/2 - \pi/2$ pulse trains to obtain the maximum NMR intensity at the optimum pulse widths $\tau_M$ in the metallic and $\tau_{sdw}$ in FISDW phases. Typically, in the metallic state the optimum $\pi/2$ pulse width was $\tau_M = 1 \mu s$ while in the FISDW region $\tau_{sdw}$ varied from 50 ns to 500 ns. Since the same pulse power level (12 W) was used for all measurements, the rf enhancement factor was obtained from the relation $\eta \approx \tau_M/\tau_{sdw}$. The spin-lattice relaxation rate $1/T_1$ was defined using a single-exponential form for the magnetization recovery, and varied by less than 10% over the spectrum in all cases. In the FISDW, there is a slight deviation (approx 5% in the initial slope) from the exponential recovery, not uncommon for these systems.

Long recycle times (0.5 to 5 s) were used to avoid sample heating.

We first discuss results at constant angle where the field (and frequency) were changed to access the different FISDW phases. In Fig. 2(a) the magnetic field dependent $1/T_1$ and the corresponding $c^*$-axis resistance ($R_{zz}$) are shown for 2 K. For constant temperature, as the metal-FISDW transition approached and $B_{sdw}$ was crossed, $1/T_1$ gradually increases. It is not until the predominant first-order sub-phase transition $B_1$ is reached that $1/T_1$ exhibits a maximum. For further increases in field $1/T_1$ decreases. The metallic pulses were optimum when $B < B_1$, but at higher fields the shorter $\tau_{sdw}$ pulses were necessary to follow the signal into the FISDW phase. In Fig. 2(b) and 2(c), the variation of $1/T_1$ with temperature is shown, along with $R_{zz}$ and the Boltzmann factor normalized NMR intensity. The temperature dependence of $1/T_1$ generally follows a critical fluctuation behavior $1/T_1 \approx (T - T_{SDW})^{-1/2}$ above the peak, and a power law behavior ($1/T_1 \approx T^{1.2}$) at lower temperatures. The spin density is constant in the metallic state, and drops exponentially starting at the second-order phase boundary. Simultaneous resistance measurements show the onset of semimetallic behavior at this same temperature. We emphasize here that the peaks in $1/T_1$ occur at temperatures that are systematically below the second-order phase boundary, as indicated by the resistance anomaly and the signal intensity. The positions of the peaks in $1/T_1$, and the corresponding phase boundaries from transport measurements, are presented in Fig. 1 for all field-dependent data.

The angular-dependent data shown in Fig. 3 at 14 T (118.32 MHz) and 1.5 K provide more detail about the second-order $B_{th}$ and first-order $B_1$ FISDW transitions. In the angle range $70^\circ < \theta < 110^\circ$, the sample is metallic, which is also shown in magnetoresistance (MR) and $\eta = 1$. There is a slight increase (10 s$^{-1}$) in $1/T_1$ as the field rotates away from the b'-axis in the metallic state. When $B_1$ reaches $B_{th} = 6.25T$, there is an increase in $1/T_1$ corresponding to a sharp feature in the resistance. Note however that for $B_{th} < B_1 < B_1^\perp$, $\eta = 1$ as in the metallic phase. As in Fig. 2a, the peak in $1/T_1$ occurs at $B_1$ where $\eta \approx 20$. Deep in the FISDW phase, the NMR lineshapes broadened and have a double-peaked struc-
ture. Since the a-axis used for the rotation data is also the symmetry axis for the hyperfine coupling, anisotropy in $1/T_1$ is indiscernible in the metallic phase, and negligible in the FISDW phase (a few percent) compared with the large variation in $1/T_1$ at the different phase boundaries. In the angular dependence there is a special symmetry position at $\theta=25^0$ where the magnetic field is parallel to the x-axis of the TMTSF donor molecule. Here $1/T_1$ exhibits a dip (marked X in Fig. 3) and the lineshape narrows to a single peak. This dip feature has been previously reported for (TMTSF)$_2$PF$_6$ and is not related to the magic angle (MA) effects reported in (TMTSF)$_2$ClO$_4$. Although FISDW data is always above the spin-flip field (<0.5 T), there is a spin rotation from the a-axis to the c*-axis when the field direction approaches B||c*, which has been associated with the dip phenomena. Likewise, there is no evidence for the MA effects in the NMR signal, in accord with previous studies on (TMTSF)$_2$PF$_6$.

In Fig. 4 the high field ($B_{max}=30$ T) angular-dependent NMR results for $1/T_1$, $B_1$, $B^*$, $B_{rec}$, and $\eta$ are shown along with the corresponding MR data at T=1.47 K and 24.93 MHz. As in Fig. 3 rotation away from the metallic phase at $B\parallel b^*$ (i.e. for increasing $B_{1\parallel}$) causes $1/T_1$ to increase as $B_{th}$ is entered, but it is not until the first-order boundary $B_1$ is crossed that $1/T_1$ reaches a maximum. Since $B_{max}=30$ T, the full field range in $B_1$ can be accessed, and additional peaks in $1/T_1$ are observed for $B_{1\parallel}=B^*$ in the range 15 to 17 T (which corresponds to a characteristic feature in the MR seen in many experiments), and also for $B_{1\perp}=B_{rec}$. Of specific note is the corresponding behavior of $\eta$ which falls from 5 to 1 when the $B_{rec}$ boundary is crossed for both positive and negative field directions. The main changes in spectral linewidth due to the internal field occur at the metal-FISDW transition, and not at the subphase transitions. Hence the enhancement factor and increase in $1/T_1$, and not a significant change in the internal field, characterizes the $B_{rec}$ phase boundary. For $\theta \approx 25^0$ in Fig. 4(a) the feature corresponding to $B_{1\parallel}=B_{rec}$ is obscured, most likely because it occurs where the dip (see Fig. 2) in $1/T_1$ appears. The results from Fig. 4 are summarized in Fig. 1.

Most significant in the present work is the location of the peaks in $1/T_1$ in the different FISDW phases, and the behavior of the NMR signal above the re-entrant phase boundary.

First, for $1/T_1$ data at constant field, the peaks in $1/T_1$ appear at temperatures as much as 30% lower than the second order phase boundary (Fig.1). Hanson et al have previously suggested that the true signature of the second order onset of the FISDW occurs at the $1/T_1$ peak, and not at the higher temperature onset of transport anomalies. However, the rapid oscillation (RO) behavior (see, e.g., Fig.4(c)) is very sensitive to the Fermi surface nesting conditions, and a transition between Stark interference behavior (metallic) and anomalous RO behavior (FISDW) is coincident with the second order phase boundary. The spin density, and the onset of semimetallic behavior in $R_{zz}$ also begins to change at this boundary (Fig. 2c); see also Fig.6(a) in Ref.[9]). We therefore assert that the FISDW phase boundary, i.e. the second order line, appears when the FS nesting (and gap) first appears, that is, above the temperature where the peaks in $1/T_1$ appear. From Fig. 2(b,c), this effect occurs approximately at the inflection point of the drop in the spin density (50%). In contrast, we note that in (TMTSF)$_2$PF$_6$ where under ambient conditions a SDW transition occurs at 12 K, the resistance shows a sharp increase which is nearly coincident with a peak in $1/T_1$ with an uncertainty of less than 10%. Hence there appears to be a difference in the way the order parameter develops below the SDW and the FISDW phase boundaries. Critical slowing of the fluctuations near a SDW transition ordinarily leads to a peak in relaxation rate at $T_c$. Below $T_c$, relaxation (dynamical) effects have been observed, which cannot be ruled out. However, a density of states effect similar to the Hebel-Slichter peak seen in superconductors is also possible.

Second, in both field dependent and rotation experiments, we find that $1/T_1$ exhibits peaks within the second-order phase boundary, in particular at the first-order phase lines $B_1$, $B^*$, and $B_{rec}$. Since the first order transitions involve transitions between the different sub-phases, domain wall effects may cause the peaks in $1/T_1$ as these boundaries are crossed.
Third, in the rotation experiments the enhancement parameter $\eta$ was monitored in detail. The effect is associated with rf-induced displacement or de-pinning of the SDW phase by the electric fields. There is no enhancement ($\eta=1$) in the metallic phase, and in the FISDW phase $\eta$ will increase according to the ability of the electric field to modulate the condensate. The electric fields associated with de-pinning are typically 5 mV/cm or less. In the present case, we estimate the ac electric field in the NMR coil to be of order 1 to 10 V/cm (see also previous estimates), well above the de-pinning field. Notably, at the transition $B_{rc}$, the enhancement factor drops to unity, but the NMR spectrum is still double-peaked and characteristic of antiferromagnetic structure.

In summary, we have correlated the transport features which describe the FISDW phase diagram of (TMTSF)$_2$ClO$_4$ with $^{77}$Se NMR. We find that the peaks in $1/T_1$ occur within (not at) the second order phase boundary. Furthermore angular dependent measurements facilitate the crossing of FISDW subphase boundaries at constant temperature where peaks in $1/T_1$ are also observed, indicating changes in the nesting configurations at these transitions. At high fields, the drop in the rf enhancement parameter upon crossing the re-entrant phase boundary is consistent with the expectation that only one FS sheet is nested above $B_{rc}$. We expect that this electronic configuration (only one FS sheet may be nested) is also the case in the broader region between the second order phase boundary and the underlying first order phases at lower temperatures.

We thank V.F. Mitrovic for helpful suggestions. This work was supported in part by NSF DMR-0602859 (JSB) and DMR-0520552 (SEB), and performed at the National High Magnetic Field Laboratory, supported by NSF DMR-0084173, by the State of Florida, and the DOE.

\begin{thebibliography}{99}
    \bibitem{T. Ishiguro, K. Yamaji, and G. Saito} Organic Superconductors II (Springer-Verlag, New York, 1998).
    \bibitem{K. Oshima, H. Okuno, K. Kato, R. Maruyama, R. Kato, A. Kobayashi, and H. Kobayashi} Synth. Met. 70, 861 (1995).
    \bibitem{N. Biskup, J. S. Brooks, R. Kato, and K. Oshima} Phys. Rev. B 60, R15005 (1999).
    \bibitem{P. M. Chaikin} J. Phys. I (France) 6, 1875, (1996).
    \bibitem{F. Pesty, F. Garoche, and K. Bechgaard} Phys. Rev. Lett. 55, 2495 (1985).
    \bibitem{W. Kang, S. T. Hannahs, and P. M. Chaikin} Phys. Rev. Lett. 70, 3091 (1993).
    \bibitem{T. Osada, N. Miura, and G. Saito} Solid State Commun. 60, 441 (1986).
    \bibitem{M. J. Naughton, R. V. Chamberlin, X. Yan, S.-Y. Hsu, L. Y. Chiang, M. Ya. Azbel, and P. M. Chaikin} Phys. Rev. Lett. 61, 621 (1988). As first described in this reference, we refer to the low temperature phase boundary near 26 T as “re-entrant”, although only one FS sheet returns to a metallic state.
    \bibitem{O.-H. Chung, W. Kang, D. L. Kim, and C. H. Choi} Phys. Rev. B 61, 11649 (2000).
    \bibitem{S. Uji, J. S. Brooks, M. Chaparala, S. Takasaki, J. Yamada, and H. Anzai} Phys. Rev. B 55, 14387 (1997).
    \bibitem{S. K. McKernan, S. T. Hannahs, U. M. Scheven, G. M. Danner, and P. M. Chaikin} Phys. Rev. Lett. 75, 1630 (1995).
    \bibitem{S. K. McKernan, S. Uji, J. S. Brooks, and P. M. Chaikin} Solid State Commun. 145, 385 (2008).
    \bibitem{S. Uji, S. Yasuoka, T. Konoike, K. Enomoto, J. Yamada, E. S. Choi, D. Graf, and J. S. Brooks} Phys. Rev. Lett. 94, 077206 (2005).
    \bibitem{G. S. Boebinger, G. Montambaux, M. L. Kaplan, R. C. Haddon, and V. Chichester} Phys. Rev. Lett. 64, 591 (1990).
    \bibitem{J. M. Delrieu, M. Roger, Z. Toffano, A. Moradpour, and K. Bechgaard} J. Phys. (France) 47, 839 (1986).
    \bibitem{T. Takahashi, Y. Maniva, H. Kawamura, and G. Saito} J. Phys. Soc. Jpn 55, 1364 (1986).
    \bibitem{M. Takigawa and G. Saito} J. Phys. Soc. Jpn. 55, 1233 (1986).
    \bibitem{F. Zhang, Y. Kuwabuki, J. Shimagawa, B. Alavi, and S. E. Brown} Phys. Rev. B 72, 060501(R) (2005).
    \bibitem{M. Takigawa and G. Saito} Physica B 143, 422 (1986).
    \bibitem{A. G. Leebed and P. Bak} Phys. Rev. Lett. 63, 1315 (1989).
    \bibitem{M. J. Naughton, O. H. Chung, M. Chaparala, X. Bu, and P. Coppens} Phys. Rev. Lett. 67, 3712 (1991).
    \bibitem{W. Wu, P. M. Chaikin, W. Kang, J. Shimagawa, W. Yu, and S.E. Brown} Phys. Rev. Lett. 94, 097004 (2005).
    \bibitem{M. E. Hanson, M. Horvatic, C. Berthier, Y. Fagot-Revurat, D. Jerame, and C. Bourbonnais} Bull. Am. Phys. Soc. 43, 130.12 (1998).
    \bibitem{S. Uji, J. S. Brooks, M. Chaparala, S. Takasaki, J. Yamada, and H. Anzai} Phys. Rev. B 55, 12446 (1997).
    \bibitem{K. Bechgaard, C. S. Jacobson, K. Mortensen, H. J. Pedersen, and N. Thorup} Solid State Commun. 33, 1119 (1980).
    \bibitem{S. Valfells, P. Kuhns, A. Kleinhammes, J. S. Brooks, W. Moulton, S. Takasaki, J. Yamada, and H. Anzai} Phys. Rev. B 56, 2585 (1997).
    \bibitem{W. G. Clark, M. E. Hanson, W. H. Wong and B. Alavi} J. Phys. IV (France) 3, C2-235 (1993).
    \bibitem{D. E. MacLaughlin} Solid State Physics 31, Edited by H. Ehrenreich, F. Seitz and D. Turbull, (Academic Press, New York, 1976).
    \bibitem{S. E. Brown, M. Pieper, W. G. Clark, A. Lacerda, and B. Alavi} J. Phys. IV (France) 9, Pr10-187 (1999).
    \bibitem{S. E. Brown, W. G. Clark, and G. Kriza} Phys. Rev. B 56, 5080 (1997).
    \bibitem{W. H. Wong, M. E. Hannah, W. G. Clark, B. Alavi, and G. Grün} Phys. Rev. Lett. 72, 2640 (1994).
\end{thebibliography}
32 T. Osada, N. Miura, I. Oguro, and G. Saito, Phys. Rev. Lett. 58, 1563 (1987).