^{77}Se NMR Probe of Magnetic Excitations of the Magic Angle Effect in $(TMTSF)_2PF_6$

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We report ^{77}Se spin-lattice relaxation rates for $(TMTSF)_2PF_6$, carried out in the regime where a set of spectacular transport anomalies known as the ''magic angle effects'' are observed. *In situ* resistance measurements (R_{zz}) were used to verify the experimental conditions and give precise sample alignment information. We found that the ^{77}Se T_1^{-1} exhibits no significant changes as the magnetic-field orientation is rotated through the magic angles, and conclude that there is no evidence for either a single-particle gap or a spin gap. The clearly observed field-induced spin-density wave transition temperature is also, unexpectedly, not enhanced at the magic angles.

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$(TMTSF)_2PF_6$ is well known as the first organic superconductor and more generally for the variety of ground states that can be stabilized using high pressure and magnetic field as tuning parameters [1]. At ambient pressure, the system undergoes a metal-insulator transition to a fully gapped spin-density wave (SDW) state at approximately $T = 12$ K. Above a critical pressure $P_c \sim 0.6$ GPa, the system is superconducting when cooled below $T_c \sim 1.2$ K. There is experimental evidence [2] suggesting a spin-triplet pairing superconductivity in $(TMTSF)_2PF_6$, which is otherwise extremely rare [3,4]. As usual, suppression of the superconductivity by a magnetic field greater than the upper critical field H_{c2} recovers a conducting phase. A cascade of field-induced SDW (FISDW) phases is observed above a threshold magnetic field $H_{\rm th}$ [5]. At intermediate fields ($H_{c2} < H < H_{\rm th}$), a phenomenon known as the magic angle effect (MAE) was discovered [6–8] after the prediction of FISDW enhancements at MAs made by Lebed [9]. The MAE is observed when a constant magnetic field is rotated in a plane orthogonal to the molecular stacking axis (**a** axis, the conducting chain). Most of the subsequent work has centered on the sharp magnetoresistance dips seen at these angles, which correspond to commensurate reciprocal-space orbits (or interchain orientations). A nonclassical background [10–12] in the resistance is seen at angles corresponding to incommensurate orbits.

The unusual angle-dependent magnetoresistance $R_{ii}(\mathbf{B})$ ($i = x, y, z$, corresponding to current along the **a**, **b**, and **c** axes) is a topic of considerable interest for a number of reasons. First, there is presently no satisfactory explanation for the experimental observations consistent with semiclassical Boltzmann transport theory [11,13–15]. Perhaps the most spectacular failure is the large resonantlike structure recently discovered in the Nernst effect as the magnetic field is rotated through the MAs [16]. Ong *et al.* [17] proposed that the origin is similar to that in the cuprate superconductors [18], where 2D superconducting phase fluctuations dominate the Nernst signal well above the three-dimensional phase transition. In fact, the temperature dependence of $R_{ii}(H, \theta)$ is metal-like ($dR/dT > 0$) at the magic angles, whereas $dR/dT < 0$ when H is applied away from the MAs [10], leading to the proposal that the normal 3D Fermi liquid metal is unstable away from the MAs [19]. Common to these proposals is the idea that the coherent interchain coupling is strongly influenced by the orientation of the magic field due to electronic correlations and large anisotropy. Interlayer coherence itself is a subject of considerable interest in layered correlated electron systems, such as the underdoped cuprates, where interlayer single-particle hopping is incoherent at all temperatures [20]. The entire set of field-induced phenomena in the Bechgaard salts $(TMTSF)_2X$ is of fundamental interest because they are orbital effects in a system lacking closed orbits. There is no energy shift or direct coupling to the transitions by the magnetic field. The cascade of FISDW transitions and the MAE have been observed primarily in the charge channel by transport measurements. The FISDW has been more thoroughly explored and spin relaxation changes have been observed (although there is no spin gap) [21].

What sorely hampers progress in understanding these unusual MA phenomena is the lack of measurements other than charge transport. While a thermodynamic probe is an obvious choice for establishing the presence of unknown phases or fluctuations in $(TMTSF)_2PF_6$, the high pressure environment makes a measurement of specific heat or dc magnetic susceptibility impractical. Magnetic torque measurements on $(TMTSF)_2ClO_4$ suggest there is a thermodynamic component to the MAE [8]. We opted to study the ^{77}Se NMR spin-lattice relaxation rate, because it is sensitive to the temperature dependence of the magnetic excitations via the hyperfine coupling. They were performed *in situ* with transport measurements in two experimental runs under different conditions. Our results give no spectroscopic evidence for a rearrangement of the density of magnetic excitations resulting from rotating the magnetic field through the MAs. That is, there is no evidence for either a spin gap or a single-particle gap. Furthermore, there is no evidence for an enhancement of the FISDW transition temperature to within experimental uncertainties. The dramatic contrast between the charge channel and the spin channel at the MAs suggests that spin and charge degree of freedom are decoupled [22,23]. The thermodynamic and suggested coherent-incoherent transitions are therefore the result of interaction and correlation effects due to subtle changes in the electronic wave function and density wave susceptibilities.

In each experiment, a high-quality single crystal of $(TMTSF)_2PF_6$ was mounted inside a small NMR coil with the **a** axis aligned with the coil axis. Four Au wires were attached with Ag paint for **c**-axis four-probe measurements of R_{zz}, which was used to verify that the experimental conditions were such that the MAE was observed, and to use that information to align the crystal axes relative to the magnetic field. The sample and the coil were mounted in a BeCu pressure cell. In the first of the experiments, the pressure $P \approx 1.0$ GPa, the magnetic field applied was $B_0 = 4.91T$, and $T \geq 1.4$ K. In the second experimental run, the parameters were changed so that we could observe changes associated with nearness to the FISDW phases: $P \approx 0.85$ GPa, $B_0 = 7.3T$, and the minimum temperature was $T = 0.3$ K. Only a weak angular dependence was observed outside the FISDW phase boundary. All magnetization recovery curves followed a single exponential form.

In Fig. 1(a), we show the angle dependence of R_{zz} for $B_0 = 4.91T$ and different temperatures. As the temperature is lowered, there is an evolution from what is expected semiclassically, $\Delta\rho \propto \sin^2(\theta)$, to where MA dips are seen. Several specific angles are labeled, such as $-1L$, $-2L$, max, etc., corresponding to where features in R_{zz} are observed. The temperature dependence of the relaxation rates for several angles is presented in Fig. 1(b), and the same data are replotted in the inset as T_1T vs T. Perhaps the most distinctive aspect of the data is the lack of any measurable angular dependence for all temperatures.

We would like to discuss these results in the context of previous measurements of the relaxation rates in $(TMTSF)_2PF_6$ [24–26] and their interpretation. It is natural to suggest that there are two regimes. At the lowest temperatures, the variation is nearly linear in temperature as it would be in any metal. However, this behavior changes at higher temperatures where it varies only weakly with temperature. Bourbonnais [27] suggested that both regimes were associated with $Q = 2k_F$ spin fluctuations, but that the distinction arose from the dimensionality: the weak temperature dependence at higher temperatures was linked to one-dimensional (1D, Luttinger liquid) behavior, and a dimensional crossover to a 2D or a 3D Fermi liquid regime was responsible for $T_1^{-1} \sim T$ at the lowest temperatures. We note that the 1D → 2D crossover is disputed [28]. Here we offer an interpretation without the need for

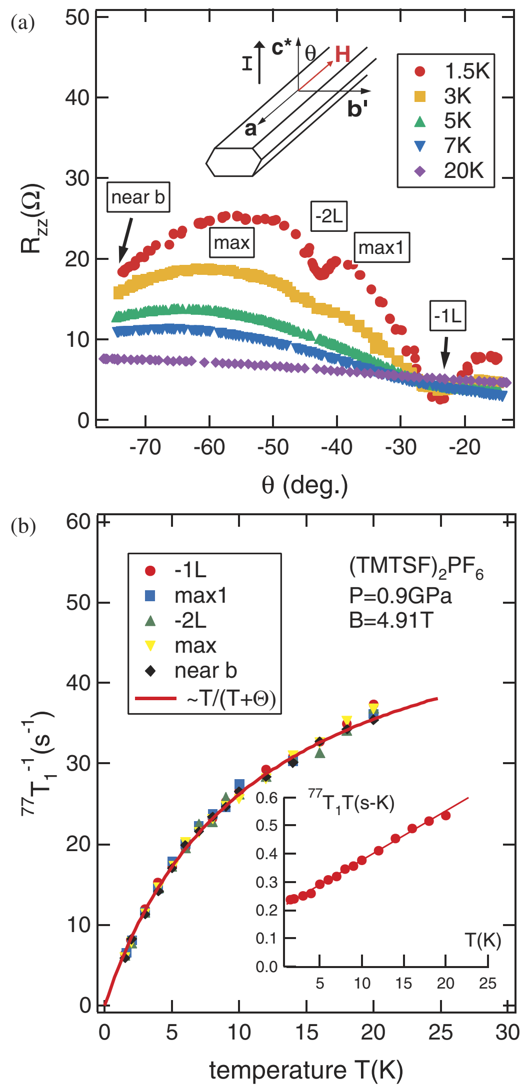

FIG. 1 (color online). (a) Angle dependence of magnetoresistance $R_{zz}(B = 4.91T)$ at selected temperatures: 1.5, 3, 5, 7, and 20 K. Several specific angles, both MAs and non-MAs in the rotation about **a** are marked. The inset shows the field orientation relative to the crystal axes. (b) T_1^{-1} vs T, for several orientations marked in (a), including MAs and non-MAs. There is no difference between them within experimental error. The inset replots the same data as T_1T vs T. The solid line indicates the data are well described by $T_1T \sim T + \Theta$ (see text).

a dimensional crossover. The linear variation of T_1T with temperature indicates the data are well described by a Curie-Weiss-like (CW) expression for the relaxation, $\frac{1}{T_1T} \sim \frac{1}{T+\Theta}$, with $\Theta \sim 11K$ a pressure-dependent constant.

Moriya and Ueda [29] and Millis, Monien, and Pines [30] have applied a spin-fluctuation model to interpreting NMR relaxation data in the high-T_c superconductors. Assuming that the susceptibility is peaked around the antiferromagnetic wave vector, the scaling gives $(T_1T)^{-1} \sim \xi^{z+2-\eta-d}$. For an overdamped response, we expect $z = 2$. A mean-field expression for the temperature-dependent correlation length $\xi \sim (T + \Theta)^{-1/2}$ and $\eta \approx 0$ leads to the CW form for $d = 2$. Perhaps the only unusual feature is the linear variation of $T_1 T$, indicating the correlation length varies to the lowest temperatures rather than saturating below $T \sim \Theta$ [29].

Our expectation is that if an angular dependence of T_1^{-1} were observed, it would be a result of magnetic-field-induced changes in the spin-fluctuation spectrum. Therefore, the absence of angular dependence of ^{77}Se NMR T_1^{-1} allows us to rule out the existence of a single-particle gap or a collective spin gap.

Missing from the data presented in Fig. 1 is any signature of the nearby FISDW phases. However, in Ref. [16], the Nernst effect about the $-1L$ angle appears to be strongly influenced by the FISDW state. Further, there is a suggestion that FISDW fluctuations result in an effective dimensionality crossover at the non-MAs, thus producing the nonclassical background in R_{zz} [31–33]. The parameters for the second experiment allowed for an exploration of the role of the FISDW in the MAE by using a greater magnetic field, $B_0 = 7.3T$, and by measuring to a lower minimum temperature, $T = 0.3$ K.

Transport data for these conditions are shown in Fig. 2(a). In the main panel is the resistance $R_{zz}(B = 7.3T)$ vs θ for two temperatures. The transition temperature to the FISDW state for $B \parallel \mathbf{c}'$ is $T_{\mathrm{FISDW}} = 1.2$ K. The principal difference between the results from $T = 0.3$ K and $T = 1.44$ K is that there is a FISDW present for much of the rotation at the lower temperature. The inset shows R_{zz} vs T at the MA $-1L$, in which the transition is very clear from the sudden increase in resistance on cooling through $T \approx 1$ K.

The accompanying data for T_1^{-1} at several angles are shown in Fig. 2(b). For four of them, the transition to the FISDW state is evident from the sharp increase on cooling through $T \approx 1$ K. Note that the onset temperature does not change noticeably when the angle is rotated through the $-1L$ magic angle. We chose several temperatures for evaluating the angle dependence of T_1^{-1}, all in the normal state. These are shown in Fig. 3. Proximity to the FISDW state coincides with a weak angular dependence of T_1^{-1}. Again, there is no significant anomaly associated with the MAs. We must conclude that there is not a FISDW enhancement at the MAs and hence that FISDWs are not responsible for the MAE. The increase in T_1^{-1} below the transition can be either from a quasiparticle contribution analogous to the Hebel-Slichter peak in superconductors or from collective magnetic fluctuations [21,34,35].

We are left with the possibility that charge and spin degrees of freedom are separated here. Consider a strong Coulomb repulsion and the resulting correlation gap at the Fermi energy (E_F) in a strictly 1D system. Sufficient interchain coupling leads to a density of extended states at the Fermi energy. A perpendicular magnetic field causes the suppression of interchain coupling and restoration of the charge gap. A large Nernst effect results if E_F lies symmetrically in the gap. However, all this happens in the charge channel and the spin channel does not participate in the MAE.

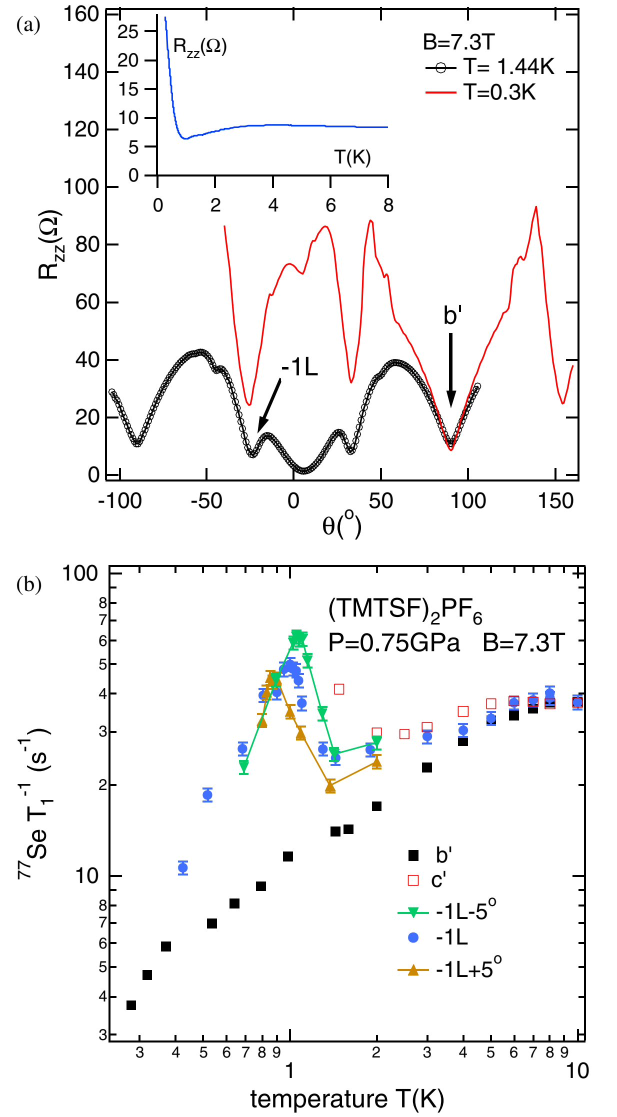

FIG. 2 (color online). (a) Angle dependence of magnetoresistance $R_{zz}(B = 7.3T, \theta)$. Shown are results for 0.30 and 1.44 K. The inset shows $R_{zz}(T)$ at the magic angle $-1L$, and the sharp rise of R_{zz} marks the FISDW transition. (b) Relaxation rates 77T_1^{-1} vs T for selected orientations of the magnetic field. There is a weak angle dependence of T_1^{-1} above 2 K. See Fig. 3. T_{FISDW} is defined by the onset of the T_1^{-1} peak. No significant change of T_{FISDW} is observed around the $-1L$ magic angle.

Finally, we comment on the proposal that the giant Nernst response is related to superconductor vortex flow [17]. The model depends on the existence of superconducting fluctuations that contribute to the conductivity in a layer of TMTSF chains; these fluctuations persist throughout the regime where the MAE is observed. A magnetic field suppresses phase coherence in the usual way *except* at the MAs, where interchain motion is parallel to the field. Assuming there exists an associated gap or pseudogap in the quasiparticle spectrum, on general grounds, some modification in the spectrum should occur under rotation

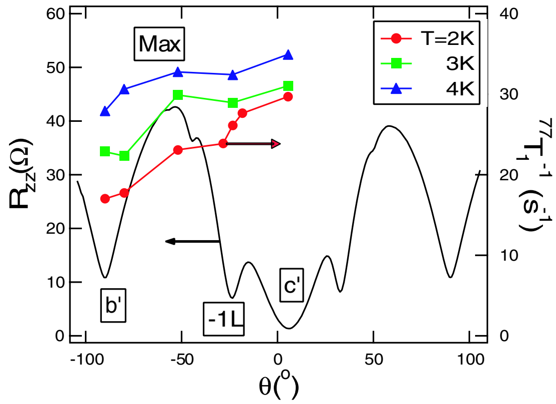

FIG. 3 (color online). Plotted using the left-hand axis is R_{zz} vs θ at $T = 1.2$ K. Plotted using the right-hand axis is ^{77}Se T_1^{-1} for several temperatures, all greater than the FISDW ordering temperature. There is no significant change of T_1^{-1} when the direction of **H** passes through the $-1L$ magic angle.

of the field, and therefore also in T_1^{-1}. To within the accuracy of our measurements, we see no significant angular dependence in T_1^{-1}, nor do we observe any evidence for pseudogap formation in $(T_1T)^{-1}$ at any temperature. If the model is applicable, then the superconducting state is gapless with very little change in density of states with angle or temperature.

In summary, we measured the ^{77}Se NMR spin-lattice relaxation rate T_1^{-1} and magnetoresistance in $(TMTSF)_2PF_6$ at both MAs and non-MAs. T_1^{-1} shows no measurable change on rotation through the magic angles. Our results demonstrate that the MAE involves neither the formation of a single-particle gap nor a collective spin gap. If there is interesting many-body physics, it is in the charge channel and not the spin channel. We searched for but did not find a shift in FISDW transition temperature when the field is rotated to the magic angles to $\pm .1K$, contradicting the original proposal [9] which led to the discovery of the magic angle effects.

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