Lebed's Magic Angle Effects in $(TMTSF)_2PF_6$

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Lebed predicted striking effects in both the metallic and field-induced spin-density-wave (FISDW) states of quasi-one-dimensional conductors when the orientation of the magnetic field causes commensurate electron motion in the least-conducting plane. We have observed sharp cusplike dips in the normal-state magnetoresistance of $(TMTSF)_2PF_6$ suggesting a giant resonance at the commensurate angles. On entering the FISDW state some of the dips become peaks, others fade, and fractional states appear. Magic angle effects are also prominent in the phase boundary of the FISDW state and in the Hall resistance.

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The organic conductor family $(TMTSF)_2X$, where TMTSF is the tetramethyltetraselenafulvalene and $X=PF_6,ClO_4,AsFe_6$, etc., has been extensively studied as the first organic superconductor [1] and more recently for the quantum Hall effect in the field-induced spin-density-wave (FISDW) state [2,3]. The compounds are highly anisotropic with the band parameters along different directions given by $4t_a:4t_b:4t_c=1:0.1:0.003$ eV. The Fermi surface consists of a pair of sheets with weak modulations along the b and c directions, and the system is an open-orbit metal. The Fermi sheets almost nest and application of a magnetic field along the least-conducting c direction enhances the nesting and induces the FISDW state. The competition between the SDW wave vector and the reciprocal of the magnetic length, $G=2\pi/\lambda=eHb/\hbar$, leads to a cascade of FISDW transitions [4].

A tilted magnetic field $\mathbf{H}=(0,H_y,H_z)=(0,H\sin\theta,H\cos\theta)$ introduces two periodicities into the system, with $G_y=eH_zb/\hbar$ and $G_z=eH_yc/\hbar$. At certain "magic" angles, when

$$\tan\theta=(p/q)b/c\,, \quad (1)$$

where p,q are integers, the two periods become commensurate with $pG_b=qG_c$, and the electronic motion in the b-c plane becomes periodic in the reduced zone scheme. Away from the magic angles, electron orbits are quasiperiodic and the entire Brillouin zone is swept out. The commensurability effect in $(TMTSF)_2X$ was originally proposed as an enhancement of the FISDW transition temperature from the disappearance of electronic dispersion along the direction parallel to the magnetic field at the magic angles [5]. In the normal state, peaks in magnetoresistance from the resonance of electron frequencies along b and c directions at the commensurate angles were predicted [6]. Subsequent works have predicted various commensurate effects in both normal and FISDW states [7–9].

Evidence for the commensurability effect in $(TMTSF)_2ClO_4$ has been recently demonstrated by angular-dependent studies of magnetoresistance [10,11] and torque [11]. Features occur at the same magic angles for different fields. However, the features were generally weak and best revealed in the second derivative. In the FISDW state, features in the magnetoresistance near the magic angles have been reported in $(TMTSF)_2ClO_4$ [12,13]. Experiments in $(TMTSF)_2ClO_4$ are complicated by an anion ordering transition at 24 K, which doubles the lattice constant along the b direction to $2b$ and thereby alters the above magic angle criterion.

We have performed commensurability-effect studies of $(TMTSF)_2PF_6$ under 10 kbar of pressure. $(TMTSF)_2PF_6$ is a less complex system which shows almost textbook FISDW behavior at this pressure. At the angles with $p/q=$integer, very sharp minima in the magnetoresistance are observed in the metallic state. The hierarchy of the resonance reflects the underlying lattice symmetry. The commensurability effects are also clearly seen in the FISDW state but with a very different character. The largest normal-state dip disappears, the dips become peaks, and fractional p/q values are observed. A dip in the FISDW transition temperature is observed as direct confirmation of the original Lebed prediction. The vast differences between behavior in metallic and FISDW states suggests that two different mechanisms are at work. The Hall effect exhibits local minima at the magnetic angles as a proof of the commensurability effect in the FISDW state.

High-purity samples were grown electrochemically using the standard technique. The experiment was performed using a miniature pressure clamp in conjunction with a superconducting split-bore magnet. The details of the pressure apparatus are described elsewhere [3]. The sample was mounted in the conventional six-probe geometry with the a axis oriented normal to the field orientation. The pressure cell was placed inside a ^{3}He refrigerator and the entire cryostat was rotated by a goniometer in the b-c plane of the sample. Angular resolution is better than 0.004°. No cracks in the resistance of the sample were observed during the cooldown. A resistivity ratio of better than 300 was obtained between room temperature and 4.2 K. The results in this Letter come from a $(TMTSF)_2PF_6$ sample under 10 kbar of pressure. Similar results were also observed for another sample under 8.5 kbar of pressure.

The angular dependence of the magnetoresistance of $(TMTSF)_2PF_6$ under 10 kbar at 0.5 K is shown in Fig. 1 for nine different magnetic fields. Riding on top of a smooth background, very sharp minima in the magnetoresistance are observed at $\pm 90°$ (the b axis or $p/q=\infty$), 7° (c axis or $p/q=0$), 33° ($p/q=+1$), and $-22°$ ($p/q=-1$) for *all* magnetic fields. In the metallic state below 5 T, the magnetoresistance features at commensurate angles sharpen under increasing magnetic field. In addition, the magic angle structures are stronger for the angles below the c axis. Weaker features at $-42°$ ($p/q=-2$) and at $-8°$ ($p/q=-\frac{1}{2}$) are observed below the c axis but not above. While the resistance at $\pm 90°$ is nearly field independent, the broad background resistance increases almost linearly with the magnetic field and shows no signs of saturation.

For fields above 5 T, the angular dependence is strongly changed by the FISDW's. The FISDW's presumably depend on the $\mathbf{c}^*$ projection of the magnetic field and onset when $H_{c^*} \geq 5$ T. We see that the commensurate effects change considerably. The largest dip at $\theta=0$ virtually disappears. The $p/q=-1$ commensurate feature changes the most with the dip becoming a peak in magnetoresistance, and the $p/q=-\frac{1}{2}$ commensurate feature at $-8°$ begins to appear.

Since the real crystal structure is triclinic and not orthorhombic, deviations from the magic angle formula of Eq. (1) are observed. The proper magic angle formula becomes [10,11]

$$\tan\theta = \left(\frac{p}{q}\right)\frac{b\sin\gamma}{c\sin\beta\sin\alpha^*} - \cot\alpha^* , \qquad (2)$$

where an ambient pressure $b=7.711$ Å, $c=13.522$ Å, $\alpha^*=95.71°$, $\beta=86.21°$, and $\gamma=71.01°$. These parameters are expected to change under pressure, and a deviation of about 1° is observed in the experimental results from the expected commensurate angles.

One striking feature of the data is the anisotropy of the magnetoresistance under tilted magnetic field. When magnetic field is parallel to the b axis, virtually no magnetoresistance is observed. As the sample is rotated toward the c axis, a smoothly increasing background resistance is observed with the dips appearing at the commensurate angles. The resistance maximum is found near magnetic field parallel to $\mathbf{c}^*$. This enormously large and nonsaturating magnetoresistance cannot be reconciled with the known band structure in terms of the relaxation-time approximation under the Boltzmann equation [14,15]. The magnetoresistance anisotropy cannot be found in the previous works [16–18]. Figure 2 shows the angular dependence of the magnetoresistance from 4 to 9 T at 2.2 K. At slightly higher temperatures, magnetoresistance can be studied to higher magnetic fields without complications from the FISDW transitions. The commensurate features are more rounded and not as strong as the 0.5-K result. There is virtually no magnetoresistance for $\mathbf{H}$ parallel to $\mathbf{b}$. At high field, the background magnetoresistance is fitted by a simple $\cos\theta$ form. At lower field the background is flatter, something like $(\cos\theta)^{1/2}$ at 4 T.

Figure 3 shows the Hall effect at 7, 8, and 9 T in the FISDW regime. Consequently, rotation near $\mathbf{c}^*$ ($\theta=0$) results in sweeping through the FISDW transitions, and the Hall effect decreases in a steplike fashion. In the ab-

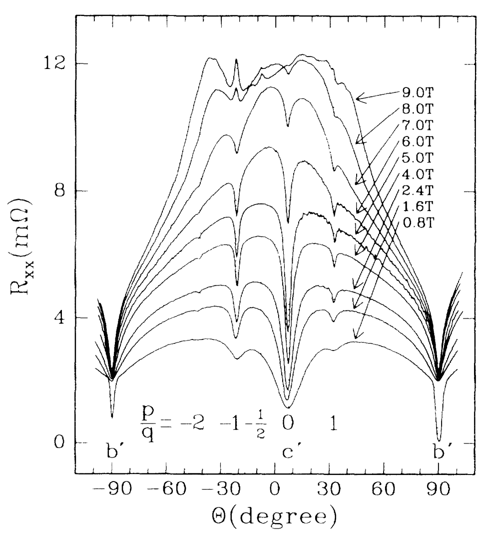

FIG. 1. Angular sweeps of magnetoresistance of $(TMTSF)_2PF_6$ under 10 kbar of pressure at 0.5 K. At the lowest field angular sweep of 0.8 T, partial superconductivity is observed near the b axis.

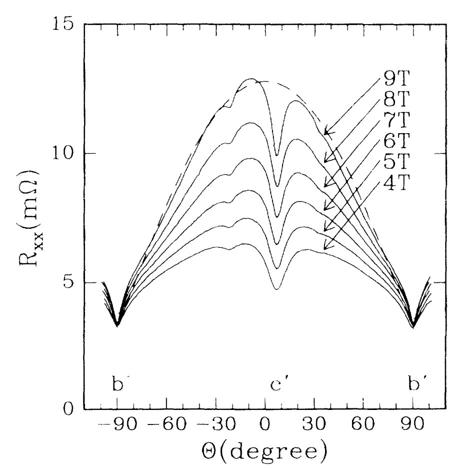

FIG. 2. Angular sweep of magnetoresistance at 2.2 K for 4 to 9 T of magnetic field. The high-field sweeps can be fitted as $\cos\theta$, as shown by the dashed curve with the resistance at the b axis as the minima.

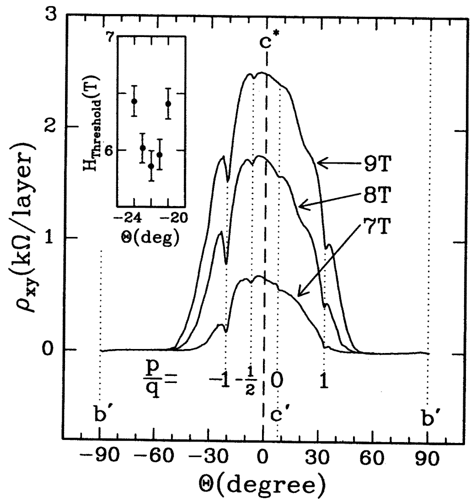

FIG. 3. Angular sweeps of Hall effect in the FISDW state at 7, 8, 9 T. Inset: The FISDW threshold field near the $p/q=-1$ commensurate state at 22°.

sence of any effect due to the bandwidth along c, the Hall resistance would be a symmetric function of $H\cos\theta$, the same function as for $\theta=0$ and changing H. In addition to the Hall effect from the FISDW, dips are found at the same magic angles (for all fields) where commensurate features in magnetoresistance are observed. Downward cusps in the Hall effect at $p/q=-1$, $-\frac{1}{2}$, 0, and 1 are seen. The $p/q=-1$ feature is the strongest. Since the Hall effect in the FISDW states are very sensitive to the phase diagram, the decrease in the Hall effect is indicative of the FISDW states at the commensurate angles differing from the neighboring states at the incommensurate angles. Similar arguments can be made on the magnetoresistance peaks in Fig. 1. Such an effect may be due to the disappearance of the third direction bandwidth at the commensurate angles [5].

In the previously reported measurements on the commensurability effect in $(TMTSF)_2ClO_4$ [10,11], the weak commensurate structures are observed along with a broad background resistance. In our experiment of $(TMTSF)_2$-PF_6, strong commensurate features are observed as the deep minima at the magic angles. The structures seen here are sufficiently strong and well defined to rule out several models and to suggest some others. Before comparing our results with the previously proposed models we make some general observations. If these striking dips are associated with commensurability effects, then the electrons must have traversed the commensurate orbit at least once before scattering, $\omega_{c,p/q}\tau > 1$, where $\omega_{c,p/q}$ is the frequency for the particular orbit. $\omega_{c,p/q} \propto B$ and decreases for higher p and q. Therefore it is surprising that more magic angles are not seen as the field is increased. All of the structure seen in the metallic state is already present at 0.8 T at 0.5 K ($p/q=0, \pm 1, -2$). This implies that the scattering time is quite long and that the structures that are seen are likely to be the only ones present at any field in the metallic state.

The observed hierarchy of the commensurate features seems to reflect the underlying anisotropy of the system. For an isotropic system, both p and q should be equally favorable. The anisotropy of the system seems to be observable for the $p/q=1$ and -1 features. The commensurate structure at $p/q=-1$ is noticeably sharper than the $p/q=1$ feature in both magnetoresistance and the Hall effect. In fact, the smaller, satellite features at $-42°$ ($p/q=-2$) and at $-8°$ ($p/q=-\frac{1}{2}$) are observed only below the c axis. We attribute the difference as simply a reflection of the underlying lattice symmetry. Since the crystalline structure is triclinic, the diagonals across the b-c zone are unequal. Consequently, the cyclotron frequency at $p/q=-1$ is smaller than at $p/q=1$ and gives rise to a greater magnetoresistance dip.

Lebed and Bak calculated the magnetoresistance of $(TMTSF)_2X$, taking account of the finite bandwidth in the c direction and the effects of a tilted magnetic field on the impurity and electron-electron scattering [6]. Resonance of the scattering rate at the commensurate angles leads to the peaks in magnetoresistance, in contrast to the clear dips seen in the experiments. In addition, the largest commensurate feature at the $p/q=0$ is not found in this model. In a clever quasiclassical model proposed by Osada, Kagoshima, and Miura, only commensurate motion allows for a finite average velocity and hence conductivity enhancement along the direction of the magnetic field [19]. The conductivity peaks correspond to transfer integrals between molecules in the commensurate directions and thus the number of peaks is limited to a few neighbors. In a recently proposed model, Maki suggests that the magnetoresistance dips arise from the periodicity of the electron frequencies at the commensurate angles [20]. The commensurability effect is expected for integral resonances only, not fractional as observed in this experiment.

In both of these models, the dominant effects are on the transport properties in the z direction. The effects should be orders of magnitude smaller along the x direction. Since our measurements along **x** show effects of order 100% at the magic angles, we do not think these models are appropriate. Moreover, the magnetoresistance along **z** in these models shows smooth dips at the magic angles and sharp peaks between them whereas the experiment shows sharp dips at the magic angles and smooth maxima between.

Another possibility, which these experiments suggest, is that the metallic magic angle effects are the results of "hot spots" on the Fermi sheets where the scattering rate is high [21]. In a magnetic field the electrons are swept into the hot spot and the average scattering rate increases, explaining the large longitudinal magnetoresistance. At the magic angles the electrons repeat their trajectory and only a fraction of the electrons are swept to the hot spot. For other angles an electron trajectory will take it everywhere on the Fermi surface and particularly to the hot spot. The shape of the hot spots determines the form of the magnetoresistance and its angular dependence.

The different characteristic behaviors of the magic angle effects in the metallic and FISDW states suggests that they have different explanations. Specifically the $p/q=0$ dip is dominant in the metal but barely observable in the FISDW. $p/q=-1$ changes from a dip to a peak and the $p/q=-\frac{1}{2}$ is only seen in the FISDW. A likely candidate for the FISDW magic angle effects are the original predictions of Lebed [5] or those of Chen and Maki [8]. In these models the dispersion along the z direction, which sets the zero-temperature threshold field for the FISDW, effectively disappears from the problem for commensurate fields which include both b and c components. For magic angles the FISDW onset field should therefore be lower, even at finite temperatures. To test this idea the FISDW onset was measured near the $p/q=-1$ state by measuring the resistance versus field at several fixed angles. The phase boundary is shown in the inset of Fig. 3 and we find the predicted dip at the magic angle. It is equally interesting to note that in Lebed's theory there is nothing which happens at $p/q=0$ and that is largely what we observe in the FISDW state.

This picture may not be quite so simple. Lebed's idea is that the FISDW is more stable at the magic angles. That would explain the peak in the magnetoresistance at the magic angles at high field. However, if anything, it would suggest that the Hall resistance should also increase, since that is the usual effect of higher field. The experimental observation that the Hall resistance has a dip suggests a more profound effect of the magic angles on the FISDW.

Calculation of FISDW transition temperature under tilted magnetic field shows a dip in transition temperature around the $p/q=\pm 1$ commensurate angles, but this is predicted for $n=0$ or $n=2$ states [8]. A field in excess of 15 T is necessary to access $n \geq 2$ FISDW states. Alternatively, the presence of a new periodicity along the c direction results in an additional freedom in the SDW vector. At p/q commensurate angle, each Landau band splits into q subbands, giving rise to a possible "fractional" quantized Hall effect [7]. It is tempting to attribute the apparent reduction in the Hall effect at the magic angles as a signature of possible fractional quantization of the Landau bands, especially the $p/q=-\frac{1}{2}$ states, since this feature is observable only in the FISDW state. However, higher magnetic field is necessary to fully distinguish the FISDW contribution from that of the commensurability effects.

In summary, we have studied the effects of commensurate motion of the electrons on the Fermi sheet of $(TMTSF)_2PF_6$ induced by the application of a magnetic field tilted to magic angles. Striking features mark the commensurability effects in both the metallic and FISDW states. The background angular dependence of the large magnetoresistance in the metallic phase coupled to the magic angle effects point to a new mechanism, possibly hot spots on the Fermi sheet, for magnetotransport. The reduction of the FISDW threshold field at commensurate fields confirms Lebed's original prediction, but the behavior of the Hall resistance in the FISDW state requires additional effects at the magic angles.

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