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H_{c2} enhancement and giant Nernst effect in $(TMTSF)_2(PF)_6$

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Abstract. We discuss two recent discoveries in the title compound: 1) A strong enhancement in the critical field as the critical pressure for the SDW-superconductor phase boundary is approached and 2) the existence of giant resonances in the thermoelectric voltage as an applied magnetic field is rotated through the "Lebed Magic Angles". 1) H_{c2} is highly anisotropic in this salt, but the enhancement is present for all field orientations. We suggest a model in which the near degeneracy of the superconducting and SDW states leads to a coexistence. The critical field is enhanced by the formation of thin slabs of superconductor (less than the penetration depth) parallel to the applied field and sandwiched by insulating SDW layers. As the temperature is lowered the slabs become thinner allowing a higher H_{c2}. 2) The thermoelectric voltage resonance indicates that the currents generated are "locked-in" to the interchain direction ("magic angle") nearest parallel with the applied field. With the field to one side of the magic angle there is a Lorentz force up, on the other side of the magic angle the Lorentz force is down. The size of the thermoelectric voltage is 10^4 larger than might be expected from conventional transport theory.

1. CRITICAL FIELD ENHANCEMENT

In the late '80's Andrei Lebed suggested an interesting way to increase the critical field of quasi-one or quasi-two dimensional superconductors[1,2]. A field applied perpendicular to the least conducting direction would decouple the conducting planes in a dimensional crossover. This would lead to a reduction in screening currents and an enhancement of the orbital critical field with a strong upward curvature in H_{c2} vs. T. His aim was to suppress orbital pairbreaking so that spin pairbreaking effects would become evident and could be used to test whether the Cooper pairs were singlet or triplet. Following his ideas we performed experiments on the title compound. We aligned the field along the b direction and found an enhancement so large that H_{c2} exceeded the Pauli limit (the spin pairbreaking field for singlet superconductors) by more than a factor of 4. A remarkable critical field of 9 Tesla for a 1.4K superconductor[3-5]. That study along with more recent NMR measurements strongly suggest triplet superconductivity[6-8]. However, Lebed's suggestion of field induced dimensional crossover (FIDC) was exciting enough that it deserved verification on it's own. We therefore performed a series of experiments to see: a)whether the enhancement vanished when the field was aligned along the c direction (perpendicular to the conducting planes), b) whether the anisotropy in critical field increased at lower temperatures as the higher applied fields decoupled the planes, and c) to observe the cusp like angular dependent critical field associated with two dimensional superconductors. What we found suggests that the H_{c2} enhancement mechanism in $(TMTSF)_2PF_6$ at pressures near P_c is not FIDC, although we suspect that dimensional crossover may well work in other regimes.

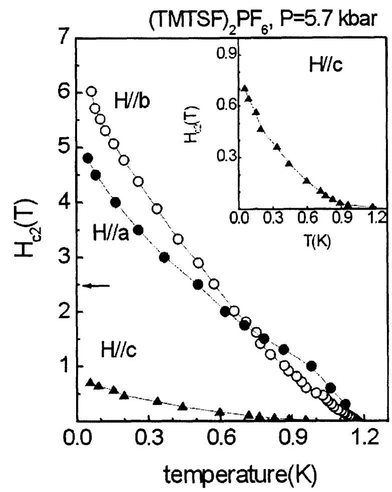


Figure 1. Resistively determined critical field along the principle axes for (TMTSF)2PF6. Insert is expansion of c axis data. All curves are well above the calculated Ginzburg-Landau values. Horizontal arrow indicates the Pauli limiting field for singlet superconductivity.

In figure 1 we show the upper critical field measured along the three crystallographic axes at a pressure of 5.7 kbar, near the SDW-superconducting phase boundary. It is clear that there is a strong upward curvature for all directions instead of the downward curvature usually found in H_{c2} measurements[9]. Dimensional crossover occurs when an orbital magnetic energy is comparable to a perpendicular bandwidth. From Lebed's work we expect the crossover field to be ~5 tesla for H//b, somewhat higher for H//a and ~90 tesla for H//c. These are untenable values for all of our data but particularly striking for H//c. The insert in figure 1 enlarges the H//c data. H_{c2} is much enhanced over the GL value of ~0.03 tesla (which is observed at higher pressure or extrapolated from the data near Tc) even though the fields are much less than the 90 tesla for crossover. We need another mechanism.

A strong hint as to mechanism comes from a study of H_{c2} as a function of pressure near Pc ~6kbar the critical pressure separating the SDW and superconducting phases. As shown in figure 2 the enhancement grows as pressure is lowered toward the SDW phase. In fact the superconducting transition temperature is essentially independent of pressure in this region and there is evidence in both our work[10] and the work of others[11] that the two phases coexist in this region. Since Tc is unaffected, the SDW is not destructive for superconductivity but merely competes with it for density of states at E_F. Coexistence and filamentary superconductivity and this can yield an enhancement of H_{c2} as long as the filaments are smaller than the penetration depth. Filaments perpendicular to the applied field would go normal at low field, but for parallel filaments the effect would be the same as for a thin superconducting film in a parallel magnetic field where H_{c2}~(λ/d) H_{c0} ~$(Tc-T)^{1/2}$, where H_{c0} is the thermodynamic critical field, which has strongly downward curvature.

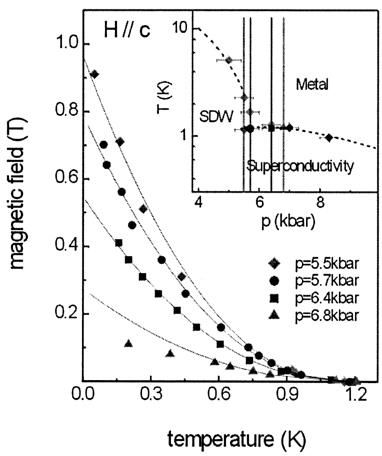


Figure 2. C axis critical field for several pressures near the SDW-superconducting phase boundary. Insert – measured phase diagram with vertical lines indicating samples shown in main plot.

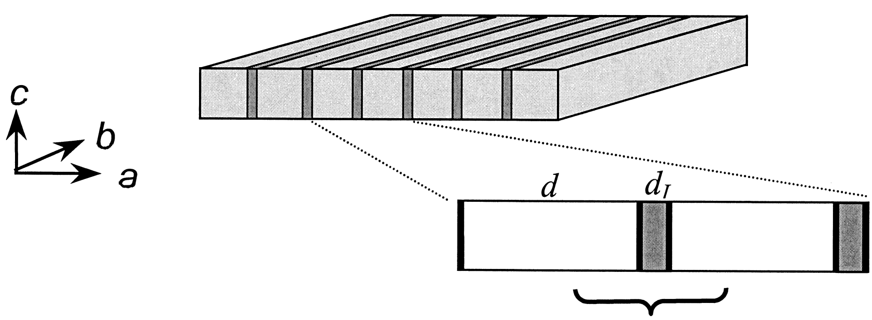
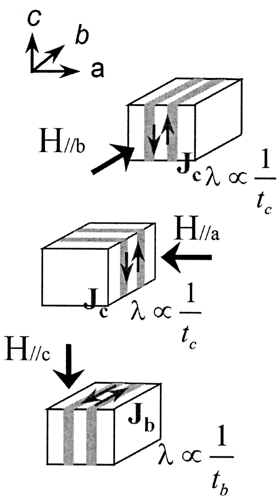


Figure 3. Cartoon of the slab arrangement of superconducting – insulating (SDW) sandwiches for applied field in the c direction.

The mechanism we propose assumes that the insulating SDW phase and the superconducting phase are nearly degenerate[10]. In that case the SDW and superconducting phases can be readily interchanged and rearranged so that the superconductor lies in thin slabs parallel to the applied field (see figure 3). We estimate the critical field by associating the reduction in Tc with the energy cost of generating screening currents plus that of introducing interfaces between super and SDW phases: $\delta T_c = T_c - T_{c0} \sim ((d/\lambda)H)^2 + \gamma/d$, where d is the slab thickness, λ the penetration depth and γ the surface tension. The last term results from the fact that the number of interfaces increases as the distance between interfaces decreases. Minimizing with respect to d and solving for H in terms of Tc we have:

$$H \propto \frac{\lambda}{\gamma} \delta T^{3/2}$$

If the two states are not degenerate then each slab costs some additional energy $\delta f_{SI} \propto P-P_c$, which effectively augments the surface tension, $\gamma' = \gamma + \delta F_{SI}/2$, as illustrated in figure 3.



The model predicts an upward curvature given by the $\delta Tc^{3/2}$ dependence (experimentally H//b $\propto \delta Tc^{3/2}$, H//c $\propto \delta Tc^2$), with the enhancement increasing as pressure is lowered toward Pc as observed in figure 2. Furthermore the relative values of the enhanced H_{c2}'s should correspond to slab configurations which yield the maximum penetration depths. As cartooned in figure 4 we want the screening currents along the least conducting directions. The penetration depth is inversely proportional to the bandwidth. In GL theory the critical field is proportional to the bandwidth in the applied field direction. For GL $H_{c2}//a >> H_{c2}//b >>> H_{c2}//c$, whereas for the present model $H_{c2}//a \sim H_{c2}//b >>> H_{c2}//c$ as observed in figure 1.

Figure 4. Screening currents, and associated penetration depths for the slabs configurations which result in the highest critical fields for applied fields along the specified directions. Note that $H_{c2}//a \sim H_{c2}//b >>> H_{c2}//c$.

2. GIANT NERNST EFFECT NEAR LEBED MAGIC ANGLES.

In the late 80's Lebed also predicted striking effects in the normal state resistance for certain magnetic field orientations relative to the crystallographic axes[12]. These magic angles correspond to commensurate k space "cyclotron" orbits of electrons on the quasi-one-dimensional Fermi surface. The b-c plane field rotation should yield resistance anomalies at these magic angles. Following Lebed's prediction striking resistance dips have been observed at these angles, but their interpretation is not yet resolved[13-18]. There is a real space interpretation which suggests that the magic angles effects are related to the dephasing of electrons tunneling between the highly conducting chains. At the magic angles (which correspond to interchain directions) electrons travel along field lines and are unaffected – the chains are coherently coupled. At other angles the tunneling is dephased and the chains become decoupled. Experiments show that at the magic angles the low temperature resistance decreases with decreasing temperature as for a metal. But off the magic angles the resistance increases with decreasing temperature as for a non-metal[19].

We decided to study this phenomena with thermoelectricity. Thermopower and Nernst effect are effectively mixed thermodynamic-transport measurements. The Nernst effect is the thermoelectric equivalent of Hall effect – a transverse electric field resulting from crossed temperature gradient and magnetic field. The exact orientation of the temperature gradient is unknown in our experiment since it was performed in a pressure cell. The largest signal we observed was odd in sign with reversal of the magnetic field and therefore indicates Nernst effect rather than longitudinal thermopower. In figure 5. we show the a axis resistance of a sample at 2K along with the Nernst voltage generated at 1K and at 2K.

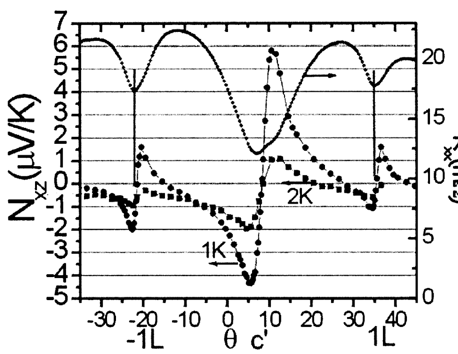


Figure 5. Angular dependence of the magnetoresistance at 2K and the Nernst signal at 2K and at 1K, H=7.5 tesla.

The resistance exhibits the dips at the magic angles c*, 11 and –11 (Lebed angles) conventionally seen. At these same angles the thermoelectric voltage undergoes a sharp resonance like feature which increases strongly as the temperature is reduced. The strong resonances can be qualitatively understood if we assume that in high fields currents "lock-in" to the nearest interchain direction to the applied field. As cartooned in figure 6, we then see that for angles less than the magic angle there is a Lorentz force up, at the magic angle the field and current are parallel and for fields slightly greater than the magic angle there is a downward Lorentz force. As we get further from the magic angles the current is reduced. In this scenario the thermally generated currents are locked in to the magic angle directions and the Nernst voltage increases as the field approaches the magic angle, goes through zero at the magic angle, changes sign and then decays as the field direction is rotated past the magic angle.

The current lockin is expected in either a recent model which attributes the magic angle effects directly to interchain currents or in Osada's model which assumes a Fermi surface with contributions from transfer matrix elements to neighbor and further neighbor chains and then uses conventional Boltzmann transport theory. As seen in figure 7 the thermoelectric voltage increases strongly with decreasing temperature. In either model the voltage expected is on the order of $N_{xy} \sim (k_B/e)(T/T_F)(\omega_c\tau \sin(\delta\theta)) \sim 1nV/K$ which is four orders of magnitude smaller than the observed value at low temperature. Here T_F is the Fermi temperature, $\omega_c = 2\pi e H v_F c/h$, τ is the scattering time and $\delta\theta$ is the difference between the field direction and the magic angle direction. This result suggest that our understanding of the magic angle effects is incomplete and that correlation effects (or perhaps spinon drag) are responsible for the formation of excess heat transport.

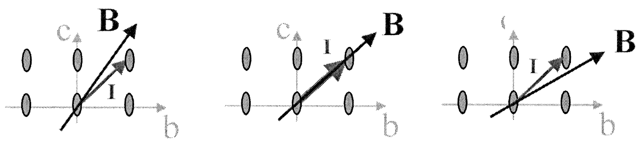


Figure 6. Looking down the conducting TMTSF chains for applied field near the 1L magic angle there is a Lorentz force between the locked in interchain current and the applied field which changes sign as the field is rotated through the 1L angle.

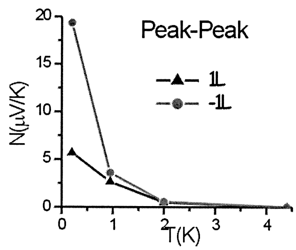


Figure 7. Temperature dependence of the peak to peak resonance in thermoelectric voltage near the 1L and −1L magic angles.

CONCLUSION

We have proposed a new mechanism for enhancement of the critical field in superconductors hear an insulating-superconducting phase boundary. The system can minimize the orbital screening energies by self-consistently subdividing into thin slabs of superconductor separated by insulating regions. The very large critical fields of (TMTSF)2PF6 near the coexistence pressure are consistent with this model. We have also described our experiments on thermoelectric voltages associated with Lebed magic angle effects in this material. These results strongly indicate a current lockin along the magic angle (interchain) directions for applied field near the magic angles. The giant voltages observed however, await further explanation and may provide a important clue as to the nature of the ground state at and away from the magic angles. We thank Phuan Ong and David Huse for helpful discussions and NSF DMR 99-76576 and 98-09483 for support.

[1] A.G. Lebed, Pis'ma Zh. Eksp. Teor. Fiz. 44, 89 (1986) [JETP Lett. **44**, 114 (1986)].
[2] L.I. Burlachkov, L.P. Gor'kov and A.G. Lebed, Europhys. Lett. **4**, 941 (1987).

[3]I.J. Lee, M.J. Naughton, G.M. Danner and P.M. Chaikin, Phy. Rev. Lett., 78, 3555 (1997)
[4] I.J. Lee, P.M. Chaikin and M.J. Naughton, Phy. Rev. B 62, R14669 (2000)
[5] I.J. Lee, P.M. Chaikin and M.J. Naughton, Phy. Rev. B 65, 180502(R)(2002)
[6] I.J. Lee, S.E. Brown, W.G. Clark, M.J. Strouse, M.J. Naughton, W. Kang, P.M. Chaikin, Physical Review Letters, 88, 017004 (2002).
[7] A.G. Lebed, Phys. Rev. B **59**, R721 (1999); A.G. Lebed, K. Machida, and M. Ozaki, Phys. Rev. B **62**, R795 (2000).
[8] C.D. Vaccarella and C. A. R. Sá de Melo , Phys. Rev. B **63**, 180505 (2001).
[9] Upward curvature is often observed in H_{c2} measurements in high Tc superconductors where the effect is related to flux motion in the temperature region between the mean field superconducting transition and the formation of a solid vortex phase. In high Tc materials H_{c2} measured resistively is substantially *lower* than what is calculated from Ginzburg Landau (GL) theory or measured by other techniques. The present measurements indicate critical fields which are substantially *higher* than the GL values, are independent of the criteria used to obtain H_{c2} from the resistive measurements and agree with bulk measurements (e.g. NMR) at the few points where the measurements overlap.
[10] I.J. Lee, P.M. Chaikin and M.J. Naughton, Phys. Rev. Lett. **88**, 017004 (2002).
[11] T. Vuletic *et al.*, Eur. Phys. J. B **25**, 319 (2002).
[12] A.G. Lebed, JETP Lett. **43**, 174 (1986).
[13] T. Osada, S. Kagoshima and N. Miura, Phys. Rev. B **46**, 1812 (1992)
[14] A.G. Lebed, J. Phys. I (France) **4**, 351 (1994)
[15] Y. Sun and K. Maki, J de Physique IV 3, 327 (1993)
[16] P.M. Chaikin, Phys. Rev. Lett. **69**, 2831 (1992).
[17] S.P. Strong, D.G. Clarke and P.W. Anderson, Phys. Rev. Lett. **73**, 1007 (1994).
[18] E.I Chashechkina, P.M Chaikin, Phys. Rev. B **65**, 12405(2002).
[19] E.I Chashechkina, P.M Chaikin, Phys. Rev. Lett. **80**, 2181 (1998).