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# On the angular dependences of the superconducting and normal state properties of the Bechgaard Salts: Triplet Superconductivity, Enhanced $H_{c2}$ near the S-I boundary, Giant Nernst Effect at Lebed Magic Angles

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**Abstract**

Our recent experiments on (TMTSF)$_2$PF$_6$ indicate that: 1) the Knight shift along both the **a** and **b** axes remains unchanged on going through the superconducting transition (suggesting triplet superconductivity with d vector along **c**), 2) Hc2 is enhanced near the critical pressure for the superconducting-insulating transition by the formation of slabs parallel to the magnetic field and the anisotropy in Hc2 is determined by penetration depth rather than coherence length anisotropies, and 3) in the normal state, for fields *near* the Lebed magic angles the current is locked into the direction of the magic angles and there is an anomalous Nernst effect which near the magic angles is ~200 times larger than what would be expected from Boltzmann transport.

*Keywords: NMR, Organic Superconductors, Metal-Insulator Transitions, Magnetotransport, Thermopower*

## 1. Introduction

The Bechgaard salts are notorious for the diversity of their low temperature phases, the richness of their phase diagrams and the inability of the scientific community to put them peacefully to rest after more than twenty years since their discovery as the first organic superconductors. Present excitement results from their potential as prototype non-Fermi liquids[1] and triplet superconductors[2]. In previous ICSM's we have presented experiments on the b axis Knight shift (suggesting triplet – p wave superconductivity), an enhancement of Hc2 which related to the formation of an inhomogeneous state with slabs parallel to the magnetic field and magnetotransport which suggested anomalous behaviour at Lebed Magic angles (fields parallel to the crystallographic axes), suggesting interlayer decoupling[3]. In the present work we show that both the **b** axis and the a axis exhibit triplet like Knight shift, that the Hc2 enhancement near Pc is a consequence of a new inhomogeneous "slab" phase and that for a magnetic field aligned near the Lebed angles the current is "locked in" to a direction connecting the one dimensional conducting chains as evidenced by the presence of a Hall-like effect, the Nernst effect [thermopower equivalent to Hall] as the field is rotated through the Lebed magic angles.

## 2. Knight Shift

Our previous work on the Knight shift which showed no change on cooling into the superconducting state with field aligned precisely along the **b** axis coupled with our

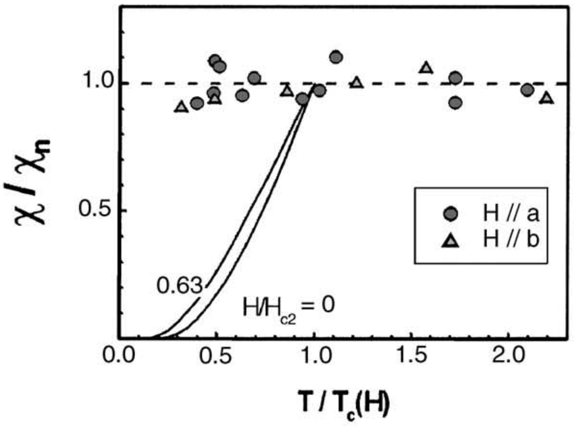

Fig.1) Spin susceptibility derived from Knight shift measurements along the a and b axes. Solid lines are the expected behaviour for a singlet superconductor at tow reduced field values. Dashed line is for a triplet superconductor

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measurements of the anisotropic critical field led to the theoretical expectation that the triplet d vector would be aligned along the **a** axis[4]. In order to test this hypothesis we studied the NMR with field aligned precisely (better than 0.01 degrees) along **a**. The d vector defines the direction along which the susceptibility is zero for an equal spin paired triplet state superconductor. Our more recent experiments with field along **a** also show the absence of a change in the Knight shift. The susceptibility is the normal metal Pauli susceptibility along both the **a** and **b** axes as the sample is cooled through Tc as shown in figure 1. We conclude that the either the d vector is aligned along the **c** axis as for $Sr_2RuO_4$ or that the d vector has realigned with the magnetic field at the 1.2 Tesla field used for the measurements.

## 3. Enhancement of $H_{c2}$ near $P_c$

Due to the highly anisotropic band parameters and particularly the small bandwidth in the **c** direction , Lebed[5] suggested a magnetic field induced dimensional crossover in the superconducting state. A field applied in the **b** direction would most effectively decouple the **a-b** planes, reduce any screening currents along the coupling, **c**, direction and lead to an enhanced Hc2.

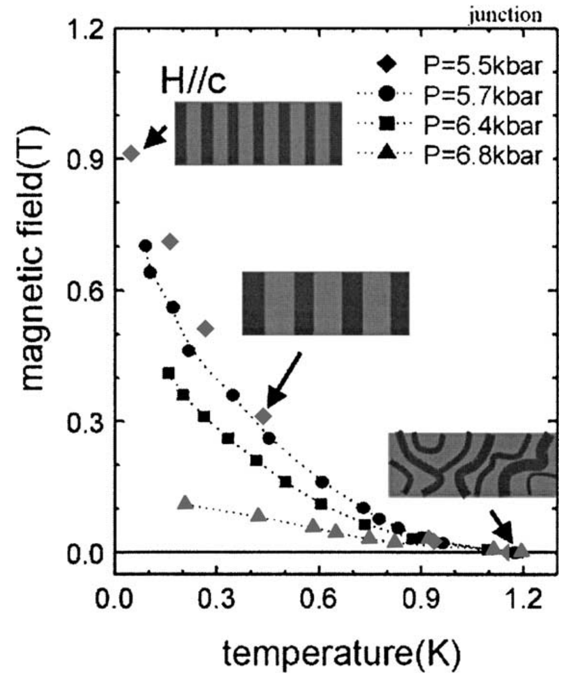

Figure 2) Hc2 vs. T for several pressures near Pc~5.5kb. Our model suggests coexisting SDW-superconducting phases (black-grey) which align with the field and thin as the field is increased.

Early experiments seemed to confirm this picture and produced an upward curvature to the Hc2 vs. T phase diagram. However, the upward curvature was also seen along the **a** and **c** directions. Moreover, the enhancement was pressure dependent and became larger as pressure was lowered toward the critical pressure separating the SDW insulator from the superconducting phase. In figure 2 we show the c axis critical field for several pressures. The low temperature Hc2 increases almost an order of magnitude as pressure is reduced from 6.8 to 5.5kbar. We developed a simple model to explain this behaviour based on the fact that Hc is enhanced in a parallel thin film[6]. If the superconductor and insulator are degenerate then an inhomogeneous state may arise with superconducting slabs separated by insulating regions. When the slabs are less than the penetration depth, λ, in thickness the field penetrates and Hc2 increases. The energy cost is in the surface tension, γ, between the two phases. When the two phases are not degenerate the energy cost in converting superconductor to SDW has the same form as a larger surface tension and vanishes at Pc. The critical field is given by : $H \sim (\lambda/(\gamma+\alpha(P-Pc))\delta T^{3/2}$, where $\delta T = Tc-T$ Experiments indicate hysteresis and hence an inhomogeneous state already at zero field. This coexistence is not sufficient to explain the critical field enhancement. As shown in the cartoons in figure 2 without applied field there is a random orientation of domains. As field is increased the slabs align parallel to the field then decrease in thickness. That is what produces the upward curvature.

## 6. Giant Nernst Effect

Lebed[7] earlier had predicted anomalies in the resistance when the magnetic field was aligned at certain magic angles in the plane perpendicular to the most conducting direction. These angles correspond to the real space translation vectors. Large dips in magnetoresistance have been observed at these angles in several of the Bechgaard salts, but despite many theoretical attempts their origin is still not clear. The effects are sharpest in the $PF_6$ saltun der pressure and therefore only resistance has been measured.

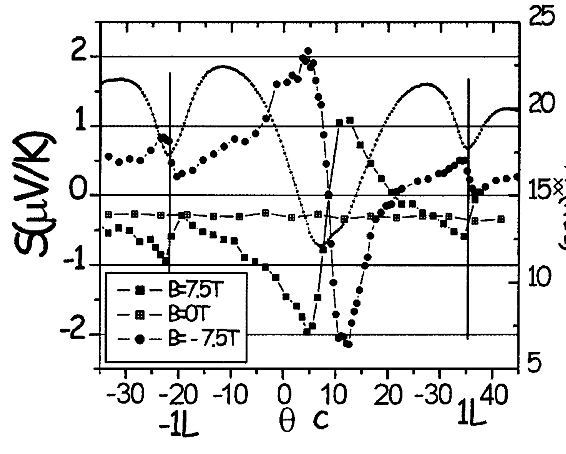

Figure 3) Resistance and Thermoelectric voltage, S, for $(TMTSF)_2PF_6$ as a function of magnetic field direction for b-c plane rotation (2K,10kb). Note that the S is essentially an odd function of applied field.

We planned to do a thermopower study to see whether the metallic state was different at these angles, but when we analyzed our results, shown in figure 3, we concluded that the thermopower (an even function of field) was too small to be observable (aside from the offset due to the gold leads), but that there were large anomalies in the Nernst effect (odd with field) at the magic angles. Although we set up to have a temperature gradient predominantly along the a direction, we must inadvertantly have gotten a component along **c**. Note that in our pressure bomb the heat current flows mostly through the pressure medium, solid Fluorinert, at this temperature. What is most striking about the data is the large resonant like structures as the field is rotated through a Lebed angle. The most straightforward and probably correct interpretation is that when the field is close to a magic angle, the current flows at the magic angle. In more conventional systems, in very high magnetic field the current follows the field lines and does not cross them. In the present case the current effectively "locks in" to the magic angle which corresponds to an interchain direction. This behavior is cartooned in figure 4. If the current locks in then the field makes an angle with the current when it "lags " the magic angle and there is a Lorentz force, a Hall effect and a Nernst signal. On the other side of the magic angle, when the field "leads" the magic angle the Lorentz force, Hall effect and Nernst change sign. (We are preparing toper form the Hall experiment which should confirm this picture.)

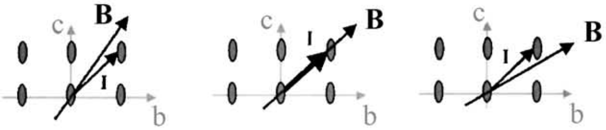

Fig. 4. Looking down the TMTSF chains. For field near the **b+c** direction the current flows only between the chains separated by **b+c**. The Lonrentz force then produces a force along a in the first figure, along –**a** in the last and no force in the middle figure when the field and the current are parallel.

The Nernst effect is the voltage generated in the $\nabla T \times H$ direction by an imposed thermal and is usually "small". In a simple Drude model for particles with mass m, charge q , one degree of freedom per carrier, density n and scattering time $\tau$, the conductivity, $\sigma = nq^2\tau/m$, the Hall coefficient, $R_H = 1/nqc$,t he thermopower, $S_1 = k_B/q$, the magnetoresistance, $\Delta R = 0$ and the Nernst coefficient, $N = 0$. On the other hand if we imagine a system with two oppositely charged but otherwise identical carriers, $\sigma = 2nq^2\tau/m$, $S = 0$, $R_H = 0$, $N = k_B\tau/m = (k_B/q)(\omega_c\tau) = (S_1)(\omega_c\tau)$ and $\Delta R = (\omega_c\tau)^2$, where $\omega_c = qB/mc$. Since most systems are neither single carrier, single mass, single scattering time nor completely electron-hole symmetric we should expect all coefficients to be non-zero and of a magnitude given by the simple expressions above. Thus the Nernst coefficient should be of the order of $N \sim (S_1)(\omega_c\tau)$. For a metal $S_1 \sim (k_B/e)(T/T_F) \sim 20$nV/K at 0.2K for $(TMTSF)_2PF_6$ in the metallic state so that the Nernst coefficient at 7 tesla should be of order 50nV/K rather than the 10μV/K which is observed at that temperature. In figure 5 we show how the Nernst structure at the magic angles increases as temperature is lowered. A Boltzmann transport model based on either Osada's model[8] or the fitting scheme recently proposed[9] can generally reproduce the shape, but not the magnitude of the effect. We should also note that the structure around **c'** disappears when the sample is cooled into the FISDW state.

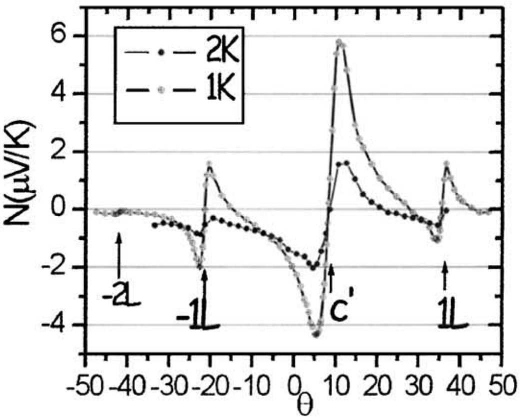

Figure 5. Angular dependence of the Nernst signal at 2K and at 1K showing the rapid growth with lower temperature.

The giant value .of the Nernst effect is not yet understood, It may result from extremely correlation narrowed bands or from something more exotic like interchain spinon drag.

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