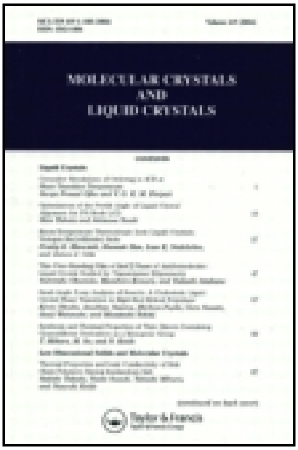

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Low-Temperature Electron Spin Resonance in $(TMTSF)_2PF_6$ in the High Pressure Metallic Phase

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LOW-TEMPERATURE ELECTRON SPIN RESONANCE IN $(TMTSF)_2PF_6$ IN THE HIGH PRESSURE METALLIC PHASE*

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The electron spin resonance of $(TMTSF)_2PF_6$ has been observed at low fields (H_o < 110 Oe) in the high pressure, metallic phase ($p \geq 6.5$ kbar) in the temperature range 1-4 K. The anisotropy in the g value is similar to that observed at ambient pressure above the metal-insulator transition. The linewidth is very narrow and the spin susceptibility strongly decreases as the superconducting transition is approached from above. We interpret this as evidence for singlet-paired superconductivity. Superconductivity is observed at 1.1 K and the critical field has angular dependence in the bc plane. These observations lead us to conclude that $(TMTSF)_2PF_6$ is a singlet paired superconductor.

INTRODUCTION

Since superconductivity was first discovered[1] in the organic metal $(TMTSF)_2PF_6$ the nature of the superconductivity has been an open question,

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particularly as to whether the pairing is singlet (s-wave) or triplet (p-wave). The properties of a p-wave coupled superconductor have been in the literature for some time.[2] As Balian and Werthamer showed, the proper p-wave state was not merely the spin-up and spin-down pairings ($S_z = \pm 1$) but the full triplet state including the non-magnetic state corresponding to $S_z = 0$. Unfortunately for the case of non-s-wave pairing, most of the experimentally observable properties of a superconductor are insensitive to the type of coupling. An exception is the behavior of the magnetic susceptibility and the Knight shift. For the case of s-wave pairing the presence of a gap at the Fermi level below the transition temperature, T_c, causes the spin susceptibility to decrease exponentially to zero below T_c. In the case of p-wave pairing the existence of the $S_z = \pm 1$ states assures that the susceptibility will remain finite below T_c. In fact, calculation shows[2] that the ratio of the superconducting to normal susceptibilities, $\frac{\chi^s}{\chi^n}$, should be 2/3 at temperatures well below T_c. Since the Knight shift of a nucleus in a metal is directly proportional to the spin susceptibility it can be a direct probe of $\frac{\chi^s}{\chi^n}$. Unfortunately, the weak coupling of the methyl protons in $(TMTSF)_2PF_6$ leads to a small Knight shift. We have therefore undertaken a study of the low field electron spin resonance in the region of T_c to directly probe $\frac{\chi^s}{\chi^n}$.

EXPERIMENTAL DETAILS

All the measurements reported here were performed with a continuous wave high frequency spectrometer[3] in the range 95-300 MHz. The spectrometer was tuned to be sensitive to absorption. The magnetic field H_o was provided by an Nb_3Sn superconducting split solenoid which was rotatable in one plane. Pressures up to 10 kbar were obtained by careful isobaric freezing[4] of the ^4He. The pressure bomb was immersed in liquid ^4He whose vapor pressure could be regulated. The magnetic resonance coil was wound directly on the sample with a rectangular cross section to maximize the filling factor. The sample was mounted with the highly conducting a axis perpendicular to the H_o plane so that the magnetic field could be rotated in the bc plane. Results were found to be independent of the temperature and pressure cycling history of a particular sample and there was no observed cracking of samples. The same setup could be used to measure the electron spin resonance, critical fields in the bc plane and T_c. The upper critical field, H_{c2}, was defined as the field at which rf absorption was first observed. The g value was measured by using a small sample of $Qn(TCNQ)_2$ outside the rf coil as a g marker.

RESULTS AND DISCUSSION

In Fig. 1 we show H_{c2} vs θ in the bc plane at a temperature ~57 mK below T_c. H_{c2} is highly anisotropic with a value of 15 Oe in the c direction and ~400 Oe along b (θ = 0 is the c direction). This anisotropy is consistent with other measurements and indicates the importance of knowing the field direction perpendicular to the a axis in this material. One should be cautious in interpretations of data taken in the bc plane if the field direction is unknown.

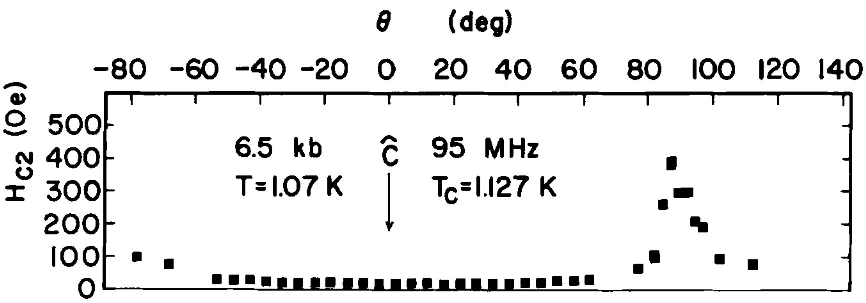

FIGURE 1 Angular dependence of the upper critical field in the bc plane 57 mK below the superconducting transition

In Fig. 2 is plotted the g value in the bc plane. The measurements agree very well with the Pedersen, et al.[6] results at X-band frequencies, ambient pressure and temperature just above the metal-insulator (MI) transition. This result indicates that the detailed nature of the metallic state is the same both near the MI transition at ambient pressure and near the superconducting state at high pressure.

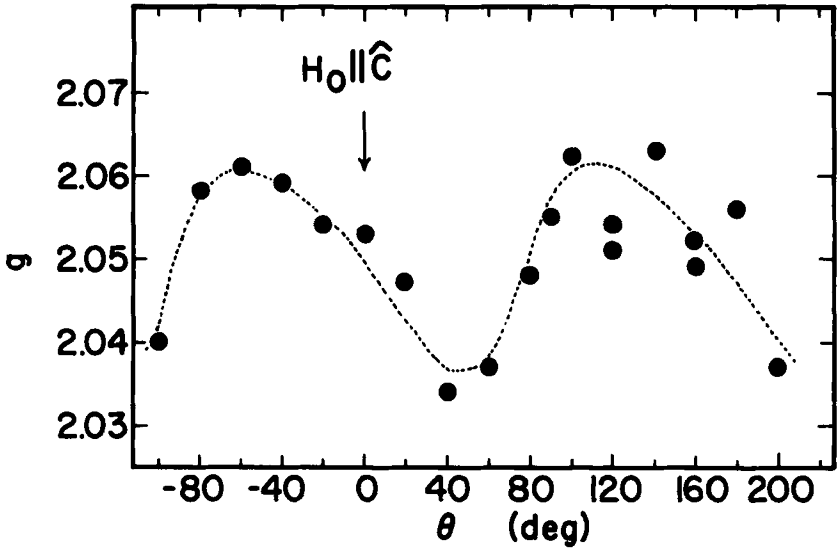

FIGURE 2 Anisotropy in the g value in the bc plane at T = 1.1 K, P = 6.9 kbar.

In Fig. 3 we show our principal result, the linewidth and absolute spin susceptibility vs temperature at a resonant frequency of 300 MHz (H_O = 105 Oe) and pressure of 6.9 kbar. The field is oriented parallel to the c axis. The absolute susceptibility was determined by using the Schumacher-Slichter method[7] with the methyl protons as the calibration.

It is important to note that the linewidth shows little broadening so that we can rule out the possibility of the onset of the MI transition as the linewidth is observed to increase strongly in that case.[6] The spin susceptibility is observed to decrease by 50% with the onset at 2 K. What is unusual is that the onset of the decrease in χ^s/χ^n is somewhat above T_c and the measuring field is above H_{c2} as can be seen by reference to Fig. 1.

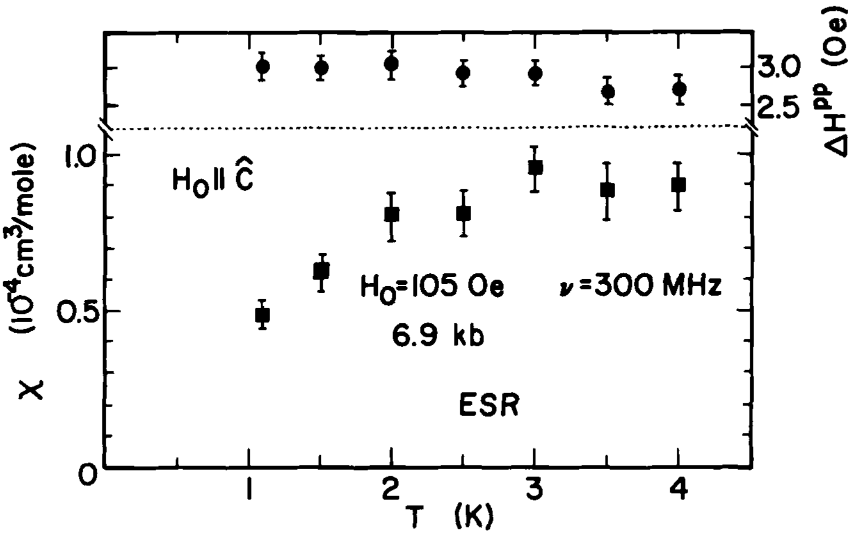

FIGURE 3 Linewidth and absolute spin susceptibility as a function of temperature at 6.9 kbar

These results suggest that there are strong fluctuations into the superconducting state above T_c. If the fluctuation interpretation is correct, then the observation that $\frac{\chi^s}{\chi^n}$ decreases by a factor of two as the superconducting transition is reached is evidence that $(TMTSF)_2PF_6$ is a singlet superconductor ($\frac{\chi^s}{\chi^n}$ for triplet pairing should be 2/3 at low temperature).[2] These effects have also been observed at low temperature at higher fields in $(TMTSF)_2ClO_4$.[8]

CONCLUSIONS

The magnetic properties of $(TMTSF)_2PF_6$ in the bc plane are highly anisotropic. We attribute the strong decrease in $\frac{\chi^s}{\chi^n}$ at low temperature due to fluctuations into the superconducting state and furthermore conclude that $(TMTSF)_2PF_6$ is a singlet superconductor.

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