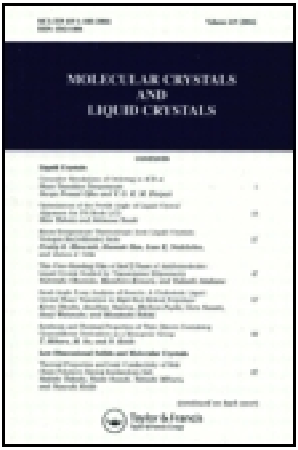

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

Magnetotransport and Nonlinear Effects In $(TMTSF)_2PF_6$

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Version of record first published: 14 Oct 2011.

Mol. Cryst. Liq. Cryst., 1982, Vol. 79, pp. 79–94
0026-8941/82/7901-0079$06.50/0

(Proceedings of the International Conference on Low-Dimensional Conductors, Boulder, Colorado, August 1981)

Magnetotransport and Nonlinear Effects in $(TMTSF)_2 PF_6$*

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Received for publication September 14, 1981

We have measured the magnetoresistance, Hall effect and the nonlinear conductivity characteristics in pure and radiation damaged $(TMTSF)_2 PF_6$. We find that the material below the metal-insulator transition temperature is best described as quasi-two-dimensional with a very high mobility (10^5 cm^2/volt-sec). The nonlinear conductivity is suppressed by the application of a magnetic field and by radiation damage. The magnetoresistance, metal-insulator transition temperature and the superconducting transition temperature are also reduced by radiation damage of order 1000 ppm for the latter and 100 ppm for the former.

A sizable fraction of this conference is devoted to the properties of the salts of TMTSF. The wide variety of the low temperature states of these materials is extroadinary[1-5]. We will concentrate in this paper on the spin density wave state (SDW) in $(TMTSF)_2 PF_6$ and in particular on its similarities and differences with the charge density wave (CDW) state previously found to be ubiquitous in these materials. We are particularly interested in the highly nonlinear conductivity[4,6] and

*Research at UCLA partially supported by NSF DMR 79-08560
Research at IBM partially supported by ONR.

whether it is similar to the depinned CDW motion[7] observed in the metal chalcogenides, or to the behavior seen in TTF-TCNQ[8]. In these studies we have performed magnetotransport measurements in the linear and the nonlinear regime and have studied the effect of radiation induced defects on both the transport properties and on the metal-insulator transition as well as the superconducting transition seen under pressure.

To sum up the main points that we wish to make: 1) The magnetotransport indicates that below the SDW transition $(TMTSF)_2PF_6$ is a quasi=two-dimensional conductor with a high mobility. 2) The nonlinear characteristics are quite different from those observed in $NbSe_3$ in the scaling with the frequency dependent conductivity, sensitivity to defects and magnetic fields, and absence of a threshold field. They are however, similar to what has been observed in TTF-TCNQ. 3) The effects of radiation damage are quite similar to what is observed in CDW systems[9] as far as the transition is concerned and the effects are those to be expected for a two dimensional band structure rather than in isolated chains[10] as far as the transport is concerned. 4) The superconducting state is easily destroyed by a small amount of damage, probably indicating that the defects are at least partially spin defects, rather than the presence of triplet superconductivity.

The original report of the nonlinear effects[4] suggested that the effects were associated with the depinning of a spin density wave in analogy with the phenomena found in the CDW system $NbSe_3$[7]. The low values of the electric field which raise the conductivity suggested that the pinning was weak, as might be expected for a SDW, but the main evidence for a SDW rather than a CDW transition came from the differences observed in static susceptibility[3] and ESR[4] measurements and more recently from the anisotropy of the static susceptibility and indication of a spin-flop transition[5]. Further investigations however indicated that the nonlinear effect is quite different from that observed in $NbSe_3$[6]. In fig. 1 we show the resistance of a sample of $(TMTSF)_2PF_6$ taken at 1.1K as a function of applied voltage which has been expressed as an electric field using the measured length of the sample. The measurement is taken with an AC resistance bridge in the presence of a DC bias voltage so that the dynamic resistance (dv/di) is recorded. The resistance characteristic is quite smooth, indicating the absence of any threshold behavior. The figure also shows a blow-up of the field region near zero where it can be seen that the behavior is non-ohmic down to fields below 0.1 mV/cm. For contrast we have also shown the resistance vs. field dependence for $NbSe_3$ taken on the same resistance bridge at 51K. For the latter material the conductivity is perfectly linear up until an onset field.

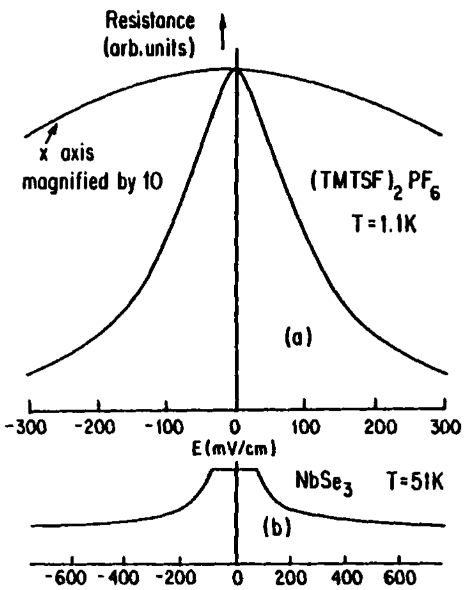

FIGURE 1 Dynamic resistance (dV/dI) as a function of electric field showing the absence (a) and presence (b) of a threshold field.

While it may be argued that we have not gone to sufficiently small fields to observe ohmic behavior, the characteristic field for depinning of a density wave should be related to the pinning force which is measured by the frequency dependence of the conductivity[11]. The characteristic frequency for $(TMTSF)_2PF_6$ is 2-3 orders of magnitude greater than that found in $NbSe_3$[6,12], whereas the threshold field for $NbSe_3$ is 2 orders of magnitude higher than our minimum field measurements for $(TMTSF)_2PF_6$. Previous measurements have revealed other differences between the bahavior of the two systems. There has been no observation of periodic "noise" associated with the nonlinear conductivity in $NbSe_3$, and the frequency dependence of the conductivity indicates a stronger pinning in the case of $(TMTSF)_2PF_6$ than would be expected for a SDW, although the frequency dependence is still at a lower characteristic frequency than can easily be explained in terms of single particle behavior involving excitations above a gap[12].

In order to better understand the ground state of $(TMTSF)_2PF_6$ with respect to both the linear and nonlinear transport we have investigated the magnetoresistance and Hall effect which are the classic measurements for analysing the conductivity. The magnetoresistance of $(TMTSF)_2PF_6$ has previously been measured by Jacobsen et al.[14] who found a large positive magnetoresistance in the metallic state, which increased considerably below the metal-insulator transition. Our measurements of the resistance in the absence of a magnetic field and in 80kGauss is shown in fig. 2 for the field aligned approximately parallel to the c axis, the direction of lowest conductivity. These results are quite similar to those reported in ref.[13]. However, their measurements indicated that the magnetoresistance was nearly isotropic in the b-c plane. To the contrary we find that the magnetoresistance along b is too small to measure. (In this discussion we will not be concerned with a rigorous alignment with the crystallographic axis, rather when we refer to the b axis we mean the axis along which the measured magnetoresistance is a minimum. This is within about 5 degrees of parallel to the crystal platelet face which from X-ray analysis is within about 5 degrees of the crystallographic b axis. When we refer to the c axis we mean the axis 90 degrees rotated from the b axis as we have defined it above. We take the b-c plane as being perpendicular to the needle axis of the crystal.) This is illustrated in fig. 2 where we show the resistance change as a function of magnetic field for low fields at 4.2K. The data were taken in a rotatable NMR magnet with the samples aligned so that their needle axis was along the axis of rotation. The magnet was rotated until the observed resistance change was minimum. A magnetic field sweep was then done, the magnet was rotated 90 degrees and another sweep taken. At low fields the ratio of the magnetoresistance in these two directions was between 200 and 1000. For high field data samples were mounted so that the platelet faces were either parallel or perpendicular to the field of a superconducting magnet. Although the alignment in this case is critical we have observed anisotropies at 80kG of up to 200. Similarly we find an anisotropy of greater than 200 between the a and c axis'.

There are several points to be made about the magnetoresistance before we procede in our analysis. The fact that the magnetoresistance is anisotropic in the plane perpendicular to the highly conducting axis implies that all measurements which claim to measure the conductivity or transfer integral anisotropy from measurements of the critical field in the directions parallel and perpendicular

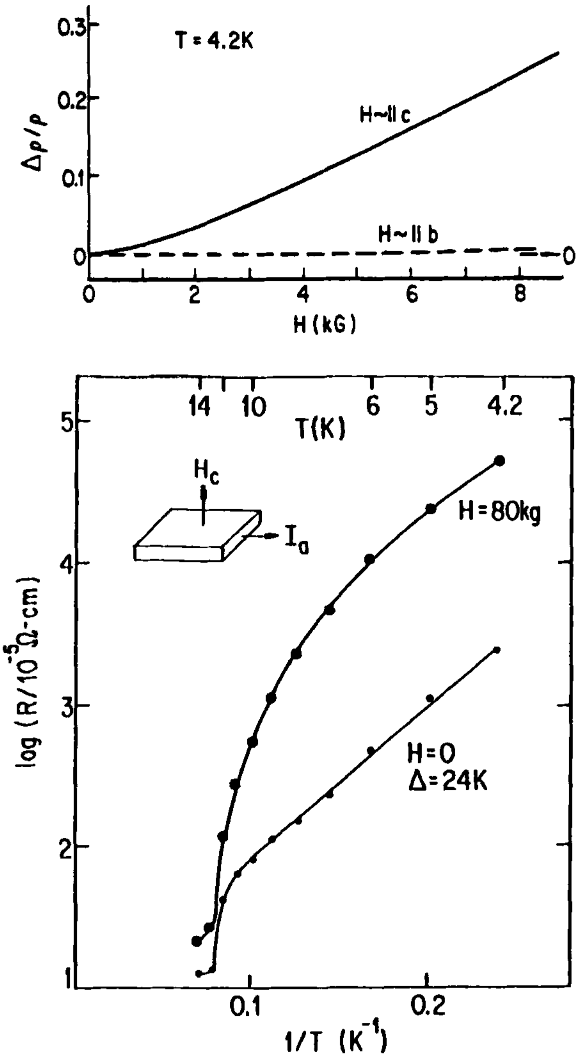

FIGURE 2a) Magnetoresistance with the field aligned along the b and c axis. b)Temperature dependence of the resistivity with and without an applied field.

to the highly conducting axis are unreliable in their conclusions about the dimensionality of the transport processes. In general, any orbital phenomena associated with the magnetic field can be expected to differ significantly in the plane perpendicular to the a axis for the compounds in this family. This is certainly the case for the critical field of the ClO_4 salt below its superconducting transition where the b-c axis critical field anisotropy is greater than the a-b axis anisotropy[14].

The large positive magnetoresistance observed in $(TMTSF)_2PF_6$ is quite different from the behavior seen in CDW organic conductors such as TTF-TCNQ where a very small negative magnetoresistance was observed[15]. This is related to the fact that in the present system the presence of an 80kG magnetic field has virtually no effect on the transition temperature (as measured resistively it shifts by less than 0.1K) indicating that the change in susceptibility through the transition is very small so that the magnetic field does not favor one state over the other. For TTF-TCNQ there is a small shift since the CDW has a sharp drop in its spin susceptibility at T_{M-I} while the (high field) susceptibility of the SDW is essentially the same as the metallic state[3,5]. A large positive magnetoresistance has also been reported for HMTSF-TCNQ[16] and for TTT_2-I_3[17]. In the former case the results are similar in origin to those that we report here. For the latter material the system is disordered and the effect is completely isotropic and thus a spin effect.

Below T_{M-I} where the material is semiconducting we expect that a two carrier description of the transport may be a good first attempt. The high field limit for the magnetoresistance is then given by

$$(R(H)-R(0))/R(0)=u_e u_h H^2/c^2 \qquad (1)$$

where u_e and u_h are the electron and hole mobilities. If we take this equation at face value, then the magnitude of the effects that we observe at 4.2K correspond to a characteristic mobility of 10^4 cm^2/volt-sec which is similar to what is observed in the purist small effective mass conventional semiconductors such as InSb[18] or in the semi-metal HMTSF-TCNQ[16]. Assuming that the anisotropies are the same for the electron and the hole bands (since before the opening of the SDW gap they belonged to the same band) we can use equation 1 to find the b-c effective mass anisotropy.

$$m_{Hb} = (m_a^* m_c^*)^{1/2} \qquad (2)$$

$$(R(H)-R(0))_c/(R(H)-R(0))_b = m_b^*/m_c^* \quad (3)$$

Where m_H is the cyclotron mass and the other masses are effective masses from the band structure. The measured anisotropy in the masses from the optical plasma frequency measurements is only 10 while the magnetoresistance would yield a value of 200-1000. The magnetoresistance with H along b is therefore well below what would be expected from band motion and we take this as evidence for diffusive motion rather than band motion in the c direction . The high value of the magnetoresistance with H in the c direction implies that electrons experience coherent motions and closed orbits in the a-b plane. Again note that from equations 2 and 3 it would be necessary to measure the anisotropic magnetoresistance along the b and a directions in order to obtain information about the mass ratio between the two highest conducting directions. Since both are extremely small and involve closing the orbits through the diffusive c direction this comparison is not possible. Almost certainly, previous anisotropy measurements from magnetotransport in most of these one dimensional conductors merely involved some component of the magnetic field being non-zero along a plane where orbits could be established and the anisotropy measured was therefore more a function of the crystal alignment than any property of the materials.

A more conventional determination of the mobility may be obtained from Hall effect measurements. Using a standard five probe technique with the magnetic field aligned parallel to the b axis to reduce the magnetoresistance, we measured the Hall coefficient from 10K t0 2K[20]. At 2K the number of carriers which we would calculate in a simple one band model is 10^{15}. The Hall mobility (u_H) is the quantity we are really after and it is shown in fig. 3 as a function of temperature. Defined as the product of the Hall coefficient (R_H) and the conductivity ( ) it is given in a two band model by:

$$u_H=R_H =u_h-u_e \quad (4)$$

The sign of the Hall coefficient is positive indicating hole carriers dominate. If we take equations 4 and 5 literally then we find mobilities of 1.2×10^5 for the holes and 0.24×10^5 for the electrons.

The mean free path for these carriers depends on their effective mass. If we take the effective mass as that of free electrons (which surprizingly is the appropriate value from a number of measurements in the metallic state),[13,19] then we find a mean free path of 5000 Angstroms. However, we would expect that the opening of a gap at the Fermi surface would increase the curvature of the bands and produce a smaller effective mass. In either a tight binding or a free electron model the pretransitional mass is reduced by a factor of the bandwidth devided by the energy gap. For $(TMTSF)_2 PF_6$ this amounts to a factor of 200. With a mass that is 1/200 of an electron mass, we would only need a mean free path of 300 Angstroms to explain the mobility. The small effective mass at the bottom of the band edges may also help explain some of the temperature dependence of the mobility. At low temperature only the small mass states are occupied, while at higher temperatures the electrons sense the much heavier mass states.

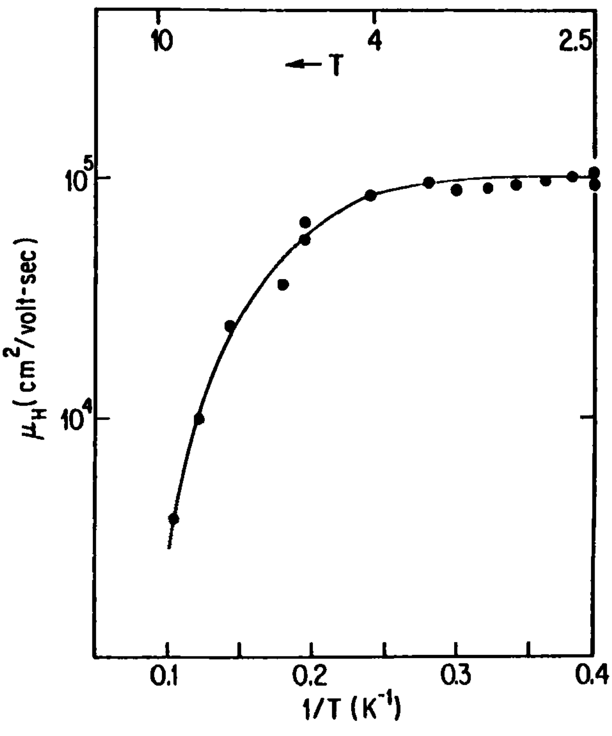

FIGURE 3 Temperature dependence of the Hall mobility.

The small mass below the SDW transition would not help in the explanation of the reasonably large magnetoresistance seen above the transition in the metallic state, nor the magnetoresistance observed in the metallic state under pressure which is comparable to what we observe in the SDW state[21]. However, it should be noted that in the present case, where there is no question of fluctuations into the superconducting state, the magnetoresistance is large and positive. It is therefore possible to observe such effects resulting entirely from normal band transport in systems with high mobility.

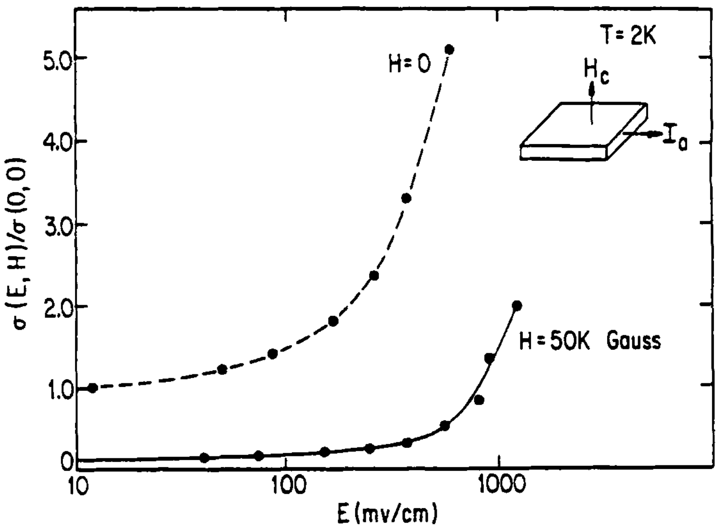

FIGURE 4 Nonlinear conductivity with and without a transverse magnetic field.

We now turn our attention toward the nonlinear characteristic and its magnetic field. Recent work on the CDW system $NbSe_3$ have shown that the nonohmic conductivity is not effected by the application of a magnetic field[22]. This is due to the highly one dimensional motion of the depinned CDW. If the depinned SDW is like the depinned CDW in that both propagate in only one direction, we would expect that the conductivity in the presence of both an electric and a magnetic field could be written as the sum of two contributions:

$$\sigma(E,H)=\sigma_{sp}(H)+\sigma_{SDW}(E) \tag{5}$$

The first term on the right refers to the transport due to the normal carriers which we have seen is highly dependent on the magnetic field but which in this model we take as independent of the electric field. The second term is the contribution of the depinned SDW which should be highly E sensitive but independent of H since its motion is presumably one dimensional. If this equation is correct then the effect of applying a magnetic field would be to shift the nonohmic characteristic down by a constant amount.

In fig. 4 we show the results of the experiment in which a magnetic field is aligned along the direction with the maximum magnetoresistance. It is clear that the magnetic field strongly suppresses the nonlinear contribution. Rather than exhibiting the behavior suggested by equation 6, the nonlinear contribution to the conductivity shows much the same reduction as does the normal contribution. This would suggest that the additional carriers have similar mobility and dimensionality to the normal carriers. In this regard we point out that one of the prerequisites for "hot electron effects" is the existence of a very high mobility carriers[23]. It is unlikely that carrier heating effects could account for the entire nonlinear characteristic, since the restoration of the metallic conductivity would require exciting all of the carriers over the gap (that is, it would require destroying the gap). It should be noted that the characteristic shown in fig.4 does not go to sufficiently high electric field, for this sample, to restore the metallic conductivity. Therefore it is possible that the low field effects are related to carrier heating and that other effects set in at higher electric fields. The fact that the nonlinear characteristics are so smooth on the other hand, suggests that this is not the case.

Radiation Damage

The radiation damage study was undertaken in order to compare the effect of defects on the SDW transtion as compared to the previous measurements on CDW systems, to see the effect on superconductivity, to test our interpretation of the high mobility and two-dimensionality seen in the magnetotransport study, and to see whether the increased pinning would effect the nonlinear conductivity in the same way as it does in $NbSe_3$.

The radiation damage was performed at the Van der Graaf accelerator at Cal State L.A. using a beam of 2.5 MeV protons. As is usual in radiation damage experiments, the homogeniety of the damage and the relative amount of damage between samples is known quite well, but the absolute value of the damage is often difficult to evaluate. We found that using the same dosages that produced neglible effect on both the transitions, transport and nonlinear effects in $NbSe_3$[24] we observed enormous effects in the $(TMTSF)_2PF_6$. This supports the conclusion reached by the group at Fontenay-aux-Rose[10] who have found that whereas Rutherford scattering is the main source of damage in the metal chalcogenides, where the electronic excitiations caused by the irradiation do not contribute to the damage, in the conducting organic charge transfer salts the electronic excitations do contribute[25]. In the case of $(TMTSF)_2PF_6$ the damage by direct scattering from the nuclei contributes only 1/300 of the damage caused by the electronic excitations.

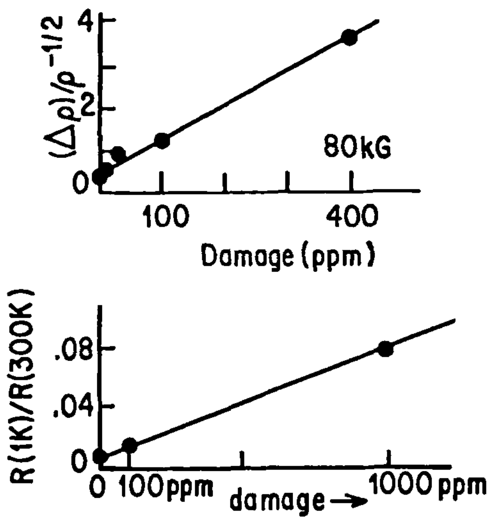

FIGURE 5 Damage fraction dependence of (a) the magnetoresistance at 1bar and (b) the resistance ratio at a pressure of 11kbar.

To calculate the amount of damage we compute the total energy loss of the incident protons in passing through the crystal and divide this by the approximate energy per dislocation as reported in ref. 25 for TMTSF-DMTCNQ (7 KeV). The result of this computation is the relationship:

$$c=1.4\times10^{-17}\times(\mathrm{protons/cm^2}) \qquad (6)$$

In order to verify these values we have also performed in situ resistance measurements during the irradiation. We find that the resistance increases by a factor of 1.3 with a calculated damage fraction of 600 ppm. We can independently obtain a value for the damage fraction (c) from the observed increase in resistance and the previously measured anisotropy. If we take the conductivity anisotropy between the two must highly conducting directions as 400[1] for $(TMTSF)_2 PF_6$ then the observed resistance increase would correspond to a damage fraction of 750 ppm. The agreement between the two values is quite satisfactory. We will therefore use equation 6 whenever we quote a value for the amount of damage in the remainder of the text.

We have measured the magnetoresistance in the SDW state as a function of the amount of damage with the basic results at 4.4K shown in fig.5. The magnetoresistance decreases drastically with the addition of only 400 ppm defects. If we assume that the main effect of the defects is to increase the scattering rate, as we would find in conventional three dimensional metals or semiconductors, then the scattering rate should be proportional to the number of defects. According to equation 1, the scattering rate (which goes as the inverse of the mobility) should be proportional to the inverse square root of the measured magnetoresistance. This is the quantity plotted in fig. 5 and indeed a linear dependence is found. Note furthermore that the value of damage which doubles the low temperature scattering rate is 100 ppm. This implies that pure material is very pure with intrinsic damage of about 100 ppm. At higher values of damage the magnetoresistance is too small to measure accurately, but there appears to be a small negative magnetoresistance at 1000 ppm.

If the additivity of scattering rates is correct then this should also show up in the resistivity ratio. Unfortunately we find many resistance jumps during cooldown for samples at ambient pressure. However the experiment is easily performed under pressure with the results shown in fig.5b. Again the scattering rate obeys Matthiessen's rule and the damage which corresponds to a doubling of the rate is 100 ppm.

The question of the applicability of Matthiessen's rule for the case of quasi-one-dimensional conductors has been treated at some length by a number of authors[10,27]. We would merely like to point out that the crossover between one dimensional and two or three dimensional behavior must occur when the time it takes an electron to coherently transfer between chains becomes shorter than the time between encounters of the defects which interupt the on chain motion. For our samples at 4.2K the scattering is largely dominated by the defects so that the condition for one dimensional behavior is that the number of unit cells between defects is equal to the anisotropy in the transfer integrals. In order to be one dimensional we would therefore need something like 3-10% defects.

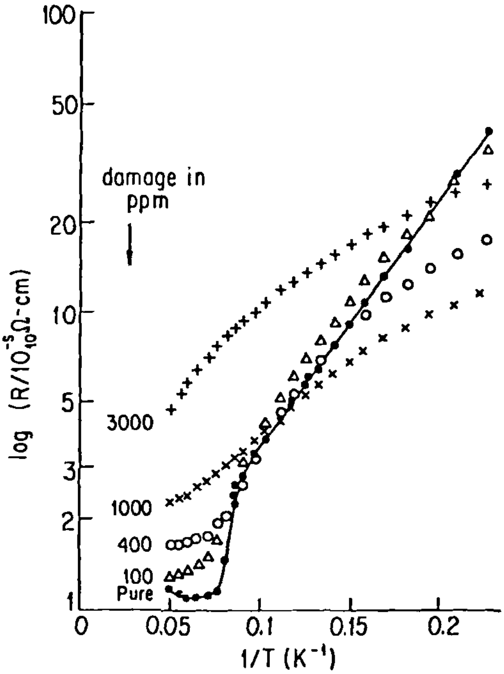

FIGURE 6 Temperature dependence of the resistivity of pure and radiation damaged samples of $(TMTSF)_2PF_6$.

In fig. 6 we show the effects of the damage on the SDW transition. The absolute values of the resistance in this figure are to be ignored. They are scaled with one another at 20K, assuming an extension of Matthiessen's rule, in the absence of data on samples with no resistive jumps in the cooldown process. What we wish to emphasize is the temperature dependences. The main efects that we observe are a raising of the temperature of the resistivity minimum, a smearing of the metal-insulator transition, a lowering of the transition temperature, and a less activated behavior to the conductivity below the transition. All of these effects have previously been seen in CDW systems and particularly in TTF-TCNQ[9]. While the first approach to the cause of these effects might be to evaluate the pairbreaking effects of the defects on the mean field transition temperature[27].

However, the broadening of the transition coupled with many previous observations suggests that the primary effect of the defects is to pin the phase of the density wave on each particular chain so that the interchain interactions are decreased and the three dimensional ordering is suppressed. Since the SDW state has no charge density variation to couple with the defect potential it had been expected that the effect of the radiation induced defects would be considerably less important for the $(TMTSF)_2PF_6$ than for systems such as TTF-TCNQ. On the contrary we find that the rate of the depression of the T_{M-I} for $(TMTSF)_2PF_6$ is dT_{M-I}/dc = 60K/%defect whereas for TTF-TCNQ the measured value is dT_{M-I}/dc = 150K/%defects[9]. The two numbers are comparable. While we expect that a good fraction of the defects created are spin defects we would still expect less exchange coupling energy between defect and SDW than potential electrostatic coupling energy between a defect and a CDW.

An evaluation of the number of spin defects and their exchange coupling is also required in order to understand the destruction of the superconducting state with irradiation. We have found that the PF_6 salt is no longer superconducting down to 20mK under 11kbar pressure when it has been damaged to 100 ppm or above. Similarly the ClO_4 salt is not superconducting at ambient pressure down to 20mK with a dosage of 100 ppm. The destruction of the superconductivity by such a low level of predominantly potential scattering sites might be taken as evidence for triplet superconductivity. However, we have recently found that the critical field of the ClO_4 salt is Pauli limited so that the superconductivity must be the usual singlet variety[14].

A quite cavalier approach would suggest that since the SDW state at 12K is supressed by 1000 ppm of defects, through their exchange coupling with the conduction electrons, 100 ppm should suppress the superconductivity at 1.2K, again a result of the exchange coupling. The problem is that for the SDW the effect of the exchange coupling is to pin the SDW on each chain. The effect we consider on the superconductivity is a mean field pairbreaking effect. Nonetheless, both effects do serve as a measure of the exchange coupling.

Finally we note that if the defects pin the SDW's and smear the transition then they should also act to pin the

SDW's against displacement by an electric field and this should decrease the nonohmic conductivity. In the case of $NbSe_3$ the defects also increased the threshold field[24]. We have found [25] that the defects in $(TMTSF)_2 PF_6$ reduce the nonlinear effects but do not produce a threshold field, the conductivity is still slightly nonohmic at all fields. Although this may be related to a pinning phenomena (with a distribution of pinning strengths), it is also consistent with an interpretation in terms of "hot electron effects" which would decrease as the mobility decreased.

In conclusion the magnetotransport and the radiation damage studies indicate that $(TMTSF)_2 PF_6$ is a two dimensional conductor with a high mobility at low temperature. The nonlinear effects are quite different than what one observes in $NbSe_3$ in that they are suppressed strongly by a magnetic field, there is no threshold field and the frequency and electric field effects do not scale. The radiation damage smears and lowers the transition temperature of the SDW system in much the same way as it does the CDW systems. Superconductivity under pressure is suppressed by the addition of very few defects (100 ppm), but the defects are at least partially spin defects with an exchange coupling to the conduction electrons which also shows up in the effects of the SDW at ambient pressure.

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