Magnetothermopower of tetramethyltetrathiofulvalenium phosphorus hexaflouride [$(TMTSF)_2PF_6$]

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We have studied the thermoelectric power and the resistance of $(TMTSF)_2PF_6$ as a function of temperature and magnetic field in the region of the spin-density-wave (SDW) metal-insulator transition. We observe slight changes above T_{SDW}. Below T_{SDW} the anomalous thermopower observed in the absence of a magnetic field is suppressed and the material behaves more as a conventional semiconductor. The effect of the magnetic field is in the orbital motion of the carriers as the significant changes are observed only when the magnetic field is aligned along the c axis. These results are compared with previous magnetotransport measurements in this family of organic conductors.

INTRODUCTION

The organic charge-transfer salts based on the donor molecule TMTSF form one of the most physically interesting families yet studied.[1] Although all of these salts have the same charge transfer and band filling, as well as virtually identical anisotropic band structures,[2] the ground states which have been found range from metallic to spin-density wave[3] (SDW) and anion order-induced charge-density-wave (CDW) insulators.[4] Additionally, the first organic superconductor was discovered in the PF_6 salt under a pressure of ~ 8 kbar.[5] There are also a number of transitions which involve the freezing of the rotational motion of the anion complexes,[6] in some cases into an ordered structure with a particular wave vector and in other cases into a disordered state.[4,7,8] The general phase diagram for many of these $(TMTSF)_2X$ compounds involves a high-temperature metallic phase at ambient pressure, followed at lower temperature by a metal-insulator transition into a SDW or a CDW state usually driven by an anion ordering transition.[9] Application of a moderate pressure ~ 10 kbar suppresses the metal-insulator transition and the metallic state remains stable down to ~ 1 K at which temperature the samples undergo a transition to the superconducting state. The great similarity of the electronic structure (and the electron-phonon interaction) in these salts is attested to by the observation that the superconducting transition temperatures are all 1.2 K (± 0.2 K) at the pressure which just corresponds to the suppression of the insulating phase. (With the exception of the FSO_3 salt which has an anion possessing a dipole moment and a $T_c \sim 2$–3 K[4].)

Band-structure calculations[2] indicate that, in the metallic state, the Fermi surface should consist of two warped planes approximately parallel to each other and with warpage considerably less than their separation. The bandwidths are $4t_a \sim 1$ eV, $4t_b \sim 0.1$ eV, and $4t_c < 0.01$ eV. Thus, there are no closed orbits expected for the tight-binding-like bands. In contrast to what would be expected from the band structure, the magnetotransport properties of these compounds is quite remarkable. The first observation of the magnetoresistance in the PF_6 (Ref. 10) salt indicated a large nonsaturating positive magnetoresistance corresponding to a doubling of the resistance in a field of 60 kG, while the sample remained in the metallic state at temperatures above 12 K. In the insulating phase below 12 K, the magnetoresistance increased still further but in a way that might be expected for a semiconductor.

The high anisotropy in the band parameters would indicate that the magnetotransport coefficients should also be highly anisotropic. The observed properties are consistent with this idea and the remarkable effects in high magnetic fields are seen with the magnetic field oriented along the c axis corresponding to orbital motion in the a-b plane which contains the highest conducting directions.[11] One of the most striking observations was that the PF_6 salt exhibited Schubnikov-deHaas oscillations[12] at low temperatures and under sufficient pressure that it remained in the metallic and presumably open-orbit state.

The magnetic field dependent properties of the ClO_4 salt have been more extensively studied, largely in relation to its superconductivity. $(TMTSF)_2ClO_4$ and $(BEDT\text{-}TTF)_2I_3$ are the only organic compounds to date which are superconducting at ambient pressure.[13] When the samples are cooled slowly the metallic state is stable to low temperature and there is no density wave transition.[8] Again the metallic state should consist of only open orbits

on the Fermi surface, and yet quantum oscillations are observed in the resistance[14] and the thermopower[15] at temperatures below ~ 2 K. At higher temperatures a magnetic field directed along the c^* axis has also been shown to have strong effects on the specific heat,[8] thermal conductivity,[16] resistance,[14] and thermopower.[15]

The object of the present paper is twofold. It has often been suggested that the ClO_4 salt is very similar to the PF_6 salt under a pressure which stabilizes its metallic state and that even at ambient pressure the two are similar above the metal-insulator transition in PF_6. We will explore resistance and thermopower in this regime for the two salts as a function of magnetic field. We find that magnetic field effects are large but different in the two salts. Secondly, the temperature dependence of the thermopower of the PF_6 salt is quite anomalous in the region below the metal-insulator transition. Instead of showing a monotonic semiconducting-like thermopower, the reported thermopower increases to a positive maximum and then decreases and crosses zero as the temperature is lowered. The thermopower is very magnetic field dependent in this regime and in 80 kG the thermopower becomes monotonic and increases negatively as $1/T$.

EXPERIMENTAL RESULTS

The experiments reported below were performed in an apparatus consisting of a brass sample holder connected to the bottom of a ^{3}He pot. The holder and pot were situated in an indium-sealed stainless-steel vacuum can so that the temperature could be varied from room temperature to ~ 0.5 K. Sample leads were heat sunk to the vacuum-can top and then to the sample holder. Since the thermopower measurement requires the establishment of a temperature gradient, it was necessary to have the samples surrounded by vacuum. In order to prevent the samples from equilibrating to a temperature other than that of the sample holder, a split copper can was placed around the sample holder and screwed to the ^{3}He pot. The use of this arrangement provided ample temperature uniformity, while allowing for a moderately rapid cycling of the magnetic field without producing a large amount of eddy current heating.

Temperatures were measured with a germanium resistance thermometer and a carbon glass thermometer which were also used to calibrate a capacitance thermometer used to control the temperature during magnetic field sweeps.

Contacts were made to the sample with silver paint. Leads were 1 mil or $\frac{1}{2}$ mil Au wires. Resistance measurements were four probe ac (~ 103 Hz) and checked ocassionally by dc. Although there was considerable sample cracking, as observed by resistance jumps, during the cooldown[17] the resistance showed no such jumps below 60 K. The thermopower was measured using the technique of Ref. 18 with the following adaptations: the quartz blocks were replaced by thin saphire slides sandwiching a thin heater wire to reduce the thermal cycling time to ~ 10 s; the data were taken by continually applying an offset square wave to one heater; and the voltage from the differential thermocouple and the sample were plotted continuously on the Y axis of two separate x-y plotters with the X axis being either the output of a temperature sensor or the output of a current sensor measuring the magnetic field. The peak-to-peak change in the thermocouple and sample voltages then supply the necessary information for the calculation of the thermopower. It is necessary to observe the relative phase of the motion of the two pens to determine a sign change in the thermopower. We find that these modifications lead to considerably more sensitive measurements of small changes as well as allowing a quasicontinuous monitoring of the thermopower as a function of either temperature or magnetic field.

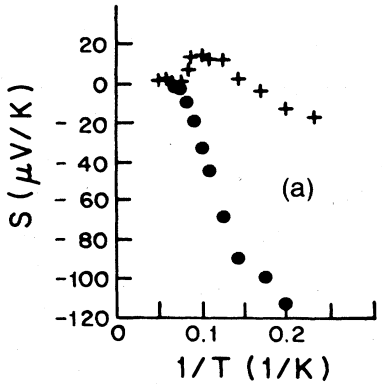
The magnetic field was generated with a superconducting magnet with a quench field of 106 kG at 1.5 K. The samples were aligned with their axes at particular orientations to the field by mounting them in different configurations at room temperature. The alignment is nominal but probably accurate to $\sim 5°$ since adjustments were made visually using the needle axis as parallel to a, and the plane of the platelet as perpendicular to $c \sim c^*$.

The temperature dependence of the thermopower is shown in Fig. 1. The results for zero field are qualitatively consistent with previous studies.[1,19] At temperatures above the SDW transition the thermopower is small and positive as expected from the band filling and bandwidth. The deviations from linearity in the metallic region have been associated with fluctuations preceding the SDW transition.[19] The transition itself is marked by the sharp positive rise of the thermopower at 12.5 K. The thermopower then increases positively, reaches a maximum, and then becomes increasingly negative as the sample is cooled. As yet, there is no conclusive interpretation of the thermopower below T_{SDW}.

The temperature dependence of the thermopower in an applied field of 80 kG along the c^* direction is also shown in Fig. 1. There are striking differences in comparing this curve with that obtained in zero applied magnetic field. The transition is not evident and the data appears as one continuous curve. There is no hint of the positive thermopower region seen below T_{SDW} in the zero field results. When the data is plotted as S versus $1/T$ [Fig. 1(a)], the thermopower in 80 kG shows the characteristic semiconducting behavior, $S \sim E_G/2k_BT + B$. The gap obtained from the slope of this curve is 35 K, similar to that observed in the zero-field resistance.

The presence of a magnetic field clearly effects the thermopower above the transition as well as below. The metallic state thermopower shifts to a more negative value and the difference with the zero-field result increases as the transition is approached. The drop in the $H=0$ thermopower near T_{SDW} is still present in $H=80$ kG. Shown for comparison in this figure are curves of the thermopower for $(TMTSF)_2ClO_4$ in the same temperature range and for the same magnetic fields. Note that the ClO_4 salt has a thermopower that increases positively in the presence of a magnetic field,[20] opposite to the effect in the PF_6 salt.

The magnetic field dependence of the thermopower is highly anisotropic. In Fig. 2 we show the thermopower as a function of temperature for 0 and 80 kG for a sample which aligned so that its needle (a) axis was parallel to



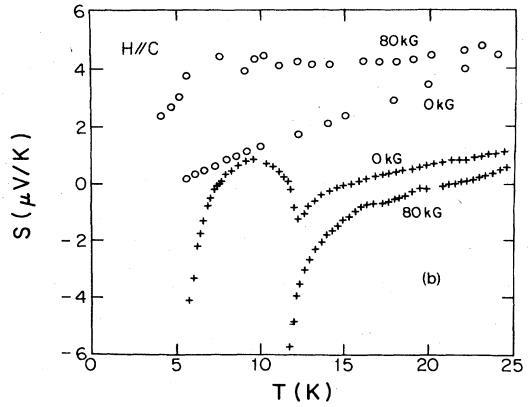


FIG. 1. (a) Thermopower vs $1/T$ for the ClO_4 salt; dots are $H=80$ kG, crosses are $H=0$. (b) Comparison of the magnetic field dependence of the PF_6 (crosses) and ClO_4 (open circles) salts thermopower.

the magnetic field. Aside from a slight shift, the high-field and zero-field curves have the same features.

The orientation which corresponds to the large changes in thermopower also corresponds to large magnetoresistance. Since it has already been established that the magnetoresistance is largest along the c^* direction and since we find comparable effects both in magnitude and in field

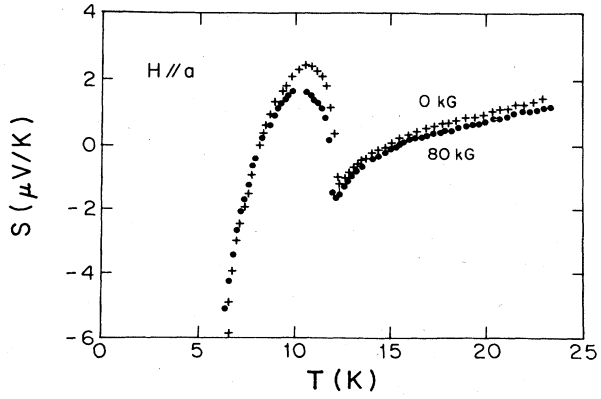


FIG. 2. Thermopower for field applied along the a axis.

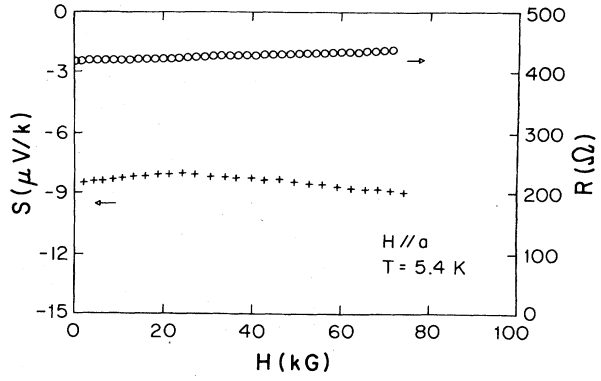


FIG. 3. Thermopower and resistance vs magnetic field along the a axis.

dependence to previous results, the alignment of the c^* axis sample is readily verified. The a axis sample shows little magnetic field dependence in either study (Fig. 3). The small effects observed, $\Delta R \sim 5\%$, probably correspond to a disalignment of $\sim 0.3°$.

In Fig. 4 we show a more detailed study of the thermopower of another crystal of $(TMTSF)_2PF_6$ using the technique of Ref. 18. The strong negative increase in thermopower at low fields and the saturation at high fields are evident in this data as is the increase in the characteristic field as the temperature is increased.

A more meaningful presentation of the magnetoresistance data is shown in Fig. 5. The Kohler plots [$\ln(\Delta\rho/\rho_0)$ versus H/ρ_0] are taken from the temperature dependence of the resistance in zero field and in 80 kG, as well as from fixed temperature sweeps of the magnetic field. Also shown for comparison are similar data obtained from the $\mathbf{H}||\hat{\mathbf{c}}^*$ magnetoresistance of $(TMTSF)_2ClO_4$.

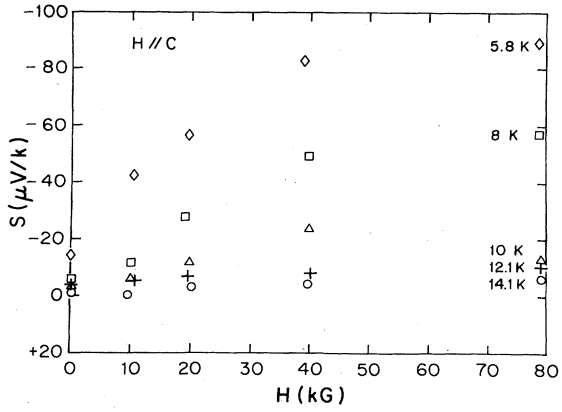


FIG. 4. Magnetic field dependence of the thermopower of $(TMTSF)_2PF_6$ for several temperatures.

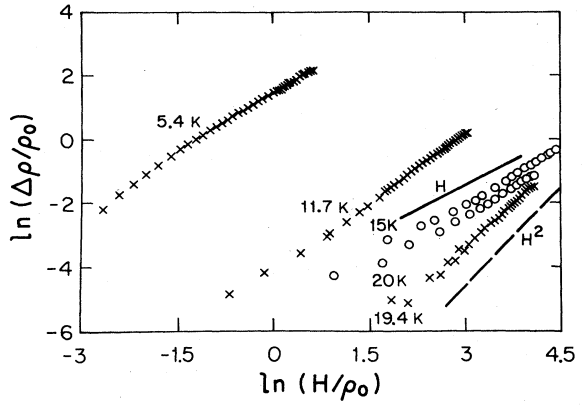


FIG. 5. Kohler plot of the magnetoresistance of the PF_6 salt (crosses) and the ClO_4 salt (open circles).

COMPARISON WITH PREVIOUS TRANSPORT DATA

There have been several previous studies of the magnetotransport properties of both the PF_6 and the ClO_4 salts of TMTSF.[9–12,14,15,21] For $(TMTSF)_2PF_6$ the transport coefficients measured include the conductivity along a, b, and c axes;[10] the magnetoresistance with the field along the principle axes and the current along a;[10,11] the Hall coefficient;[11] and the thermopower in zero field along the a and the b axes.[19] For $(TMTSF)_2ClO_4$ the x axis conductivity and thermopower and their magnetic field dependences have been investigated. Some work on the anisotropic conductivity has also been reported.

The magnetoresistance and zero-field thermopower reported in this paper are qualitatively consistent with previous data. The absolute value of the magnetoresistance with the field along c^* is very sensitive to the scattering rates and therefore varies appreciably (up to a factor of 4 at low temperatures) even in nominally pure samples. The general aspects are quite similar and involve a $\Delta\rho/\rho \sim 1$ at 80 kG in the metallic phase below about 30 K, with a good fit to an H^2 behavior, and a much larger nonsaturating field dependence with $\Delta\rho(80\text{ kG})/\rho \rangle 10$ and a lower power in H at high fields for the semiconducting phase.

The zero-field thermopower is the same in the metallic state (to within $\sim 20\%$ for results from different groups, samples, and techniques), but there seems to be a larger variation in the absolute value of the positive anomaly directly below the metal-insulator transition temperature. This suggests that the anomaly may also be impurity dependent.

Since it has often been suggested that the ClO_4 salt is similar to the PF_6 salt in the metallic phase, from the Kohler plots shown in Fig. 5 the field dependences are different with a H^2 behavior for the PF_6 salt and a H dependence for the ClO_4 salt. The differences in transport are also seen in Fig. 1 anticipating the lower-temperature behaviors.

Another of the surprising features of the magnetotransport has already been noted in Ref. 22. The Hall coefficient of the PF_6 salt in the semiconducting phase has been measured as large and positive for temperatures down to 2 K. The zero-field thermopower becomes negative below ~ 8 K. However, the thermopower measured along the b axis remains positive at all temperatures.[19] Thus the sign of the carriers is ambiguous in any simple model.

DISCUSSION

The unusually large magnetoresistance of the TMTSF salts,[9] especially in the metallic state, is a matter of considerable interest. We review briefly previous interpretations. The magnitude of the magnetoresistance was so large in these salts that it suggested to some researchers that the origin was in the quenching of superconducting fluctuations.[9] The initial interpretation of the magnetoresistance in the PF_6 salt suggested that there might be anomalous scattering from pieces of the Fermi surface which were almost nested.[10,20]

However, the magnetoresistance of the ClO_4 salt attains values of $\Delta\rho/\rho > 10$. Extensions of the above model suggest that such large values are possible if the scattering is extremely strong in very small regions of the open orbit. In such a case the magnetoresistance does not saturate until $w_c\tau_{\text{anomalous}} \sim 1$, where $\tau_{\text{anomalous}}$ is the scattering time in the region of the anomalously large scattering. Moreover, the field dependence of the magnetoresistance for this mechanism has a large linear region ($\Delta\rho/\rho \sim H$) and hence may be particularly appropriate for the ClO_4 salt. Although this is a viable explanation there is a more direct one. For the TMTSF salts, the a and b axes are not orthogonal. Therefore the a axis conductivity has a component along the direction of the open orbit. In this case the a axis resistance becomes a nonsaturating function of H and is related to the b axis magnetoresistance by

$$R_a = R_b \cos^2\theta \,, \tag{1}$$

where θ is the angle between the current direction and the open-orbit direction.[23,24] Recent experiments on the b axis magnetoresistance support this idea.[25]

In the semiconducting state of the PF_6 salt, the interpretation of the large nonsaturating magnetoresistance with values of $\Delta\rho/\rho \sim 10$—100 involved the existence of closed, compensated electron and hole bands with a large mobility.[11] The Hall mobility from these studies was $\sim 10^5$ cm^2/volt s. Moreover, the dependence on scattering rate was checked by radiation damage studies which showed that $\Delta\rho/\rho \sim (w_c\tau)^2$.[26] However, the deviations from H^2 dependence are not easily explained. Parameters characterizing the electron and hole bands suggest that for fields above ~ 60 kG at temperatures below 5 K, one is entering the quantum regime.

Some general comments are necessary before trying to interpret the new field-dependent thermopower results. In a semiconductor in the quantum regime, the thermopower and magnetoresistance are nonsaturating functions of field.[27] Since, in the present case, with the PF_6 salt the thermopower is definitely saturating at low fields (Fig. 4), we are probably not in the quantum regime.

On the other hand, the classical treatment of the high-field thermopower, for both metals and semiconductors, gives a saturating thermopower independent of whether

the magnetoresistance is saturating or not.[28] Therefore the observation of a saturating thermopower is to be expected.

Thermoelectric effects arise from two contributions: diffusion thermopower from the energy dependence of the density and mobility of the carriers, and boson drag thermopower from the electron-boson (phonon-magnon) interaction and the heat of the boson gas. Both of these contributions can change in the presence of a magnetic field as carriers are swept across the Fermi surface and experience different mobilities and different interactions with the bosons.[29] The fact that no identifiable phonon-drag contribution is observed in the zero-field thermopower suggests that this is not the dominant effect. However, studies of the magnetic field dependent thermopower in noble metals have shown that the phonon-drag thermopower may be enhanced in the presence of a magnetic field.[29]

One of the usual features of the magnetothermopower in the metallic phase is that it has a different sign for the ClO_4 salt than for the PF_6 salt. This may result from the slightly different low-temperature band structures of the two materials. The former compound has an anion-ordering transition at 24 K which splits the two sides of the Fermi surface into two pieces each but leaves only open orbits. Also, the PF_6 salt presumably has a contribution to its thermopower from fluctuations into the SDW state at temperatures just above 12.5 K.[19]

We now turn to an interpretation of the new results. The most striking and unusual effect that we see in the magnetotransport properties is the change in the thermopower of the PF_6 salt when a field is applied along the c^* axis. The characteristic temperature dependences for metals and insulators are

$$S=(k_B/e)(k_BT/E_F) \quad \text{for metals}, \tag{2a}$$

$$S=(k_B/e)(E_g/2k_BT) \quad \text{for insulators}. \tag{2b}$$

Since $k_B/e \sim 87\ \mu V/K$, a metal should have a considerably lower thermopower than $87\ \mu V/K$, while a semiconductor should have a comparable or larger thermopower. Moreover, a semiconductors thermopower should vary as $1/T$.

Thus the thermopower of $(TMTSF)_2PF_6$ is not characteristic of the semiconducting behavior which is seen in the resistance. Although the zero-field data in itself would not be considered striking, and could be explained in terms of a particular set of parameters for the band structure, energy dependent scattering, and donor or acceptor concentration, the fact that a much more characteristic semiconducting behavior is restorted with application of a magnetic field is quite striking. The effect of a magnetic field on an SDW transition is quite small as is known both theoretically and experimentally. Note, for example that the transition temperature has changed by less than 0.1 K in the presence of a field of 80 kG.

It should be emphasized at this point that the large effects which are seen are only present for the configuration in which the field is aligned with the c^* axis (compare Figs. 1 and 2). This orientation causes the electrons to circulate in the a-b plane, in which the transfer integrals and bandwidths are maximum. Thus it appears clear that the effects which are observed are orbital in origin. In the absence of strong spin-orbit coupling we would expect spin effects to be isotropic.

The general behavior of the thermopower suggest that there are at least two types of carriers present. Predominantly positive carriers with energy close to the chemical potential at temperatures just below the transition and in the absence of a magnetic field, and negative carriers with higher energy at low temperatures or in the presence of a magnetic field.

Since the properties of quasi-one-dimensional systems have proven to be a very attractive topic for theoretical physicists, there are many possibilities to be explored. Aside from the thermally excited band carriers, there have been various suggestions that transport may have sizable contributions from sliding SDW, fractionally charged solitons related to the SDW transition, and the presence of superconducting fluctuations. In addition, there are more mundane explanations possible in terms of an impurity band, localized impurity states, and phonon drag. Another possibility is antiferromagnon drag from the magnetic excitations below the SDW transition.

The case for fluctuating superconductivity is the weakest since the supercurrent should have no thermopower of its own and should not participate in interactions with the other carriers. Likewise, the sliding SDW contribution should also have no entropy term unless there were strong interactions with normal carriers or other elementary excitations.[30]

Conwell[22] has suggested that the discrepancy in sign between the Hall coefficient and the a axis thermopower (at zero field) may result from the presence of solitons. In her model the solitons are of sufficiently high concentration that they overlap along a single chain and thus have their dominant conductivity along the a axis with little conductivity along the c or b axis. The Hall effect sees only the band carriers and remains positive as does the b axis thermopower. The thermopower sees the solitons and the band carriers. At low temperature the Fermi level lies below the soliton states so that they appear as electron-like (i.e., negative) carriers. If this model is correct, then its extension to the present observations would go approximately as follows. The band carriers producing a positive contribution are frozen out by the magnetic field. This leaves only the solitons with negative effective charge to contribute to the thermopower.

The freeze out of the normal carriers may be due to the classical magnetoresistance effects or to Landau quantization raising the energy of the band states. What is unusual in this picture is that the thermopower should have a similar field dependence to the magnetoresistance.[27] Also, the solitons must be thermally activated to give the exponential resistance that is observed and the soliton states must be close to the band-gap edge in order to give a thermopower with a $1/T$ dependence and a value consistent with the gap that one expects from the observed transition temperature.

While an explanation in terms of phonon drag is probably inappropriate simply because the temperatures are too low (phonon drag peaks usually occur at about

$\Theta_D/5 \sim 40$ K for these salts), it may be that scattering or drag effects may be associated with the new modes created by the SDW, the phasons, and the antiferromagnons. The drag effect would have to be positive and its quenching by the magnetic field may come from the increased scattering rate from other processes caused by the orbital circulation of the electrons into high scattering regions.

Among the more mundane explanations, the contribution of an impurity band is unlikely since the defect concentration is low as evidenced by the high mobility ($> 10^4$ cm^2/volt s).[11] The impurity band would have to contribute positively to the thermopower and freeze out at low temperatures or in high fields to explain our results. This is contrary to what would be expected.

In more traditional semiconductors the magnetothermopower has been investigated in Ref. 27. The diffusion thermopower in the classical regime saturates as field is increased to a value which is $\frac{1}{2}(k_B/e)$ for acoustic scattering and is reduced by a value of $\frac{3}{2}(k_B/e)$ for ionized-impurity scattering in the case of an n type semiconductor. While these changes are the same order as those which are observed in our samples, they cannot explain the change of sign with temperature or field that we see. The classical dependence of the phonon-drag thermopower on field is simply proportional to the magnetoresistance change and is therefore not consistent with our observations.

CONCLUSIONS

The magnetotransport measurements on the $(TMTSF)_2X$ compounds points out two major characteristics: the high mobility of the band carriers at low temperature and the quasi-two-dimensional nature of the band structure. Although these properties can account qualitatively for the similarity in the magnetoresistance in the different salts, we see that the thermopower behavior is different specifically between the PF_6 and the ClO_4 salts even in the metallic state.

The most unusual discovery revealed in this paper is the strong dependence of the thermopower in the semiconducting state on applied magnetic field along the c^* direction. Unless the band structure and scattering mechanisms have conspired to produce unexpected results, the straightforward interpretation of our findings is that low energy (that is residing close to the Fermi energy) positive carriers dominate the transport at temperatures just below the metal-insulator transition. These carriers freeze out upon lowering the temperature or applying a magnetic field along the c^* direction. What remains are negative carriers with relatively large energy (distance to E_f large compared to k_BT). Comparison with Hall measurements and transverse thermopower measurements indicate that only the positive carriers are mobile in the perpendicular (b) direction.

Another possibility deserving further investigation is that the thermopower indicates a drag effect with the relevant dragged bosons being the excitations from the spin-density-wave state.

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