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## Molecular Crystals and Liquid Crystals

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### Magnetic and Spectroscopic Properties of $(\text{TMTTF})_2(\text{SbF})_{1-x}(\text{AsF}_5)_x$

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MAGNETIC AND SPECTROSCOPIC PROPERTIES OF  
 $(\text{TMTTF})_2(\text{SbF}_6)_{1-x}(\text{AsF}_6)_x$

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**ABSTRACT**  $(\text{TMTTF})_2\text{SbF}_6$  shows an anomaly on the temperature dependence of its electrical conductivity at 154K and at low temperature an antiferromagnetic ground state. On the other hand  $(\text{TMTTF})_2\text{AsF}_6$  ground state is non-magnetic. We present the spectroscopic and magnetic studies of these two salts and of their alloys  $(\text{TMTTF})_2(\text{SbF}_6)_{1-x}(\text{AsF}_6)_x$ .

The knowledge of the low temperature ground state is an important asset in the study of an organic conductor. The nature of this ground state is often related with the high temperature behaviour of the compound as for example in  $(\text{TMTTF})_2\text{SCN}^+$ . More generally the competition between the ground states is at least partially driven by the interaction between anions and organic stacks as proposed by Bruinsma et al<sup>2</sup>

In order to study the influence of the anion we have performed spectroscopic (I.R.) and magnetic measurements of the two pristine salts  $(\text{TMTTF})_2\text{SbF}_6$  and  $(\text{TMTTF})_2\text{AsF}_6$  and of their alloys.

The D.C. electrical conductivity of  $(\text{TMTTF})_2\text{SbF}_6$  shows an anomaly at 154K<sup>3</sup>. A possible explanation of this behaviour is an ordering of the centrosymmetrical  $\text{SbF}_6^-$  anion. A recent specific heat study<sup>4</sup> of this compound fails to detect any anomaly of  $C_p$  in the range 10K-300K. This result together with recent X-ray measurements which show no evidence for either  $4k_F$  or  $2k_F$  scattering<sup>3,5</sup> probably indicate that the electronic localization is not related with an order-disorder phase transition.

The ordering of non-centrosymmetrical anions is clearly evidenced by infrared spectroscopy. For example the intensity of the  $\nu_4$  vibronic line increases at the order-disorder transition in  $\text{ReO}_4^-$  or  $\text{BF}_4^-$  salts of both TMTF and TMTSP<sup>6</sup> and the  $\nu_4$  frequency shifts at the temperature transition in the SCN<sup>-</sup> and  $\text{ReO}_4^-$  TMTSP salts<sup>7</sup>. In both  $\text{AsF}_6^-$  and  $\text{SbF}_6^-$  salts of TMTTF no frequency shift is detected on the  $\nu_4$  vibronic line between 15K and 300K.

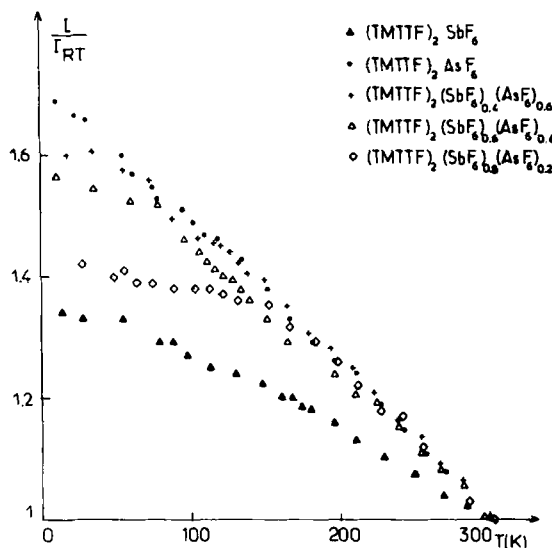


FIGURE 1 Temperature dependence of the normalized intensity of the  $\nu_4$  vibronic line

The intensity of this line increases monotonously in the  $\text{AsF}_6$  and  $\text{SbF}_6$  salts but however with a saturation at low temperature for this last salt (figure 1). By alloying, the behaviour of the I.R. spectrum evolves regularly between that of the two undoped samples.

These results are in agreement with the hypothesis that in  $(\text{TMTTF})_2\text{SbF}_6$  the electronic localization occurs without any condensation of a new long range order.

The E.P.R. results and static magnetic susceptibilities are given figure 2 and 3 for the pristine salts. The temperature dependence of the E.P.R. signal of  $(\text{TMTTF})_2\text{SbF}_6$  shows two characteristic features. First a bump of the linewidth around 154K reminiscent of the electrical anomaly is observed. Then a divergence of the linewidth is found at low temperature together with a slight decrease of the g-factor. At the same time the static magnetic susceptibility remains finite as the temperature approaches zero. This behaviour already found for the SCN salt<sup>1</sup> usually indicates the condensation of an antiferromagnetic ground state. The occurrence of such a ground state below  $T_C \approx 8\text{K}$  has been recently confirmed through antiferromagnetic resonance<sup>8</sup> and magnetic anisotropy measurements<sup>9</sup>.

On the other hand the  $\text{AsF}_6$  salt shows a broadening of the E.P.R. linewidth, but the static magnetic susceptibility is activated below  $T_c \approx 12\text{K}$ . These results are most likely related to the condensation of a non-magnetic ground state induced by a Spin-Peierls transition as confirmed by magnetic anisotropy measurements<sup>9</sup>.

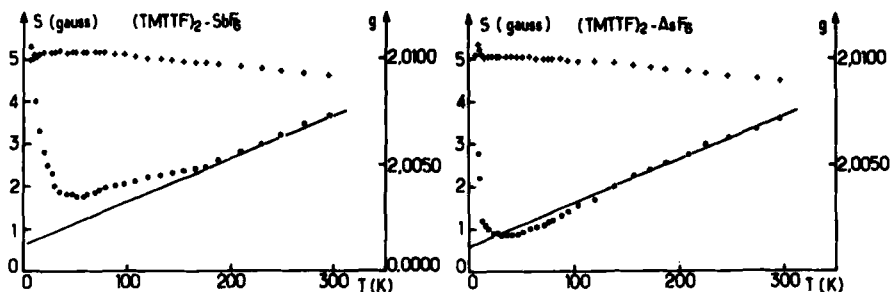


FIGURE 2 E.P.R. linewidth (...) and g-factor (++++ ) T dependences. The static magnetic field was parallel to the short axis of the TMTTF molecule.

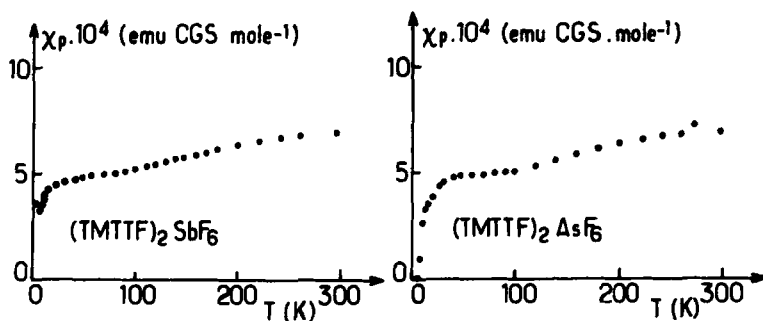


FIGURE 3 Temperature dependence of the static magnetic susceptibility

The  $(\text{TMTTF})_2(\text{SbF}_6)_{1-x}(\text{AsF}_6)_x$  alloys behave in the same way as the undoped salts when  $x < 0.2$  (close to  $\text{SbF}_6$ ) and  $x > 0.8$  (close to  $\text{AsF}_6$ ). Around  $x \approx 0.5$  neither an A.F. nor a N.M.

ground state is observed above 5K suggesting that the disorder induced by the anions dominates the low temperature properties of these materials. However a continuous behaviour versus  $x$  is found as far as the high temperature anomaly is concerned. This anomaly is reflected in both the conductivity, E.P.R. linewidth and I.R. properties. That means that the  $\text{SbF}_6$  salt is not singular among the TMTF series. On the contrary a similar anomaly is observed for several compounds even prepared with anions of different symmetries<sup>10</sup>. Among the salts with octahedral anions the  $\text{SbF}_6$  compound is only the material for which the electronic localization occurs at the highest temperature.

In this picture it may be suggested that there is a correlation between the nature of the low temperature ground state and the high temperature electronic localization, even if no real phase transition is associated with this effect. A more extensive discussion together with new experimental data concerning these samples will be presented in forthcoming publication.

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