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MAGNETIC INSTABILITIES IN TMTTF SALTS

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The static magnetization and magnetic susceptibility anisotropy of $(\text{TMTTF})_2 \text{X}$ (where $\text{X} = \text{AsF}_6$ and SbF_6) have been investigated between 3 and 300 K for magnetic field strengths up to 50 kG. A spin Peierls phase transition is observed in AsF_6 salt around 11 K, whereas in the $(\text{TMTTF})_2 \text{SbF}_6$ compound an antiferromagnetic (AF) transition is detected around 7 K.

I - Introduction

Since the discovery of the superconductivity (SC) and the SDW in $(\text{TMTSF})_2 \text{PF}_6$ under and at ambient pressure respectively (1,2), an intensive study has been undertaken on $(\text{TMTSF})_2 \text{X}$ series and their sulfur analogs $(\text{TMTTF})_2 \text{X}$. The recent experimental and theoretical progress show evident similarities between the physical properties of these series (3,4,5,6). It has been shown that the observed low temperature instabilities are related to the electron-electron and electron-phonon Umklapp scattering processes (4,5). In this paper we present and discuss some results on the magnetic properties of $(\text{TMTTF})_2 \text{X}$ ($\text{X} = \text{AsF}_6$, SbF_6).

II - Experimental

The parallelepipedic needles of crystals were obtained with the electrochemical method (7).

The magnetization anisotropy was measured as function of temperature (3-300 K) and magnetic field strengths up to 50 kG using a SQUID magnetometer (S.H.E. Corp.). The measurements were carried

out on ~ 25 mg of crystals. The crystals were placed in a small pocket, and aligned parallel to the *a*-axis (axis of the needles). Their *b* and *c* axes were pointing randomly in the perpendicular plane. The pocket was suspended to a glass fiber with the *a*-axis parallel or perpendicular to the applied magnetic field direction.

III - Results

In figure 1 are plotted the magnetizations of the AsF_6 and the SbF_6 salts respectively as a function of the magnetic field strength at different temperatures, with the field perpendicular to the *a*-axis. The respective contributions of the mylar pocket and the glass fiber were substrated. The error barr in relative value is about 10 %. In the $(\text{TMTTF})_2 \text{AsF}_6$ case, the magnetizations measured parallel and perpendicular to the *a*-axis are linear function of the field strength up to 50 kG and at all temperatures. The deduced susceptibilities are constant with temperature from 25 K down to about 14 K (figure 2) like the $(\text{TMTTF})_2 \text{PF}_6$ behavior (8). Below this temperature an abrupt decrease in both directions ($a//H$ and $a \perp H$) is observed, indicating the occurrence of a spin Peierls (SP) phase transition at about 11 K. The $(\text{TMTTF})_2 \text{SbF}_6$ behavior is more complex. The magnetic susceptibilities χ_{\perp} and $\chi_{//}$ decrease monotonously with temperature down to 20 K like the $(\text{TMTSF})_2 \text{PF}_6$ behavior (9). At lower temperature an important decrease is observed with a magnetic field dependence of the susceptibility (figures 3 and 4). This is clearly evidenced by the magnetization behavior (see figure 1). These results, completed by those obtained with a torque method, indicate the occurrence of an antiferromagnetic phase transition (AF) at about 7 K and a spin-flop transition at $H_{\text{SF}} \approx 3.2$ kG. The easy axis *b'* is found along the transverse molecular axis.

IV - Discussion and conclusion

The measured magnetic anisotropy at high temperature is essentially issued from diamagnetism (10). The paramagnetic anisotropy is very

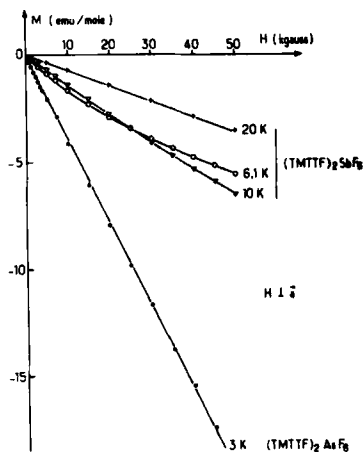


Fig. 1 : Magnetization of $(\text{TMTTF})_2 \text{AsF}_6$ and $(\text{TMTTF})_2 \text{SbF}_6$.

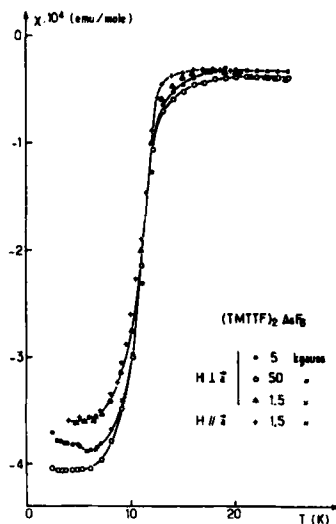


Fig. 2 : T. dependence of the magnetic susceptibility of $(\text{TMTTF})_2 \text{AsF}_6$.

weak and can be neglected. The behavior of the AsF_6 salt is clear. The observed anomaly is related to the spin Peierls phase transition, like the one detected in $(\text{TMTTF})_2 \text{PF}_6$ (8) and confirm the recent observation of $2 k_F$ diffuse scattering below 40 K (11). However no evidence for $2 k_F$ diffuse scattering was found down to 15 K in the SbF_6 salt (11). The results of magnetic anisotropy show the occurrence of an antiferromagnetic phase transition at about 7 K and a spin-flop transition near 3 kG in agreement with antiferromagnetic resonance experiments of COULON et al. (12). However the behavior of the magnetic susceptibilities above 7 K is particular. (fig. 3 and 4), relative to those observed in SCN and Br salts (10, 13, 14). Below 18 K a sharp decrease of magnetic susceptibility occurs and at 7 K almost half of the paramagnetism has been lost ($\chi_{\text{dia}} \sim -3,7 \times 10^{-4}$ emu/mole).

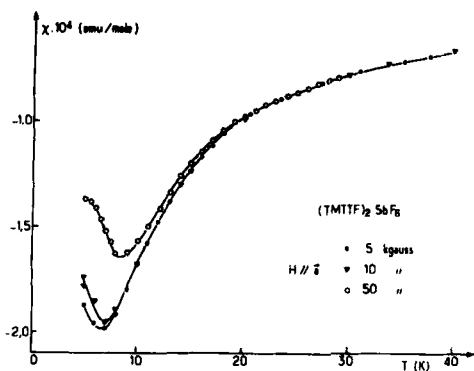


Fig. 3 : T. dependence of the magnetic susceptibility of $(\text{TMTTF})_2 \text{SbF}_6$.

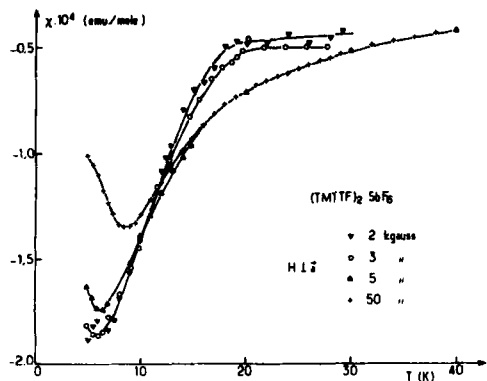


Fig. 4 : T. dependence of the magnetic susceptibility of $(\text{TMTTF})_2 \text{SbF}_6$.

A similar behavior has been observed in the first AF compound investigated, $(\text{TMTSF})_2 \text{PF}_6$ (2). This compound showed a decrease in χ (although of less importance) from 15 K down to the (AF) transition at 11 K. One of the explanations suggested was a cross-over from one-dimensional to three-dimensional ordering. This could be valid for $(\text{TMTTF})_2 \text{SbF}_6$, which could be more one dimensional than the PF_6 and AsF_6 analogs, due to larger interstack distances. Another argument could be found in the recent experiments of CREUZET *et al.* (3), who showed that $(\text{TMTTF})_2 \text{PF}_6$ undergoes an (AF) transition under pressure instead of the (SP) transition observed at ambient pressure (8).

This decrease could result from a (SP) transition occurring at about 18 K. Then the field dependence of the magnetic susceptibility at low temperature could be related to the onset of short range magnetic order leading to a uniform (non-distorted) phase, as observed in other (SP) systems, where a field-induced transition appears analogous to a spin-flop transition (15). However the absence of diffuse scattering down to 15 K is not in favour of this interpretation.

Another possibility would be the occurrence of precursor effects preceding an (AF) transition at 7 K.

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