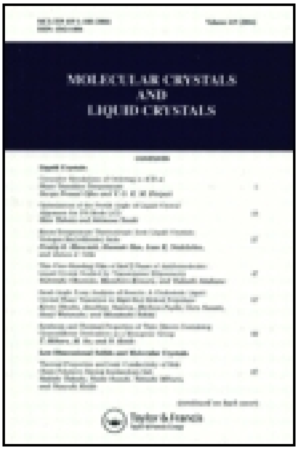

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THE CONDUCTIVITY AND THERMOPOWER OF $Ta_{1-x}W_xS_3$

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Abstract The conductivity and the thermopower of samples synthesized from various mixtures of $Ta_{1-x}W_xS_3$ have been measured. Doping is more efficient and the doped samples have more reproducible (orthorhombic-like) properties after growth at high temperatures (750-800C). All samples are p-type at room temperature.

INTRODUCTION

TaS_3 appears in two structures[1,2], orthorhombic (o) and monoclinic (m) easily distinguished by their phase transitions at : T_c=215 K in TaS_3(o) and T_H = 240 K and T_L = 160 K in TaS_3(m). The growth conditions that produce the (o) and (m) structures (or their mixture) are not yet under control. TaS_3 (m) is isostructural with $NbSe_3$ [3] which has a 1/4 filled energy band.[4] The fourfold periodicity[5] of the CDW along the chain axis (below the transitions) which is a common feature of the three types of crystals indicated that TaS_3 (o and m) has also a 1/4 filled band at high temperatures. It was therefore surprising to find[6] that according to the sign of the thermopower (S):

- TaS_3 (o) is p-type above and below T_c
- TaS_3 (m) is p-type above T_H, n-type between T_H and T_L and p-type below T_L

The sign of S above the transitions is in contradiction with a 1/4 filled band model.

Hall measurements were prevented so far by the samples' dimensions and therefore the clue to this puzzle must come from a different approach. The present approach is to study the effect of impurities on the TaS_3 samples. Tungsten (W) was chosen first because it is the closest atom to Ta having one extra d electron. The first step was to experimentally answer the following questions:

- Can TaS_3 be doped with tungsten?
- If so, can W change the sign of S above or below the transitions?
- Can W stabilize one of the two structures?

Preliminary results of this study are presented here.

EXPERIMENTAL METHOD AND RESULTS

The method of preparation and the properties of samples obtained from two growth runs (1 and 2) are summarized below:

1. Three evacuated quartz tubes, containing mixtures of Ta + 3.05 S (a), 0.9 Ta + 0.1 W + 3.05 S (b) and 0.95 Ta + 0.05 W + 3.05 S (c) were heat treated at 750-800C for two weeks, cooled to 400C during two weeks and cooled further to room temperature during several hours. The products contained long fibers (characteristic of MS_3), flakes (characteristic of MS_2) and excess sulfur. The temperature dependence of the resistance and the Seebeck coefficient (S) of fibers from tube (a) showed that they have a mixture of the (o) and (m) structures. The fibers from the tubes (b) and (c) were orthorhombic like (see Fig. 1a and 1b) with highly reproducible properties. Samples b were superior to all TaS_3 (o) samples grown in the past in this laboratory. Their activation energy and threshold field for the onset of nonlinear conductivity (1 V/cm or less) are smaller than in pure TaS_3 (o). Short samples of this tube could be easily treated to show large current oscillations[7] in the MHz regime. Auger analysis of some of these samples showed that the concentration of W in the bulk is smaller than the nominal concentration by about a factor of two.

2. Four evacuated quartz tubes containing mixtures of MS_4 in which 1/8 (A), 1/6 (B), 1/4 (C) and 1/3 (D) of the Ta atoms were substituted by W, were heat treated at 500-550C for a month and then cooled to room temperature during several hours. The products

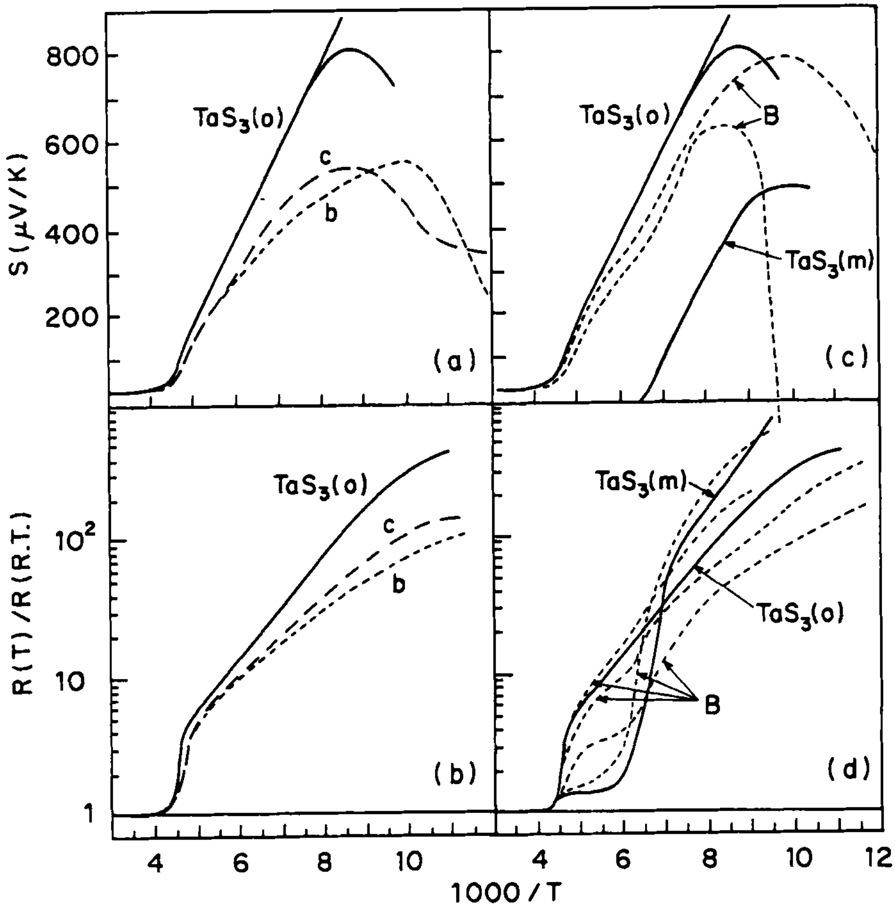

FIGURE 1 Seebeck coefficient and normalized resistance vs the inverse temperature for $Ta_{1-x}W_xS_3$ samples (see text). The results for pure TaS_3 (o) and TaS_3 (m) (solid lines) are shown for reference.

contained fibers (inferior to the previous ones) very few flakes and excess sulfur. The properties of the fibers are sample dependent (see Fig. 1c and 1d for samples of tube B). They are intermediate between those of (o) and (m) structures with a tendency towards those of the (o) structure enhanced by the higher nominal concentration of W. All the samples are p-type at high temperatures. A small negative S was found between T_H and T_L in only one sample (of tube A). In all samples S exhibited below T_L a maximum of variable magnitude. In some samples (see one example in Fig. 1c) the maximum was followed by a steep drop towards large negative values (-4000 μV/k at 80K). This effect was reproduced after cycling the sample through the whole range of temperatures. Auger analysis was done only in samples of tube D. It showed that the ratio W/(W + Ta) in the bulk is 0.09 (much lower than the nominal concentration).

It should be stressed that although W was found in the bulk (by analysis after sputtering) it is very important to find if it substitutes Ta in the lattice. In such a case the results presented above may have interesting implications on the defect structure and the electronic structure of TaS_3 above and below the transitions.

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