Investigation of $Fe_xCo_{1-x}Sb_3$ Thin Films as a Skutterudite and Determination their Specific Resistance

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□ ABSTRACT □

In this paper the thin films $Fe_xCo_{1-x}Sb_3$ are investigated as a skutterudite and showed some of its properties and applications: the specific resistance. The aim of this paper is determination the specific resistance of $Fe_xCo_{1-x}Sb_3$ thin films using the four probe method. This method was employed by using two different devices in two different countries and the results was compared.

Keywords: skutterudite, thin film, specific resistance, four probe method.

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دراسة الأفلام الرقيقة $Fe_xCo_{1-x}Sb_3$ باعتبارها بلورات سكوتيروديت وتحديد مقاومتها النوعية

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□ ملخّص □

تمّ في هذا البحث دراسة الأفلام الرقيقة $Fe_xCo_{1-x}Sb_3$ باعتبارها بلورات سكوتيروديت كما تمّ عرض بعض خصائصها وتطبيقاتها: المقاومة النوعية. الهدف الرئيسي من هذا البحث هو تحديد المقاومة النوعية للأفلام الرقيقة $Fe_xCo_{1-x}Sb_3$ باستخدام طريقة المجسات الأربعة. طُبِّقت هذه الطريقة باستخدام جهازين مختلفين في دولتين مختلفتين وتمت مقارنة النتائج.

الكلمات المفتاحية: بلورات سكوتيروديت، أفلام رقيقة، المقاومة النوعية، طريقة المجسات الأربعة.

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Introduction:

Due to the flexible application possibilities of thermoelectric components, industrial interest in the use of thermoelectricity has increased more and more in the last two decades. With the thermoelectric generators (TEG) it is possible to convert thermal energy into electrical energy. The problem is that the used thermoelectric materials achieve to date only an efficiency of less than 4.5% [1]. Therefore, further applications are not economical, for which the efficiency of these materials would have to be increased fivefold.

In this study, four probe method was used to study and determine the specific resistance of $Fe_xCo_{1-x}Sb_3$ thin films. Four probe device is one of the most commonly used apparatus for the measurement of resistivity of semiconductors.

Materials and Methods:

Four probe method

This method can be used when the sample is like thin film, such as in this study used samples. Four peaks arranged linearly in a straight line at equal distance s from each other are considered. A constant current I flows through the two outer probes and the potential drop V can be measured through the middle tow probes as shown in figure (1).

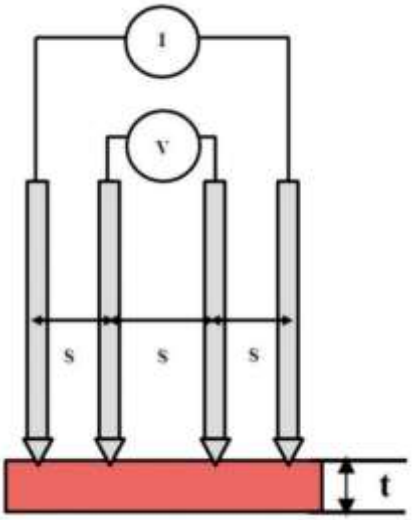

FIG. (1): Schematic diagram showing the four point probe electrical resistance measurement technique.

We assume that the sample thickness is greater than the distance between the probes, which is the same. It turns out [2, 3]:

$$\rho = \frac{V}{I} 2\pi s$$

ρ is the specific resistance. In 1995, the realization of a solid-state system " Phonon-Glass / Electron- Crystal " (PGEC) was by G.A. Slack described. In this model solid, the electrical conductivity should not be reduced by scattering of the charge carriers at point defects or other disturbances in the lattice periodicity (ideal crystalline behavior). The skutterudites are considered a good candidate for a PGEC. They got their name from the Skutterud site in Norway, where in 1928 the mineral $CoAs_3$ was found as a natural mineral. Skutterudites have the general formula TX_3. T is the transition metal from the 9th group of the periodic table (cobalt, rhenium, iridium) and X from the 5th main group (phosphorus, arsenic,

antimony). In this work, T = cobalt and X = antimony. $CoSb_3$ has a body-centered cubic structure. The structure is often illustrated with the cubic unit cell from figure (2).

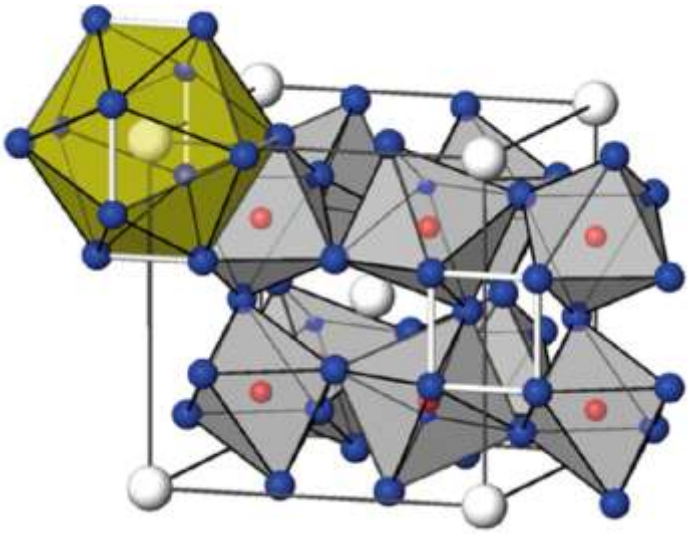

FIG. (2): structure of $YbCo_4Sb_{12}$, red: Co, bleu: Sb, white: Yb [4,2].

Two in their design different devices were used in this work. One device is located at the Technical University of Chemnitz, Germany. A picture of the structure of the four point measurements can be found in figure (3).

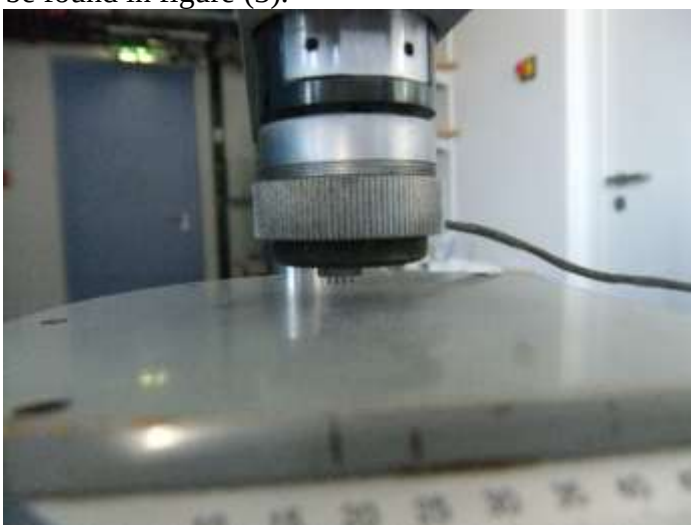

FIG. (3): Photo of the four probe arrangement at the Technical University of Chemnitz.

The distance between the tips is 1.3 mm and the tips are spring-loaded to ensure that the tips do not destroy the layer. The current I is generated by the power source of the device *POWER SUPPLIES D3D-5* from the company *DELTA ELEKTRONIKA*. The voltage drop is read using the *Prema 6000* digital voltmeter device.

The other different device is located at Al-Manara University, Syria. These two devices differed between each other in the distance of the probes. It is 1.3 mm in the device of Chemnitz University and 1.8 mm in the device of Al-Manara University. A picture of the structure of the four point measurements at Al-Manara University can be found in figure (4). The current I is generated by the power source of the device *HIGH CURRENT*

POWER SUPPLIES D3D-5 from the company *LEYBOLD LD Didactic GmbH*. The voltage drop is read using the *Multimetrix Dmm 121* digital voltmeter device.

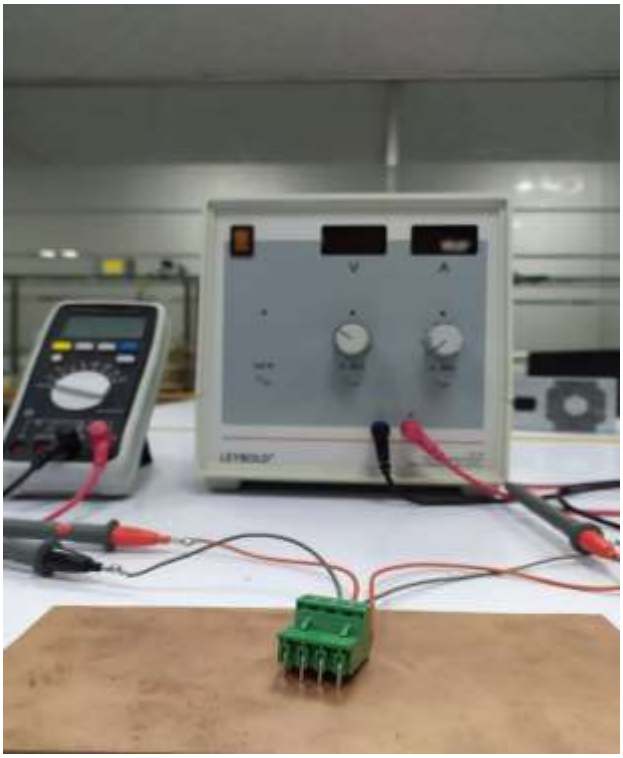

FIG. (4): Photo of the four probe arrangement at Al-Manara University.

In an ultra high vacuum (UHV) chamber were 8 $Fe_xCo_{1-x}Sb_3$ films co-deposited by molecular beam deposition. The base pressure was between 7×10^{-10} mbar and 7×10^{-11} mbar. As substrate was used $(SiO_2(100\,nm)/Si(100))$. The deposition chamber consists of an electron beam evaporator for the evaporation of Cu and Fe and an effusion cell for Sb. An optical detector can controls the flux of Co and Fe immediate. On the other hand the deposition rate of Sb was controlled by the temperature of the effusion cell. The Co deposition rate was dominated to achieve the desired nominal composition $Fe_xCo_{1-x}Sb_3$. The film thickness was modified by the deposition time. To guarantee homogeneous films, the holder of sample was rotating during the deposition, for more details see e.g. Ref. [5,6]. The compositions of the film series are summarized in Table (1). The optimal atomic ratio Fe + Co:Sb = 1:3. On average, the Sb content is around 70 at.%. All layers are between 30 nm and 35 nm thick.

TABLE (1): Compositions of the investigated film series.

samples	real	wondered
1	$Fe_{0,02}Co_{0,98}Sb_{3,1}$	$Fe_{0,01}Co_{0,99}Sb_3$
2	$Fe_{0,03}Co_{0,97}Sb_{2,58}$	$Fe_{0,02}Co_{0,98}Sb_3$
3	$Fe_{0,06}Co_{0,94}Sb_{2,71}$	$Fe_{0,05}Co_{0,95}Sb_3$
4	$Fe_{0,13}Co_{0,87}Sb_{2,82}$	$Fe_{0,1}Co_{0,9}Sb_3$

5	$Fe_{0,26}Co_{0,74}Sb_{2,48}$	$Fe_{0,2}Co_{0,8}Sb_3$
6	$Fe_{0,35}Co_{0,65}Sb_{2,59}$	$Fe_{0,3}Co_{0,7}Sb_3$
7	$Fe_{0,58}Co_{0,42}Sb_{2,44}$	$Fe_{0,5}Co_{0,5}Sb_3$
8	$Fe_{0,76}Co_{0,24}Sb_{2,46}$	$Fe_{0,7}Co_{0,3}Sb_3$
9	$FeSb_{2,81}$	$FeSb_3$

Results and Discussion:

I. Measurements by the device at the Technical University of Chemnitz

At room temperature RT, the specific resistance ρ was determined for all films using the four point method. For low Fe content, it decreases very sharply and then also goes into stability as shown in figure (5). This result corresponds with the result obtained by M. Daniel et. all [6].

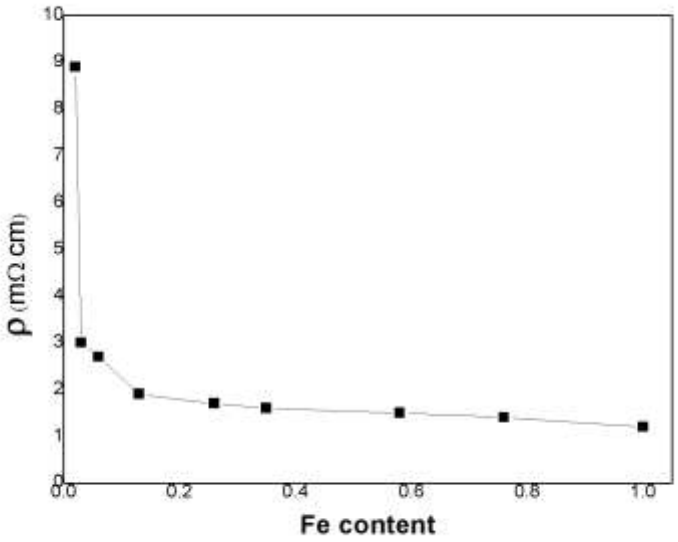

FIG. (5): The dependency of the specific resistance ρ on the Fe content x. Measurement at TU-Chemnitz.

The reason for the decrease in ρ may be the increased charge carrier density p, and this needs to be investigated by further research. The stabilities can be attributed to the formation of parasitic phases [7].

II. Measurements by the device at Al-Manara University

In the same situation, the specific resistance ρ was determined for all films using the four point method. It is easy to see the same bahavior of ρ with increasing the Fe content (see figure (6)). Actually, there is very small differents in the both values of ρ (between the measured values at Al-Manara University and the measured values at Chemnitz University). Measured values of ρ at Chemnitz University may be more accurate and close to thiers real values. In any case, we can use the both and get good results.

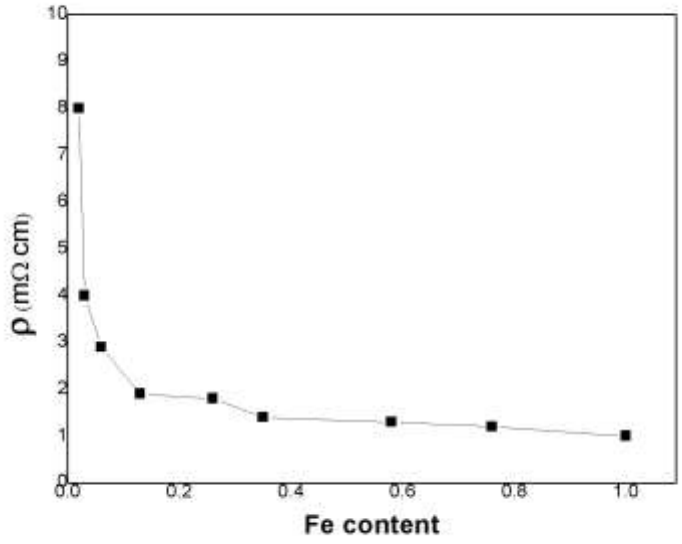

FIG. (6): The dependency of the specific resistance ρ on the Fe content x. Measurement at Manara University.

The results obtained at Chemnitz University were compared with the results obtained at Al-Manara University, as shown in figure (7).

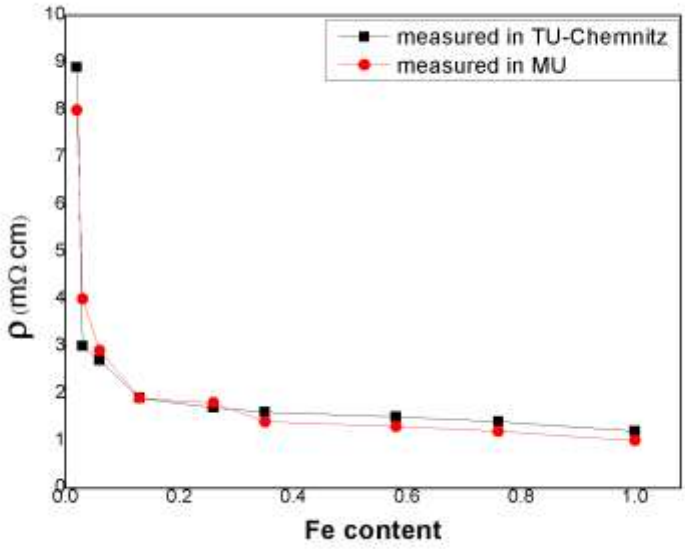

FIG. (7): comparing the results of the specific resistance, which we obtained in the two universities, red: the specific resistance measured in TU-Chemnitz, black the specific resistance measured in Al-Manara university.

From Figure (7), it can be said that the specific resistance ρ is almost the same. Thus, it is possible to use the device at Al-Manara University to obtain good results.

Conclusions and Recommendations:

The specific resistance of $Fe_xCo_{1-x}Sb_3$ thin films using the four probe method was determent. Two different devices were used to determination the values of ρ. These both devices shows the same values of ρ. For low Fe content, it decreases very sharply and then also goes into stability. The reason for the decrease in ρ may be the increased charge carrier density p. The stabilities can be attributed to the formation of parasitic phases.

Conflicts of Interest

The authors, the authors declare that there are no conflicts of interest.

Acknowledgments

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