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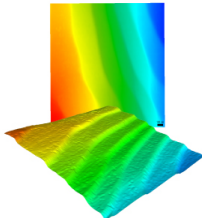
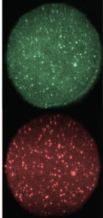
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# Experimental setup for the measurement of the electrical resistivity and thermopower of thin films and bulk materials

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An experimental setup was developed for the simultaneous measurements of the electrical resistivity and the thermopower in the temperature range going from 77 up to 330 K. The system combines accuracy, rapidity, few efforts for the sample mounting, and is fully automated. Pressure contacts are used for contacting the thermocouples to the sample and the sample to the heat sink, making possible the measurement of both thin films and bulk materials with a great flexibility on their geometrical dimensions. Tests of the system have been made on a 99.98% pure Ni bulk sample and on PbTe/BaF<sub>2</sub> thin films for illustration. © 2005 American Institute of Physics.

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## I. INTRODUCTION

During this last decade, intensive effort has been made in the search for high efficient thermoelectric materials.<sup>1,2</sup> Both low dimensional structures and bulk materials have been synthesized and their thermoelectric performance determined. Their evaluation includes the measurement of the electrical resistivity ( $\rho$ ), the thermoelectric power or thermopower ( $\alpha$ ), and the thermal conductivity ( $\lambda$ ) in order to estimate the dimensionless figure of merit  $ZT$ , defined by

$$ZT = \frac{\alpha^2}{\rho\lambda} T, \quad (1)$$

where  $T$  is the absolute temperature. High performance thermoelectric devices designed either for refrigeration or for power generation involve materials having high  $ZT$  values.

The search for advanced bulk thermoelectric materials requires testing numerous samples in order to gain information about their transport properties. Similarly, the electrical and thermal properties of thin films depend on many experimental parameters (post-treatment, growth conditions, etc.), so that it is necessary to investigate a large series of samples. For these reasons, an experimental setup is recommended for measurements of the transport coefficients combining high accuracy with high flexibility and where the sample preparation is minimized. Our endeavor has been to measure simultaneously two properties, i.e., the electrical resistivity and the thermoelectric power. This simultaneity should be an advantage because of the rapidity of execution, a smaller number of sample handlings, and the certainty about identical chemical and physical conditions during measurements.

Accurate thermopower measurements require a very careful sample preparation and well-defined thermal conditions. Generally, in the literature, very few experimental details are given concerning the measurement procedure and the setup. Usual measurement methods include the connection of the sample to the heat sink and the heat source with the thermocouples either attached directly to the sample or connected to the source and the sink.<sup>3</sup> Alternated heating with two heaters is also used.<sup>4</sup> Some details about the fabrication of sample holders for wide temperature range measurements can be found elsewhere.<sup>5-7</sup>

In this article, we describe the development of an experimental setup extended from that originally fabricated by Stölzer *et al.* for thin films of fixed size.<sup>8</sup> The measurement cell allows the simultaneous measurement of the electrical resistivity and the thermopower of both thin films and bulk materials of various sizes for temperatures ranging from 77 to 330 K. The sample holder in which only pressure contacts for the thermocouples and the heat sinks are used is first described. Then, the procedure to extract the electrical resistivity and the thermopower is developed in detail. Finally, experimental results obtained on a reference bulk sample (Ni) and on PbTe thin films are presented.

## II. DESCRIPTION OF THE SAMPLE HOLDER

The experimental setup can be roughly divided into three parts: (i) the sample holder, (ii) the cryostat, and (iii) the computer-controlled data acquisition and management of both the input data and measurement apparatus. We will emphasize here only our specific sample holder that can be modified and adapted to any kind of cryostat.

The sample holder is represented in Figs. 1 and 2. On the photo (Fig. 1), we can see that it is mainly made of a copper cylindrical body. Two small copper blocks are located at one end on which the sample is mounted. Note that a notch [not

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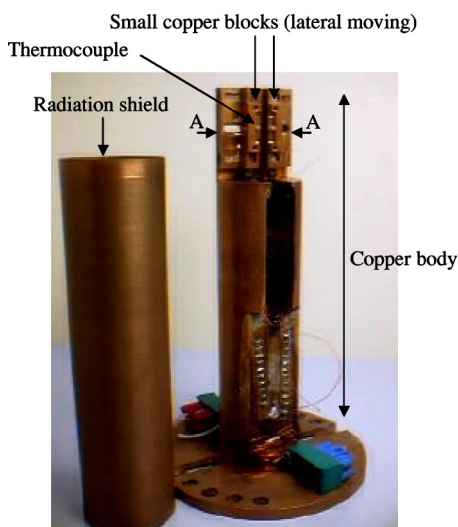


FIG. 1. (Color online) Photo of the measurement cell together with its radiation shield. The cell is devoted to the simultaneous measurement of the thermoelectric power and the electrical resistivity.

shown for simplification in Fig. 2(a)] separates the two small copper blocks in order to prevent the mechanical stress due to the difference of thermal expansion coefficient between the sample and the sample holder that could break the sample or damage the thermal contacts. The other end of the cylindrical sample holder is connected to the heat sink of the cryostat with screws. The measurement cell can therefore be easily disconnected from the cryostat making the mounting of the sample easy.

The two small copper blocks, electrically isolated from the main copper body, are placed in a symmetrical way [Fig. 2(a)]. Small resistors (typically 100  $\Omega$ ) that are used as heaters are embedded inside. The heat resistance induced by the electrical insulation determines the power consumption of the small heaters in the small copper blocks and the cooling rate of the sample. A value of 10 K/W is optimum for acceptable measurement speed and accuracy. On the other hand, this insulation layer also has to withstand large changes of temperature. These requirements are fulfilled by the use of cigarette paper as an insulation layer. It is glued on the copper part (Fig. 2) with special low temperature varnish (GE 7031 varnish).

The two small copper blocks serve both as thermal and electrical contacts to the sample. One original feature of our measurement cell is the possibility of easily changing the distance of these two blocks by a lateral sliding as illustrated in Fig. 1. In order not to damage the cigarette paper, the sliding is not performed directly on it but on an intermediary copper piece (Fig. 2). The motion allows adjusting the measurement cell to the sample size. A great flexibility with respect to the sample size can therefore be gained. Typically, samples with length varying between 5 and 22 mm can be measured. The width can vary between 1.5 and 6 mm. The thermal contact of the sample to the heat sink is usually ensured by pressing the sample between polished copper surfaces. The pressure is controlled through small screws [Fig. 2(b)]. For contacting hard and brittle samples or substrates like glass and silicon, we used indium foils inbetween.

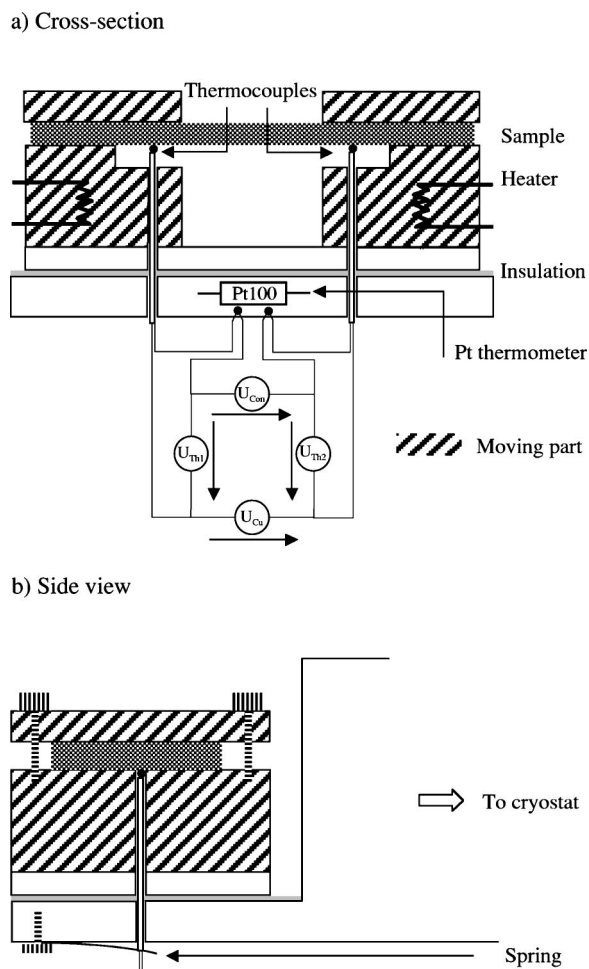


FIG. 2. Schematic view of the sample holder for the thermopower and the electrical resistivity measurements: (a) cross section taken at level A in Fig. 1 and (b) side view. When the sample is a film deposited on a substrate, the film is faced toward the thermocouples.

Differential copper–constantan thermocouples (0.127 mm diameter) sense the temperature differences between the sample and the copper body. All the leads in contact with the sample holder, including those of the differential thermocouples, are carefully thermally anchored to the copper body. The contacts of the hot junction of the thermocouples to the samples are made as follows: the thermocouple wires are threaded through two round hole alumina ceramics and their junction at one end of the alumina tube is spot welded and pressed against the sample by means of springs [Fig. 2(b)]. In order to reduce parasitic heat fluxes through the thermocouples, we used thermal grease to improve the thermal contact between the copper pieces and the alumina tubes on one hand and the thermocouple wires and the alumina tubes on the other hand. The cold junction of the thermocouples is soldered on dedicated prints of fined-down printed circuit. This circuit is located near the place where the reference temperature is measured by a platinum resistive thermometer inserted in the copper body.

All the voltages are measured using a Keithley 2000 multiscan meter via a scanner card. The current injected in the heaters and in the sample is controlled by a Keithley 224 digital source power supply.

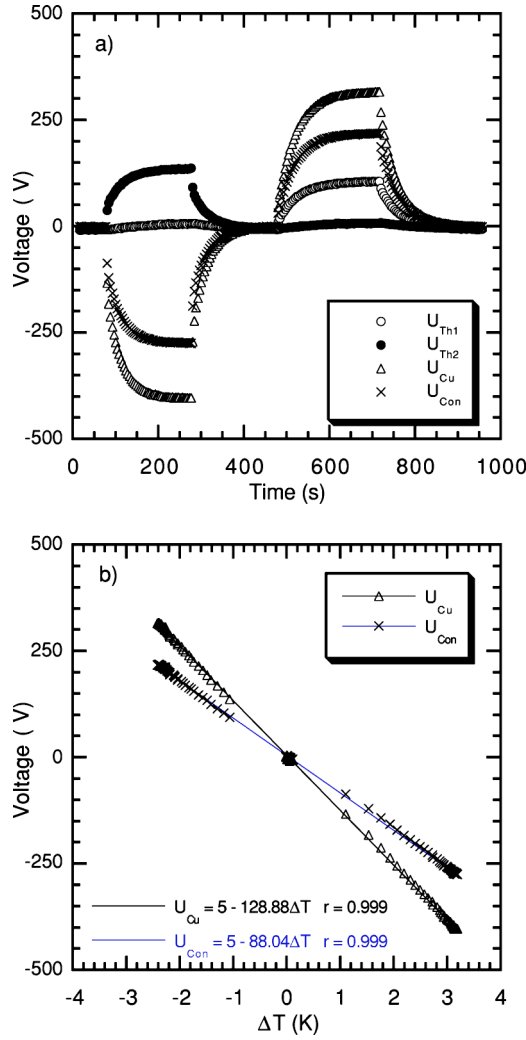


FIG. 3. (Color online) Measurement procedure to extract the thermopower from data corresponding to a measurement performed at 300 K on a PbTe thin film: (a) voltages  $U_{Th1}$ ,  $U_{Th2}$ ,  $U_{Cu}$ , and  $U_{Con}$  as a function of time and (b) determination of the slope  $a_{Cu}$  and  $a_{Con}$  to calculate the thermopower by Eq. (7).  $r$  is the coefficient of regression of the linear fit.

### III. MEASUREMENT PROCEDURES

#### A. Electrical resistivity

The measurement of the electrical resistivity of the sample is made with the standard four probes method. In this technique, the electrical current ( $I$ ) flows through the sample and the voltage drop ( $\Delta V$ ) is measured. The value of the resistivity is given by

$$\rho = \frac{\Delta V S}{I L}, \quad (2)$$

where  $L$  is the distance between the two voltage probes and  $S$  is the surface of the sample. We used the two copper branches of the thermocouples as a voltage probe. In order to reduce the influence of the thermoelectric voltage on the determination of the electrical resistivity, the voltage occurring on the sample was measured instantaneously after the application of the electrical current through the sample. Each resistivity value results from the average of several currents  $\pm I$ .

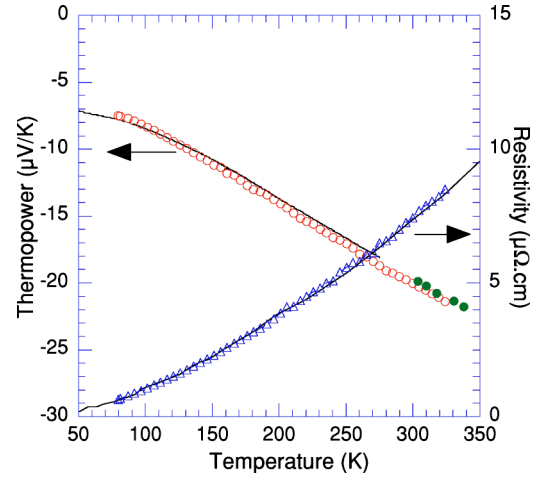


FIG. 4. (Color online) Thermopower and electrical resistivity data on bulk Ni as compared to other data. Open circles and triangles represent measured data from this work. Solid line (thermopower) from Blatt *et al.* (Ref. 9); filled circles from Burkov *et al.* (Ref. 6); solid line (resistivity) from Lewis *et al.* (Ref. 10).

#### B. Thermopower

When we apply a small temperature difference  $\Delta T$  ( $-5 < \Delta T < 5$  K) on the sample, the four voltages,  $U_{Th1}$ ,  $U_{Th2}$ ,  $U_{Cu}$ , and  $U_{Con}$  as depicted in Fig. 2(a), are recorded as a function of time.  $U_{Th1}$  and  $U_{Th2}$  represent the thermal voltages in the two thermocouple circuits and  $U_{Cu}$  and  $U_{Con}$  the voltages in the copper circuit and in the constantan circuit, respectively. From  $U_{Th1}$  and  $U_{Th2}$ , the mean temperature  $T_{av} = (T_1 + T_2)/2$  of the sample and  $\Delta T = T_1 - T_2$  are calculated.  $T_1$  and  $T_2$  represent the temperatures at the location of the thermocouple junction on the sample.

The voltages  $U_{cu}$  and  $U_{Con}$  measured between the input terminals of a voltmeter can be expressed as

$$U = \int dV = - \int \alpha dT \quad (3)$$

where the integral is between terminal 1 and terminal 2 of the voltmeter.

If the terminals are at the same temperature, we get

$$U_{Cu} = - \int_{T_1}^{T_2} (\alpha_{Cu} - \alpha) dT, \quad (4)$$

$$U_{Con} = - \int_{T_1}^{T_2} (\alpha_{Con} - \alpha) dT,$$

where  $\alpha_{Cu}$ ,  $\alpha_{Con}$ , and  $\alpha$  are the thermopower of the copper, the constantan, and the sample, respectively.

When  $\Delta T \ll T_{av}$ , the system (4) reduces to

$$U_{Cu} = (\alpha_{Cu} - \alpha) \Delta T, \quad (5)$$

$$U_{Con} = (\alpha_{Con} - \alpha) \Delta T.$$

In fact, we often have to consider an offset voltage ( $U_{off}$ ) in the copper and constantan circuits.  $U_{off}$  may rise from different sources: instrumentation, parasitic thermal gradient, but also from a nonlinear temperature distribution coming together with an inhomogeneous thermopower anywhere in

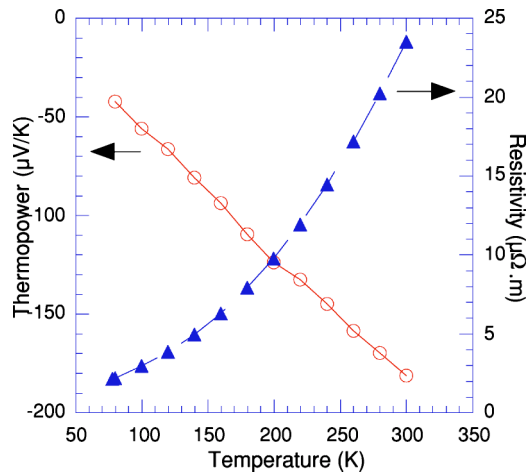


FIG. 5. (Color online) Temperature dependence of the thermopower and the electrical resistivity of a PbTe thin film deposited on BaF<sub>2</sub>(111).

the electrical circuit.<sup>8</sup> If we suppose that  $U_{\text{off}}$  is independent of  $\Delta T$  and has the same value for  $U_{\text{Cu}}$  and  $U_{\text{Con}}$ , which is well verified experimentally, Eq. (5) becomes

$$U_{\text{Cu}} = (\alpha_{\text{Cu}} - \alpha)\Delta T + U_{\text{off}} = a_{\text{Cu}}\Delta T + b, \quad (6)$$

$$U_{\text{Con}} = (\alpha_{\text{Con}} - \alpha)\Delta T + U_{\text{off}} = a_{\text{Con}}\Delta T + b,$$

where  $a_{\text{Cu}}$  is the slope of the curve  $U_{\text{Cu}}(\Delta T)$  and  $a_{\text{Con}}$  is the slope of the curve  $U_{\text{Con}}(\Delta T)$ .

From Eq. (6), it is possible, after some calculations, to extract the thermopower  $\alpha$  independently of  $\Delta T$ :

$$\alpha = \frac{a_{\text{Cu}}}{a_{\text{Con}} - a_{\text{Cu}}} \alpha_{\text{Cu/Con}} + \alpha_{\text{Cu}}, \quad (7)$$

where  $\alpha_{\text{Cu/Con}} = \alpha_{\text{Cu}} - \alpha_{\text{Con}}$  is the thermopower of the copper-constantan thermocouple.

The relation

$$U_{\text{Th1}} - U_{\text{Th2}} + U_{\text{Cu}} - U_{\text{Con}} = 0, \quad (8)$$

which is evident from Fig. 2(a), is used as a verification of the measured data.

In the testing phase, we used the following extended procedure for measuring the thermopower:

- (1) Step 1: heating the left hand side with constant power until  $\Delta T = \text{constant}$ ;
- (2) Step 2: heater off, waiting until  $\Delta T = \text{constant}$ ;
- (3) Step 3: heating the right hand side with the same power until  $\Delta T = \text{constant}$ ; and
- (4) Step 4: heater off, waiting until  $\Delta T = \text{constant}$ .

The four voltages are plotted as a function of time as illustrated in Fig. 3(a). From a linear regression of  $U_{\text{Cu}}$  and  $U_{\text{Con}}$ , the coefficients  $a_{\text{Cu}}$  and  $a_{\text{Con}}$  are calculated [Fig. 3(b)] and the thermopower is calculated using Eq. (7). The points used for the calculation of the two slopes ( $a_{\text{Cu}}$  and  $a_{\text{Con}}$ ) corre-

spond to  $\Delta T = 0$  and  $|\Delta T| > 1$  K. The coefficient of regression of the linear fit is used as a mark of quality of the measurement. Generally, it is higher than 0.995. A low value of this coefficient or a long time to get the thermopower measurement are indicators of a bad thermal contact of the sample or the thermocouples. The error in the determined thermopower is usually below 5%. On high resistive films ( $> 2$  k $\Omega$ ), the error is larger due to noise.

## IV. RESULTS

The best way to validate a measurement system is to use a reference material for which the absolute thermopower and the electrical resistivity are well established. From the point of view of the reliability and precision of the measurement, we selected nickel. The dimensions of the Ni parallelepipedic-shaped sample were  $9.00 \times 3.90 \times 0.25$  mm<sup>3</sup>. The experimental results are reported in Fig. 4 and compared with literature data.<sup>6,9,10</sup> The measured thermopower and resistivity values are in excellent agreement with these published values.

To see the capability of our system on thin films, we present results obtained on a PbTe thin film. The layer was prepared by pulsed laser deposition from stoichiometric PbTe target onto a BaF<sub>2</sub>(111) substrate. The substrate temperature was kept at 300 °C during the growth. We used a rectangular mask ( $2 \times 8$  mm<sup>2</sup>) to fix the geometry of the deposit. Figure 5 displays the results obtained on a 1  $\mu$ m thick *n*-type PbTe layer. As expected, the thermopower data are negative in the whole temperature range investigated and the values are in good agreement with the values reported by Ravich *et al.*<sup>11</sup> for similar carrier concentration ( $n \sim 2.10^{18}$  cm<sup>-3</sup> at 300 K). Moreover, the temperature dependence behaviors of both the resistivity and the thermopower are typical of those of a degenerated semiconductor and that are expected for such a high carrier concentration.

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