International Journal of Thermophysics, Vol. 17, No. 6, 1996

Note

Thermal Conductivity and Electrical Resistivity of Pure Polycrystalline Cobalt in the Temperature Range 2.5–30 K

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Received January 30, 1996

Thermal conductivity and electrical resistivity of 99.99% pure Co sample were measured in the temperature range 2.5–30 K. The annealing procedure of the sample (either above or below Curie temperature), followed by cooling it down to room temperature at a slow cooling rate, caused an unexpected increase in its thermal resistivity and residual electrical resistivity, contrary to the results obtained for most pure metals. Co samples either not thermally treated or annealed consist only of a H1 phase as proved by X-ray and electron diffraction analyses. The result led to the conclusion that changes of grain structure and physical defects appearing in the Co at Curie temperature and at 690 K, when phase transitions take place, should be taken into account. The electron-magnon scattering is significant in electrical conductivity but the electron-physical defect and impurity scattering plays a dominant role in thermal conductivity. The electron-physical defect and impurity scattering is elastic (validity of the Wiedemann–Franz law) as demonstrated by the value of $\beta_{th}/\beta_{el} = 1.0$, obtained in this work.

KEY WORDS: cobalt; electrical resistivity; low temperatures; thermal conductivity.

1. INTRODUCTION

Annealing of metal samples (with a very slow cooling rate) before thermal conductivity and electrical resistivity measurements is a standard way to

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obtain reproducible values of thermal and electrical properties of a metal. This procedure reduces both the mechanical strains and the physical defects (such as point defects, dislocations, etc.) in the metal under investigation. For a pure tin sample in which a phase transition from f.c.c. to b.c.t. takes place at 286.2 K [1], reproducibility of thermal conductivity and electrical resistivity values at low temperatures was established also when the sample was repeatedly heated above and cooled below this temperature [2, 3].

The main objective of this work was to determine the influence of thermal treatment on the thermal conductivity and electrical resistivity of a pure cobalt sample at low temperatures. The effect of mechanical deformation as well as the annealing procedure on elastic properties of cobalt was considered in Ref. 4, and elastic properties of pure cobalt in the proximity of spin-reorientation phase transition was described in Ref. 5.

2. EXPERIMENTS

A 99.99% pure rod-shape cobalt sample (5-mm diameter and 54-mm length) supplied by Johnson–Matthey was used in the experiments. Electrical resistivity measurements were performed in the temperature range from 2.5 to 30 K. A thermomagnetic superconducting tantalum modulator as a null detector of sensitivity 5×10^{-12} V was applied [6]. Random errors of measurements were estimated to vary from 0.2 to 2%, depending on the temperature, while the systematic error due to determination of the sample geometry was estimated to be 3%. A typical longitudinal heat flow, steady-state method [1] for measuring thermal conductivity in the temperature range from 2.5 to 30 K was used. Random errors depending on the sample temperature varied from 0.2 to 0.6%; the systematic error in the determination of the sample geometry was 1.8%.

The following measurements were performed.

(a) Thermal conductivity and electrical resistivity in the temperature range 2.5–30 K of Co samples not thermally treated (curve 1 in Fig. 1 and curve 1 in Fig. 2, respectively).

(b) Thermal conductivity of the same sample after annealing at about 1430 K (slightly above three/quarters of its melting point), followed by slow cooling to room temperature, at an average cooling rate of 20 $\text{K} \cdot \text{h}^{-1}$ (5 $\text{K} \cdot \text{h}^{-1}$ in the vicinity of the phase transition temperature). The thermal conductivity of the sample decreased by about 15% in this experiment (curve 2 in Fig. 1).

(c) Dependence of thermal conductivity on temperature for the above Co sample after annealing at 1230 K (Curie temperature:

1400 K), followed by slow cooling to room temperature. A further decrease in thermal conductivity was observed (curve 3 in Fig. 1).

(d) Dependence of thermal conductivity on temperature for the above Co sample but after annealing for a week at 680 K (a phase transition in Co takes place at 690 K), followed by slow cooling to room temperature. In comparison to experiment c, an increase of about 3% in the thermal conductivity was observed (curve 4 in Fig. 1). The dependence of electrical resistivity of the sample on temperature was also determined. In comparison to the results obtained in experiment a, the electrical residual resistivity increased by about 30% (curve 2 in Fig. 2).

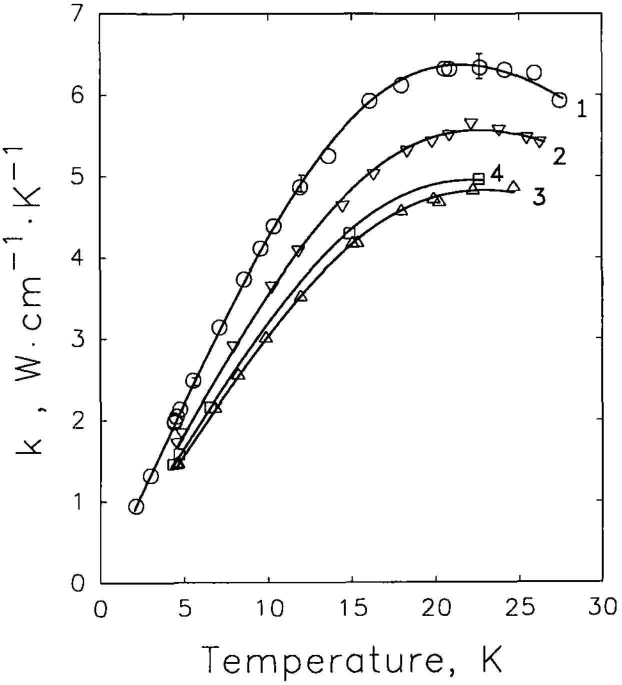

Fig. 1. Thermal conductivity vs temperature for Co sample. Curves 1–4—see the text, Section 2. The curves are described by the equation $1/k = \alpha T^2 + (\beta/T)$, where $\alpha = 1.14 \times 10^{-4}$, 1.18×10^{-4}, 1.32×10^{-4}, and 1.36×10^{-4} $W^{-1} \cdot cm \cdot K^{-1}$ and $\beta = 2.24, 2.70, 3.16$, and 2.99 $W^{-1} \cdot cm \cdot K^2$, for curves 1, 2, 3, and 4, respectively.

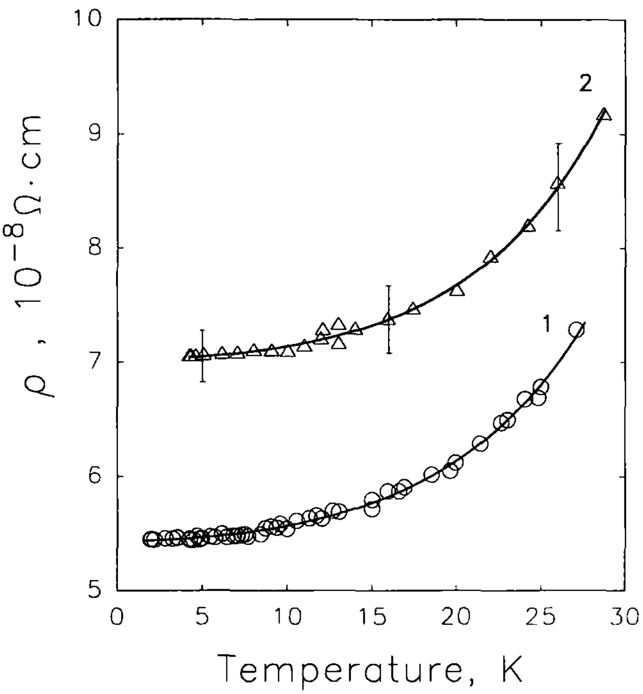

Fig. 2. Electrical resistivity vs temperature for Co sample. Curves 1 and 2—see the text, Section 2. The curves are described by the equation $\rho = \rho_0 + A_1 T^2 + A_2 T^5$, where $\rho_0 = 5.44 \times 10^{-8}$ and $7.02 \times 10^{-8} \Omega \cdot \mathrm{cm}$, $A_1 = 1.13 \times 10^{-11}$ and $1.12 \times 10^{-11} \Omega \cdot \mathrm{cm} \cdot \mathrm{K}^{-2}$, and $A_2 = 5.80 \times 10^{-16}$ and $6.34 \times 10^{-16} \Omega \cdot \mathrm{cm} \cdot \mathrm{K}^{-5}$ for curves 1 and 2, respectively.

Additionally, to test the method of thermal conductivity measurements, dependence of the thermal conductivity of a polycrystalline Nb sample (99.99% pure) on temperature was determined in the following two experiments with the same experimental setup:

(i) after annealing of the Nb sample as in item b for Co and

(ii) after annealing of the Nb sample as in item c for Co.

There are no phase transitions in the Nb within the temperature range of the thermal treatment.

Agreement of the results of experiments (i) and (ii) was obtained within the limit of experimental error; this validates the proper operation of the experimental system.

3. RESULTS AND DISCUSSION

Thermal conductivity k versus temperature T data have been described using the following equation for $k(T)$ for pure metals and dilute alloys around the thermal conductivity maximum at low temperatures:

$$1/k = W = \beta/T + \alpha T^2 \tag{1}$$

where W is the total electronic thermal resistivity, β is the electron-physical defect and impurity scattering parameter, and α is a constant.

Curves representing the data according to Eq. (1) are presented in Fig. 1. Deviations of the curves from the experimental points do not exceed 3%. To answer the question whether electron-physical defect and impurity scattering in the investigated Co sample is elastic or inelastic, β_{th} calculated from Eq. (1) and β_{el} calculated from the Wiedemann–Franz equation were compared. It has been found that for the sample not thermally treated and after thermal treatment, $\beta_{\text{th}}/\beta_{\text{el}}$ was equal to 1.0. The results indicate that electron-defect and impurity scattering is elastic (see Ref. 7).

Dependence of the electrical resistivity of the Co sample (either not thermally treated or annealed at 680 K) on the temperature is presented in Fig. 2 (see items a and d, Section 2). Residual electrical resistivity ρ_0 increased from $5.44 \times 10^{-8} \Omega \cdot \text{cm}$ for the non-annealed sample to $7.02 \times 10^{-8} \Omega \cdot \text{cm}$ for the thermally treated sample, which suggests an increase in the lattice disorder as the result of annealing. Both ρ_0 values are smaller than the $\rho_0 = 9.05 \times 10^{-8} \Omega \cdot \text{cm}$ recommended in Ref. 1.

The dependence of electrical resistivity on temperature was described satisfactorily by the following equation:

$$\rho(T) = \rho_0 + A_1 T^2 + A_2 T^5 \tag{2}$$

where ρ_0 is the residual electrical resistivity, $A_2 T^5$ is the Bloch–Grüneissen term related to electron–phonon scattering, and $A_1 T^2$ is the electron-magnon scattering term [8, 9]. Attempts to describe the dependence by the above equation but without the $A_1 T^2$ term was not satisfactory. However, $\rho(T^2)$ can also arise for electron–electron scattering but in the lower temperature range (below 1 K) [10]. Similarly, the term $\rho(T^2)$ for vibrating impurity atoms [11] can be omitted because of the high purity of our Co sample. We have also considered the term $\rho(T^2)$ as resulting from electron-point defect scattering, but it should significantly change its value after the annealing procedure, which we have not observed. Finally, we can state that A_1 values for the non-thermally treated and for the annealed Co sample are equal to $1.1 \times 10^{-11} \Omega \cdot \text{cm} \cdot \text{K}^{-2}$. This value is in good agreement with the $A_1 = 1.3 \times 10^{-11} \Omega \cdot \mathrm{cm} \cdot \mathrm{K}^{-2}$ reported in Ref. 8 for electron–magnon scattering in Co.

The transmission electron micrograph of the non-thermally treated Co sample prepared as a thin foil is shown in Fig. 3. Several grains with sizes of a few micrometers can be distinguished (scale bar is shown in Fig. 3). Selected area electron diffraction patterns obtained from many grains in the sample could be indexed only in the H1 crystal structure of Co (an example of the indexed pattern is shown in Fig. 4). Grain sizes in the annealed sample increased significantly so that sample areas thin enough to be observed by TEM contained one grain only. However, electron diffraction patterns from various grains could again be indexed only in the H1 structure. This result is consistent with XRD spectra, where also only H1 peaks were observed.

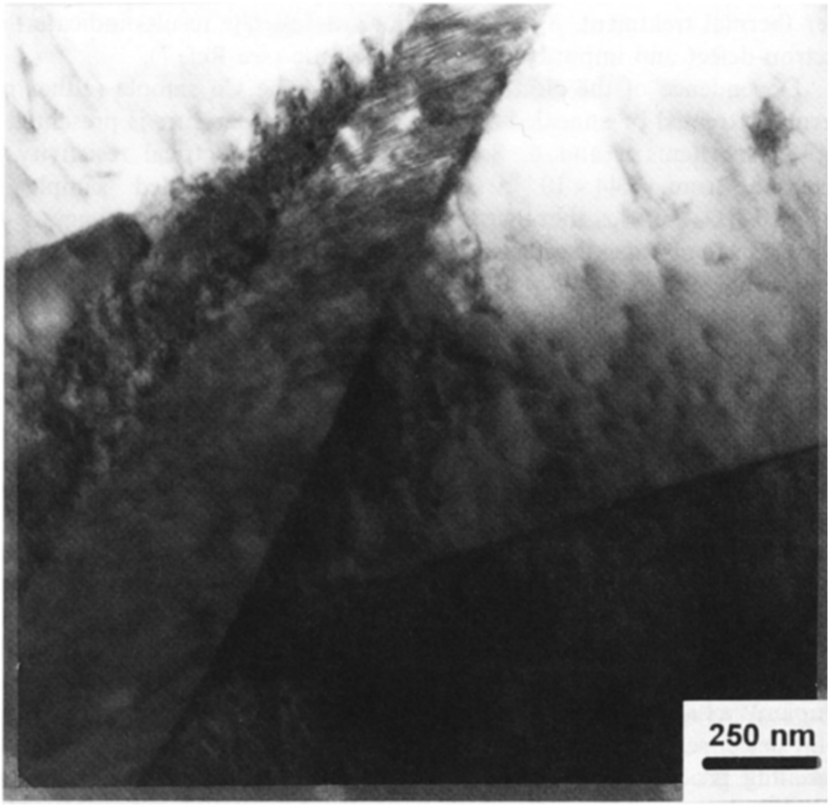

Fig. 3. Transmission electron micrograph of the non-thermally treated Co sample.

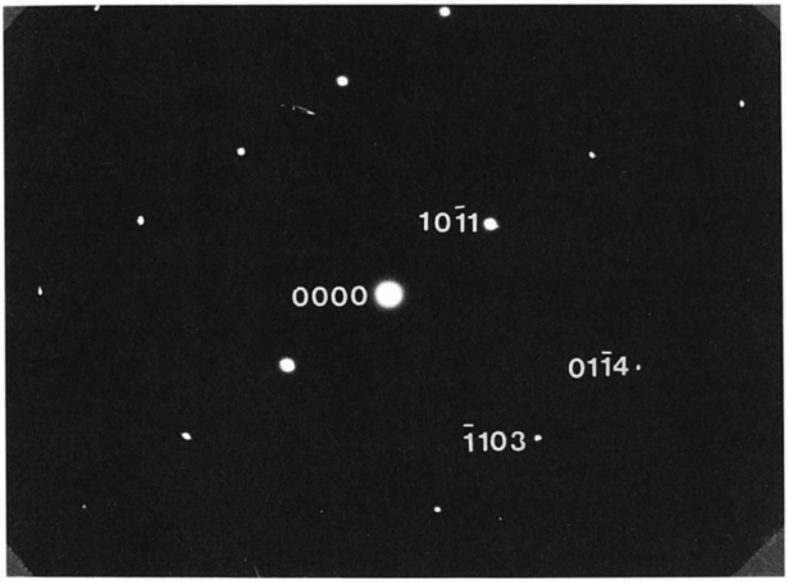

Fig. 4. Selected area electron diffraction pattern obtained from one grain in Fig. 3. The electron beam is parallel to the [2 − 7 5 3] direction of H1 Co.

4. CONCLUSIONS

The dependence of thermal conductivity and electrical resistivity on temperature measured for the Co sample in comparison to the recommended curve of the thermal conductivity given in the literature [1] indicates that our sample was of a lower physical defect concentration and a higher chemical purity. This results from the higher thermal conductivity maximum and the lower residual electrical resistivity of our Co sample.

Annealing of the Co sample either above or below the Curie temperature, followed by slow cooling to room temperature, caused an unexpected increase in its thermal resistivity and an increase in its residual electrical resistivity, which is in contradiction to the results obtained for most pure metals. Annealing of the sample at 680 K for 1 week caused only a slight increase in its thermal conductivity. However, only the H1 phase was present in either the non-thermally treated or the annealed Co sample, as can be seen from the results of X-ray and electron diffraction analyses. The changes of grain structure and physical defects appearing in the Co at Curie temperature and at 690 K, at which phase transitions take place, should be taken into account.

In the investigated Co sample, electron–magnon scattering is significant in electrical conductivity, whereas electron-physical defect and impurity scattering is dominant in thermal conductivity. The electron-physical defect and impurity scattering is elastic, which verifies the value $\beta_{th}/\beta_{el} = 1.0$, obtained by us.

ACKNOWLEDGMENT

The authors thank Prof. Z. Henkie for his valuable remarks during the preparation of the manuscript.

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