# MEASURING THE THERMOPOWER
# OF ORDERED AND DISORDERED ALLOYS

by

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# ABSTRACT

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A number of atomic systems display an atomic order-disorder transition. These alloys have a low conductivity so that a direct measure of the Fermi surface is not possible. An alternative to a direct measurement is the comparison of electron transport and equilibrium properties. In this paper, thermopowers will be compared. A method of obtaining the thermopower over a range 150°K to 400°K is discussed. Data for 5 order-disorder systems $Cu_{0.83}Pd_{0.17}$ ($\equiv$ CuPd*), CuPd* Mn, CuPd* Ni, $Cu_3Au$, and $Cu_3AuMn$, plus data for 3 homogeneous alloys, CuMn, AgMg, and AlAg (10%) are given. Direct comparison of the thermopower of samples with the thermopower of copper does not give consistent results. The total thermopower can be separated into two separate constituents, lattice vibration, and residual. After this separation trends do appear.

# ACKNOWLEDGEMENTS

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# TABLE OF CONTENTS

## LIST OF FIGURES

# LIST OF TABLES

Page

# INTRODUCTION

A number of alloy systems have an atomic order-disorder transition. The influence of such a transition upon the properties of a given alloy are difficult to predict. The effects for a specific property can range from discontinuous value changes in one system to nearly imperceptible changes in another system. For a single alloy system, the same range of behavior can occur for the different properties of the alloy. Thus, characterization of atomic order-disorder systems requires systematic measurements of numerous properties.

Since the thermopower is sensitive to the details of the electron scattering process, it is reasonable to expect some rather striking changes in this property when atomic ordering occurs. Although interpretation of such changes is likely to be difficult and to be dependent upon knowing other properties of the alloy, this property may serve as a valuable guide in selecting systems deserving more detailed study. Therefore, this thesis deals with the thermopower of several alloys having atomic order-disorder transitions.

The thesis is divided into three major sections. Section I deals with the development of an apparatus to perform thermopower measurements. Since these measurements are part of a survey of properties the apparatus had to satisfy two primary conditions with respect to the samples. First, small samples of similar but not identical sizes had to be accomodated. Second, the thermopower measurements could not contaminate the samples; such contamination would render the samples unsuitable for subsequent measurements of their other properties. The fact that the thermopower is a sum of individual contributions, each of which has a significant temperature dependence, added an additional requirement: the thermopower measurements had to be made over an extended temperature range. Section I provides a detailed description of how these requirements were satisfied, how the apparatus was calibrated, and the final procedure used to collect data.

Section II presents data for the systems studied. The most systematic results are for $Cu_{0.83}Pd_{0.17}$ (this system does not have a $Cu_3Pd$ composition to eliminate a consideration of tetragonal distortion effects) and $(Cu_{0.83}Pd_{0.17})_{1-C}Ni_C$ alloys. More limited data for this same host with Mn impurities and for the isomorphic system $Cu_3Au$ are given.

Section III deals with interpretation of the data. As expected, other properties are needed for a convincing interpretation. However, if the thermopower is interpreted as a sum of individual contributions, patterns arise that question a previous interpretation for $Cu_3Au$. This section concludes with an outline of a consistent method for identifying the individual contributions of the thermopower and some preliminary results.

# SECTION I

# APPARATUS

As mentioned in the introduction, the device to measure the thermopower of samples with an atomic order-disorder transition had to meet the following requirements: (1) Small samples of varying dimensions had to be accomodated by the device. (2) Because the samples were to be used for further measurements the method of mounting the samples had to be noncontaminating. (3) The measurements had to be made over as wide a temperature range as possible.

The size of a "typical" sample was a compromise between a long wire, ideal for transport measurements, and a sphere, ideal for magnetic susceptibility measurements. Thus actual samples were small rods 1-2 cm in length and 0.1-0.2 cm in diameter. This size in combination with the temperature range requirement indicated that a differential method of thermopower measurement was needed. In this method a small temperature gradient is produced across the sample. Both the temperature gradient and the accompanying voltage are measured and the thermopower is obtained from the slope of a voltage vs temperature gradient plot. This thermopower value is associated with the average temperature of the sample.

The sample was mounted onto a copper reference block as shown in Figure 1. The sample was supported in two directions. In the vertical direction a screw with sufficient travel to support

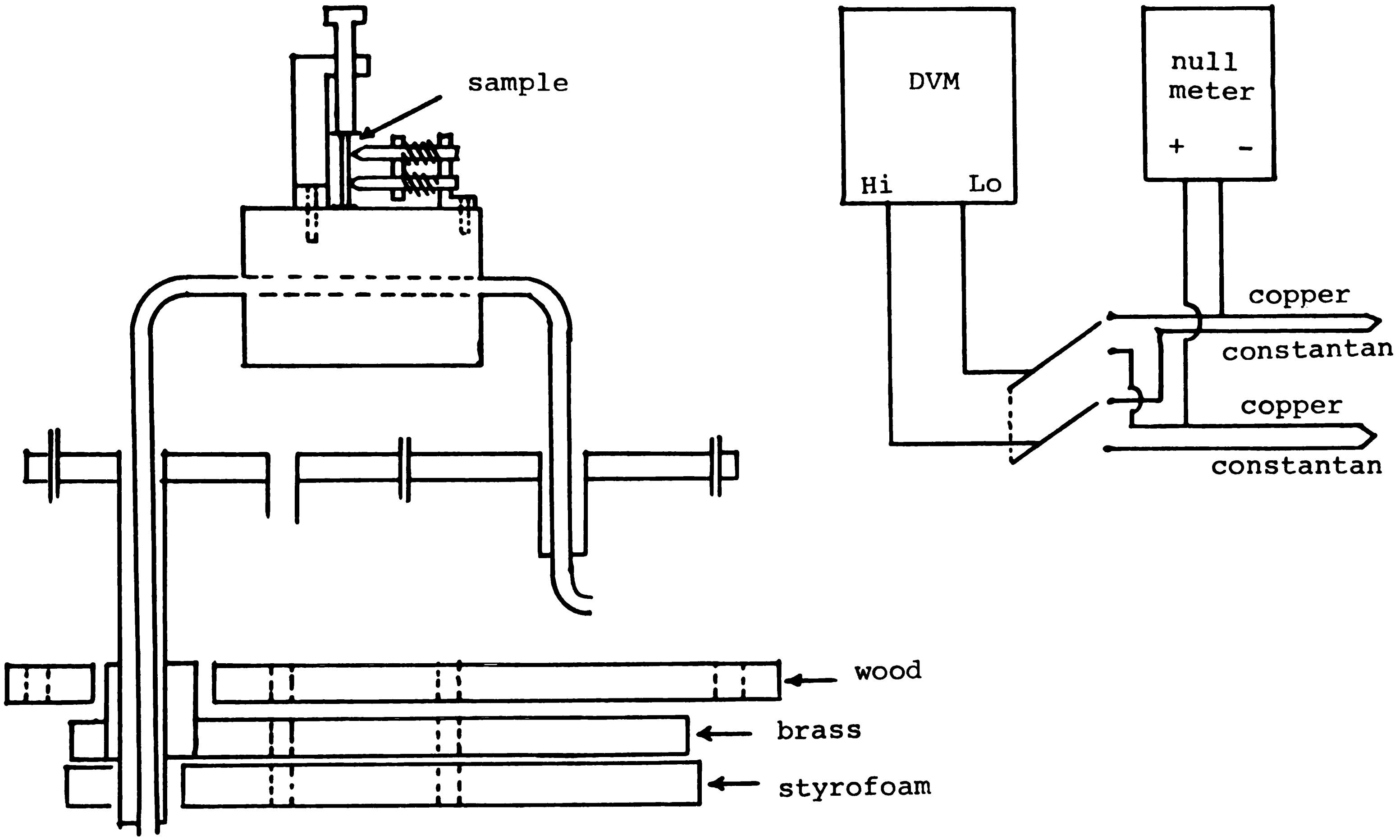

Figure 1 Apparatus for Measuring Thermopower

any sample was used to accomodate the various lengths. To adapt to the variable diameters of the samples and to provide non-contaminating electrical contacts, horizontal probes of copper slid in and out through holes in the rear support pressing the sample against a nylon back. Both of these (copper probe) press fittings were spring loaded.

To make the temperature measurements a copper constantan thermocouple was connected to the tip of each of the two horizontal probes. Initial spot welds proved too fragile so low thermal solder was used to attach the thermocouples to the horizontal probes because its properties should minimize the effects of small temperature gradients across the pressed contacts. The thermocouples had to run through the support plate without any interruption to eliminate uncontrolled stray voltages which would be produced at connections. The thermocouples were connected to a double pole double throw switch and then to a single digital voltmeter. The voltmeter needed a range of ±5.000 mV with an accuracy of .001 mV to cover the temperature range used in this experiment. For ease in operation the thermocouples were referenced to room temperature.

The double pole double throw switch enabled a rapid measurement of both temperatures. The difference of two temperatures gave the thermal gradient along the sample. From the switch, a connection was made so that the two copper wires of the thermocouples and the sample formed a thermocouple whose output was constantly monitored by a null detector. Thus, temperature and voltage measurements were nearly simultaneous. On the finest scale the null detector could read ±30.0 microvolts, full scale.

The entire assembly was mounted under a bell jar with fore-pump vacuum providing thermal insulation. Major temperature changes were achieved by altering the temperature of the copper reference block. For sample temperatures below room temperature, vapor from liquid nitrogen was used for a coolant because it was easily available, capable of reaching reasonable low temperatures, and easy to work with. A dewar, filled with liquid nitrogen, had a small air space at the top where a vertical tube was placed. As seen in Figure 1, this tube would collect the nitrogen vapors and carry them through a copper tube to a temperature reference block on which the sample stood. The vapors would then be exhausted into the room.

Controlled variation in the temperature of the reference block was obtained by sealing the dewar top and varying the rate at which the nitrogen vapors flowed. The dewar was sealed with a one inch block of styrofoam which fit tightly enough on the dewar to withstand the pressure of the nitrogen vapors. The styrofoam also acted as insulation. A brass plate above the styrofoam held the tube for the vapors, wires for a liquid nitrogen heater and a port to add liquid nitrogen. Bolts ran through a block of wood on top of the brass plate to another block at the bottom of the dewar to secure the apparatus. A heater connected to a variable power supply controlled the rate at which the nitrogen boiled. This controlled the vapor flow rate and thereby controlled the reference block temperature. With this set up a sample temperature of 150°K could be obtained.

To obtain a more complete picture of the thermopower variation, temperatures above room temperature were also desirable. The low thermal solder that had been used on the apparatus had a low melting point and, to prevent damage to the soldered connections, the maximum sample temperature was limited to 400°K. A heat lamp connected to a variable power supply was used to obtain these temperatures. This method of heating was easy to use, since the lamp was shone through the bell jar directly onto the sample and reference block.

In addition to controlled variation of the temperature of the reference block, it was necessary to have a controlled variation in the temperature gradient of the sample. The power required for the needed size of the gradient would vary, according to the physical size of the sample and the thermopower it produced. The gradient had to be large enough to produce a measurable voltage. This gradient was generated with a 10 ohm resistor which was attached to the top of the vertical support screw. This heater was electrically isolated from the screw. See Figure 1. The resistor was connected to a variable power supply which passed a maximum of 500 mA through the resistor. The leads were then connected to the resistor with alligator clips so the connections could be removed while the screw was raised or lowered to accomodate different sample sizes.

Making measurements over the range 150°K to 400°K required good electrical and thermal contacts. Without good electrical contact between the sample and the horizontal probes, the voltages from the thermocouples would not be reliable. During early runs of the experiment, electrical contact between the sample and the horizontal probes was lost, due to thermal contractions of the probes and sample. Individual springs for each of the probes were needed to maintain good contact throughout the cooling. A piece of plastic was placed on top of the copper reference block to prevent a short between it and the bottom probe. On one occasion, due to contaminated electrical contact, there was an electrical potential at the point of contact of the probes and the sample. To eliminate this electrical potential the tips of the probes were swabbed with sulfuric acid but this process destroyed the soldered connection of the probes to the thermocouples. Emery paper and care should be used to clean the tips of the probes.

The sample needed good thermal contact with the copper reference block and the heater to facilitate the flow of heat and to produce a smoothly changing, even, temperature gradient. Initial experiments indicated that this contact was crucial. Electrically insulated contacts and even direct press fit contacts at the screw-sample and sample-reference block junctions gave unreliable results. To provide this good thermal contact between the sample and the reference block, and the sample and the vertical screw securing it, lead foils were placed at each end of the sample.

Further evidence for the importance of good thermal contact comes from the thermal gradients which occurred when no power was supplied to 10Ω heater. The support for the vertical screw was nylon so that the only contact of the sample with the reference block was through one end of the sample. Despite several modifications of the heater leads there was always an initial temperature gradient across the sample. As the reference block temperature dropped, this initial gradient increased. At room temperature this initial difference was less than 1°K, at 150°K the difference across the sample was around 10°K. When a sample of larger diameter was used this initial temperature difference across the sample decreased.

To obtain the thermopower as a function of temperature a set of data points was taken every 25°K to 30°K over the range 150° to 400°K. These data would supply enough information to discern how the thermopower varied as a function of temperature. With this spacing the measurements did not overlap. At each selected temperature power was applied to the resistance heater on top of the vertical screw causing the gradient across the sample to increase. As the voltage produced by the sample increased by 0.5 microvolts, the voltage from the upper and lower thermocouple would be recorded as well as the sample voltage. Once the maximum temperature gradient was achieved, the power to the heater would be shut off, the gradient allowed to decrease, with data being taken in the same way. For each point in the temperature range about forty groups of the three measurements were produced.

The average temperature of the sample over these forty readings was the specific point that a given thermopower was identified with. The thermocouple voltages were converted to temperatures and the difference of the two temperatures found. The slope of the line, voltage vs temperature gradient, would give the total thermopower of the system of sample and copper leads. A least squares fit of the data was used to find the slope of the line. The total thermopower of the system was subtracted from the thermopower of copper at the point of interest to obtain the thermopower of the sample. When wired as in Figure 1, with the top thermocouple having a greater temperature, a negative deflection of the null meter would produce a positive total thermopower.

The accuracy of the results from the samples and the consistency of results between samples had to be insured, therefore calibration measurements were made periodically between sample runs. These calibration measurements were made with a sample of pure aluminum.

The data from Al at 162°K is shown in Figure 2. A least squares fit of the data gives a intercept of $-0.54$ μV, a slope of 3.10 μV/°K, and a correlation coefficient of 0.98. Within 5% our results, Figure 3, were reproducible and consistent with the results of Gripshover, et. al.(1)

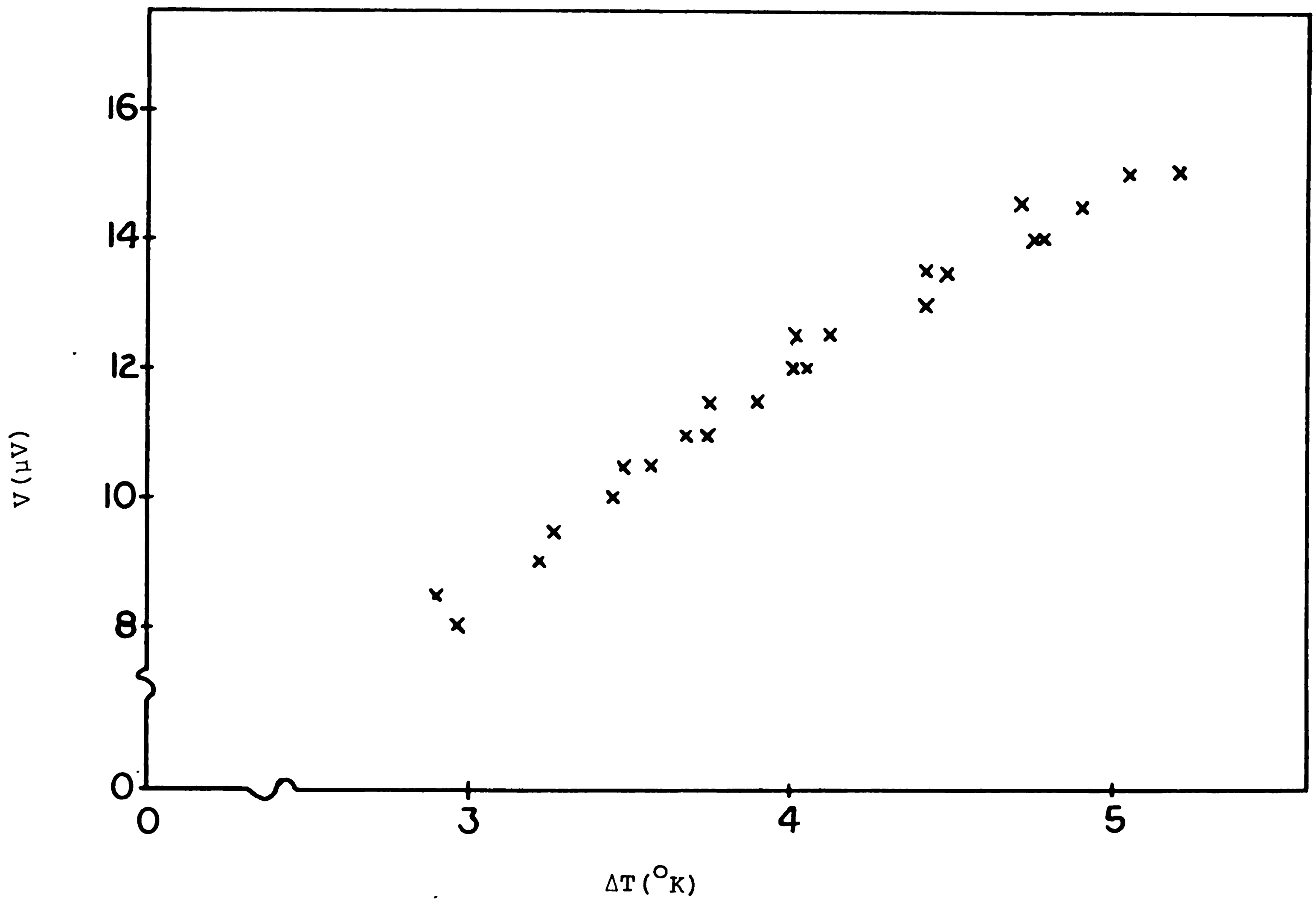

Figure 2 Data For Al at 162°K

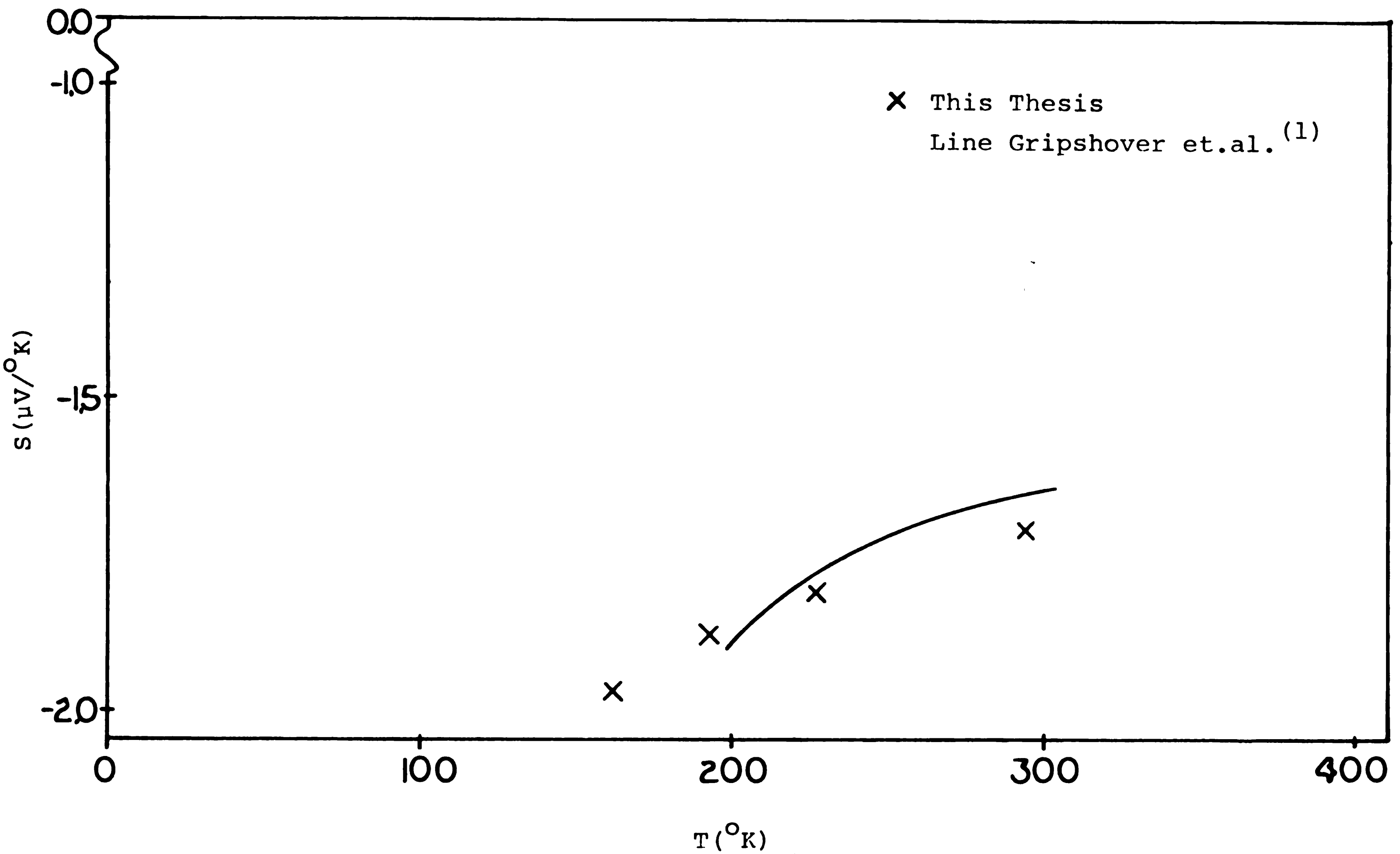

Figure 3 Thermopower of Al

# SECTION II

## DATA

The tables and graphs in this part of the thesis are a summary of the data taken. In the tables, the first column lists the sample which was tested. The second column gives the average temperature over which a set of data, consisting of a sequence of two temperature measurements and the associated voltages produced by the sample, was taken. From each set of data a plot of voltage vs temperature gradient was made. The slope of this line is given in the third column. To find the thermopower of the sample, the slope had to be corrected for the thermopower of copper. The copper thermopower is given in the fourth column and the absolute thermopower of the sample is given in the fifth column. Following the tables are graphs of the data more pertinent to this paper. The thermopower of these samples are plotted vs temperature in the graphs.

There were two sources of uncertainty, one in the temperature associated with each thermopower measurement, and the other in the slope of the line from the plot of sample voltage vs temperature gradient. The sample temperature is the average temperature of the sample over the gradient. The gradient was approximately 10°K.

Therefore the uncertainty in the sample temperature is ±5°K. As for the uncertainty of the slope, the majority of the straight line fits had correlation coefficients of .98 or better.

Table 1. Data for disordered and annealed $Cu_{0.83}Pd_{0.17}$

| Sample | Average °K Temperature | Slope μV/°K | Correction for Cu μV/°K | Thermopower Sample μV/°K |
|---|---|---|---|---|
| $Cu_{0.83}Pd_{0.17}$ | 160 | 3.13 | 1.125 | -2.00 |
| Disordered | 178 | 3.34 | 1.18 | -2.16 |
| | 183 | 3.44 | 1.21 | -2.23 |
| | 193 | 3.50 | 1.26 | -2.24 |
| | 207 | 3.85 | 1.34 | -2.51 |
| | 208 | 3.65 | 1.34 | -2.31 |
| | 233 | 3.97 | 1.48 | -2.49 |
| | 246 | 4.46 | 1.54 | -2.92 |
| | 263 | 4.18 | 1.63 | -2.55 |
| | 282 | 4.97 | 1.74 | -3.23 |
| | 283 | 4.55 | 1.74 | -2.81 |
| | 300 | 4.77 | 1.83 | -2.94 |
| | 318 | 5.66 | 1.93 | -3.73 |
| | 327 | 5.03 | 1.98 | -3.05 |
| | 184 | 3.37 | 1.21 | -2.16 |
| | 216 | 4.07 | 1.38 | -2.69 |
| | 260 | 4.88 | 1.62 | -3.26 |
| | 285 | 5.12 | 1.76 | -3.36 |
| | 324 | 5.19 | 1.97 | -3.22 |
| | 364 | 6.46 | 2.19 | -4.27 |
| $Cu_{0.83}Pd_{0.17}$ | 152 | 2.26 | 1.12 | -1.14 |
| Annealed | 195 | 2.55 | 1.27 | -1.28 |
| | 223 | 2.78 | 1.42 | -1.36 |
| | 251 | 2.89 | 1.57 | -1.32 |
| | 285 | 2.99 | 1.76 | -1.23 |
| | 313 | 3.20 | 1.91 | -1.29 |
| | 352 | 3.09 | 2.12 | -0.97 |
| | 381 | 3.37 | 2.28 | -1.09 |

Table 2. Data of ordered and disordered $\underline{Cu_3AuMn}_{.01}$

| Sample | Average °K Temperature | Slope μV/°K | Correction for Cu μV/°K | Thermopower Sample μV/°K |
|---|---|---|---|---|
| $\underline{Cu_3AuMn}_{.01}$ Ordered | 163 | 2.57 | 1.13 | -1.44 |
| | 189 | 2.75 | 1.24 | -1.51 |
| | 217 | 3.04 | 1.39 | -1.65 |
| | 247 | 3.30 | 1.55 | -1.75 |
| | 279 | 3.40 | 1.72 | -1.68 |
| | 304 | 3.35 | 1.86 | -1.49 |
| | 339 | 3.66 | 2.05 | -1.61 |
| | 382 | 3.74 | 2.29 | -1.45 |
| $\underline{Cu_3AuMn}_{.01}$ Disordered | 162 | 1.10 | 1.13 | +0.03 |
| | 191 | 1.09 | 1.25 | +0.16 |
| | 223 | 1.17 | 1.42 | +0.25 |
| | 246 | 1.18 | 1.54 | +0.26 |
| | 282 | 1.22 | 1.74 | +0.52 |
| | 294 | 1.34 | 1.80 | +0.46 |
| | 334 | 1.36 | 2.02 | +0.66 |
| | 373 | 1.41 | 2.24 | +0.83 |

Table 3. Data for $Cu_3Au$, ordered and disordered.

| Sample | Average °K Temperature | Slope μV/°K | Correction for Cu μV/°K | Thermopower Sample μV/°K |
|---|---|---|---|---|
| $Cu_3Au$ | 155 | 0.93 | 1.12 | +0.19 |
|  | 170 | 0.98 | 1.15 | +0.17 |
| Disordered | 208 | 1.00 | 1.34 | +0.34 |
|  | 230 | 1.18 | 1.46 | +0.28 |
|  | 261 | 1.21 | 1.63 | +0.42 |
|  | 287 | 1.42 | 1.77 | +0.35 |
|  | 320 | 1.50 | 1.95 | +0.45 |
|  | 346 | 1.55 | 2.09 | +0.54 |
|  | 379 | 1.69 | 2.27 | +0.58 |
| $Cu_3Au$ | 138 | 1.80 | 1.13 | -0.67 |
|  | 173 | 2.13 | 1.16 | -0.97 |
| Ordered | 201 | 2.07 | 1.30 | -0.77 |
|  | 226 | 2.53 | 1.44 | -1.04 |
|  | 260 | 2.72 | 1.62 | -1.10 |
|  | 282 | 3.01 | 1.74 | -1.27 |
|  | 318 | 3.39 | 1.93 | -1.46 |
|  | 350 | 3.86 | 2.11 | -1.75 |
|  | 374 | 4.35 | 2.24 | -2.11 |

Table 4. Thermopower for $\underline{\mathrm{CuPd}}*\mathrm{Mn}_{.022}$ and $\underline{\mathrm{CuPd}}*\mathrm{Mn}_{.010}$

| Sample | Average °K Temperature | Slope μV/°K | Correction for Cu μV/°K | Thermopower Sample μV°K |
|---|---|---|---|---|
| $\underline{\mathrm{CuPd}}*\mathrm{Mn}_{.022}$ | 148 | 3.03 | 1.12 | -1.91 |
| | 183 | 3.84 | 1.21 | -2.63 |
| | 212 | 4.14 | 1.36 | -2.78 |
| | 232 | 4.85 | 1.47 | -3.38 |
| | 263 | 4.70 | 1.64 | -3.03 |
| | 293 | 4.88 | 1.80 | -3.08 |
| | 333 | 5.82 | 2.02 | -3.80 |
| | 350 | 4.86 | 2.11 | -2.75 |
| $\underline{\mathrm{CuPd}}*\mathrm{Mn}_{.010}$ | 164 | 3.12 | 1.14 | -1.98 |
| | 190 | 3.18 | 1.25 | -1.93 |
| | 227 | 3.45 | 1.44 | -2.01 |
| | 266 | 3.95 | 1.65 | -2.30 |
| | 287 | 4.26 | 1.77 | -2.49 |
| | 314 | 4.84 | 1.91 | -2.93 |
| | 337 | 4.95 | 2.04 | -2.91 |
| | 357 | 5.79 | 2.15 | -3.64 |

Table 5. Thermopower of $\underline{\text{Cu}}\text{Mn}_{.01}$, $\underline{\text{Ag}}\text{Mg}$, and $\underline{\text{Al}}\text{Ag}_{0.1}$

| Sample | Average °K Temperature | Slope μV/°K | Correction for Cu μV/°K | Thermopower Sample μV/°K |
|---|---|---|---|---|
| $\underline{\text{Cu}}\text{Mn}_{.01}$ | 188 | -0.91 | 1.24 | +2.15 |
| | 227 | -1.20 | 1.44 | +3.64 |
| | 259 | -1.82 | 1.62 | +3.44 |
| | 288 | -2.60 | 1.77 | +4.37 |
| | 300 | -3.68 | 1.84 | +5.52 |
| | 356 | -2.55 | 2.14 | +4.69 |
| | 373 | -2.34 | 2.24 | +4.58 |
| $\underline{\text{Ag}}\text{Mg}$ | 139 | 0.50 | 1.13 | +0.63 |
| | 166 | 0.52 | 1.14 | +0.62 |
| | 196 | 0.51 | 1.27 | +0.76 |
| | 222 | 0.52 | 1.42 | +0.90 |
| | 254 | 0.58 | 1.59 | +1.01 |
| | 288 | 0.70 | 1.77 | +1.07 |
| | 328 | 0.67 | 1.99 | +1.32 |
| | 350 | 0.72 | 2.11 | +1.39 |
| | 380 | 0.69 | 2.28 | +1.59 |
| $\underline{\text{Al}}\text{Ag}_{0.1}$ | 168 | 2.34 | 1.14 | -1.20 |
| | 201 | 2.55 | 1.30 | -1.25 |
| | 236 | 3.20 | 1.49 | -1.71 |
| | 265 | 3.25 | 1.65 | -1.60 |
| | 274 | 3.40 | 1.70 | -1.70 |
| | 307 | 3.26 | 1.87 | -1.39 |
| | 339 | 3.43 | 2.05 | -1.38 |
| | 374 | 3.61 | 2.17 | -1.44 |

Table 6. Data for thermopower of $(Cu_{.83}Pd_{.17})_{1-c}Ni_c$

| Sample | Average °K Temperature | Slope μV/°K | Correction for Cu μV/°K | Thermopower Sample μV/°K |
|---|---|---|---|---|
| $(Cu_{.83}Pd_{.17})$ $Ni_{.025}$ | 166 | 6.31 | 1.14 | -5.17 |
| | 196 | 7.38 | 1.27 | -6.11 |
| | 230 | 8.45 | 1.46 | -6.99 |
| | 251 | 9.32 | 1.58 | -7.74 |
| | 270 | 10.09 | 1.68 | -8.41 |
| | 305 | 11.39 | 1.86 | -9.53 |
| | 315 | 11.43 | 1.91 | -9.52 |
| $(Cu_{.83}Pd_{.17})$ $Ni_{.05}$ | 168 | 8.56 | 1.14 | -7.42 |
| | 180 | 8.95 | 1.20 | -7.75 |
| | 204 | 9.77 | 1.32 | -8.45 |
| | 228 | 11.19 | 1.45 | -9.74 |
| | 273 | 13.47 | 1.69 | -11.78 |
| | 299 | 15.16 | 1.83 | -13.33 |
| | 300 | 14.72 | 1.83 | -12.89 |
| | 323 | 14.74 | 1.96 | -12.78 |
| | 325 | 14.73 | 1.97 | -12.76 |
| | 351 | 17.43 | 2.12 | -15.31 |
| | 386 | 24.57 | 2.31 | -22.26 |
| $(Cu_{.83}Pd_{.17})$ $Ni_{.10}$ | 163 | 14.03 | 1.13 | -12.90 |
| | 192 | 15.28 | 1.26 | -14.03 |
| | 211 | 17.18 | 1.355 | -15.83 |
| | 234 | 19.70 | 1.48 | -18.22 |
| | 282 | 21.81 | 1.74 | -20.07 |
| | 297 | 21.54 | 1.82 | -19.72 |
| | 323 | 21.31 | 1.96 | -19.35 |
| | 350 | 21.74 | 2.11 | -19.63 |
| | 270 | 20.73 | 1.675 | -19.06 |

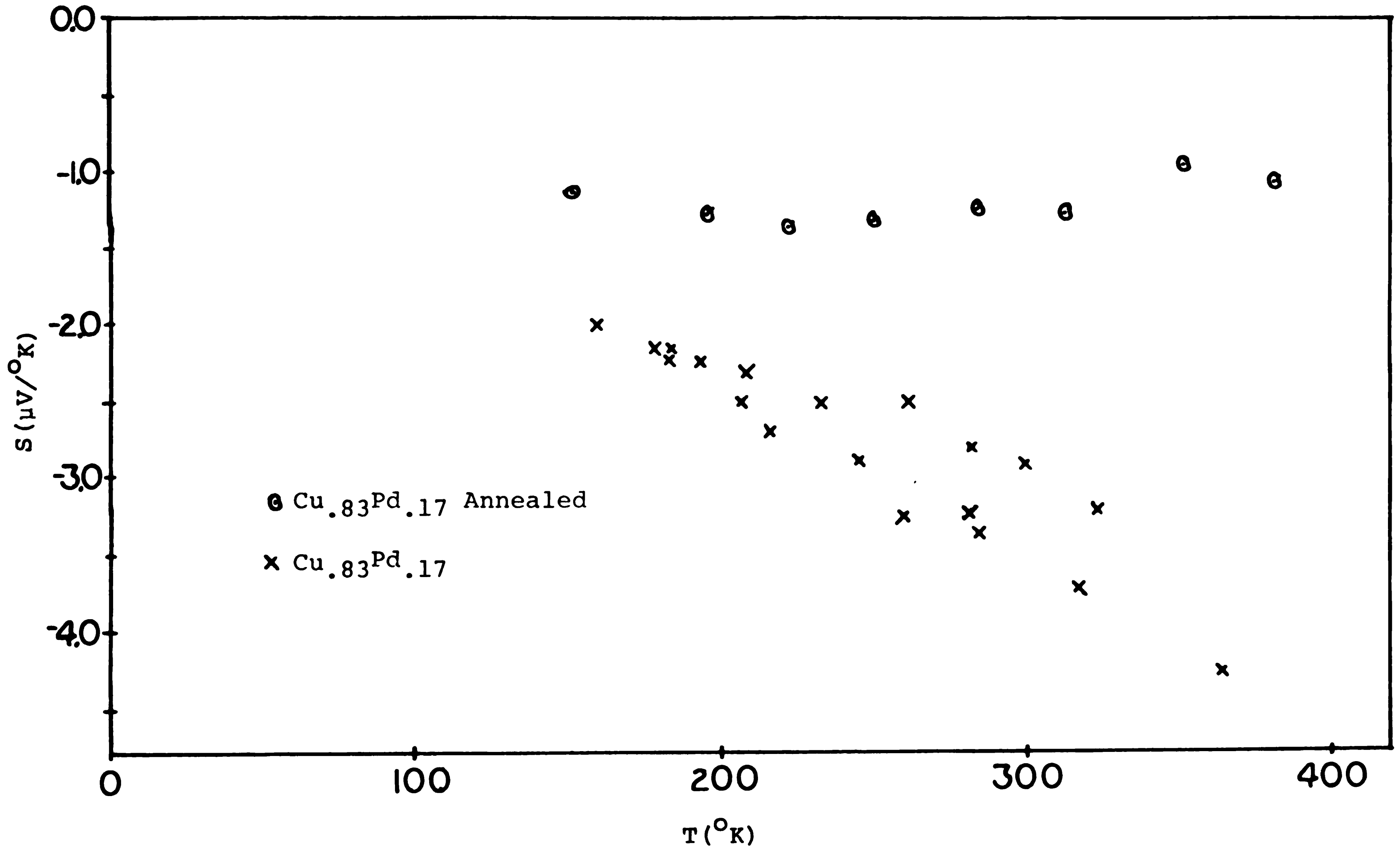

Figure 4 Thermopower, Effects of Atomic Ordering

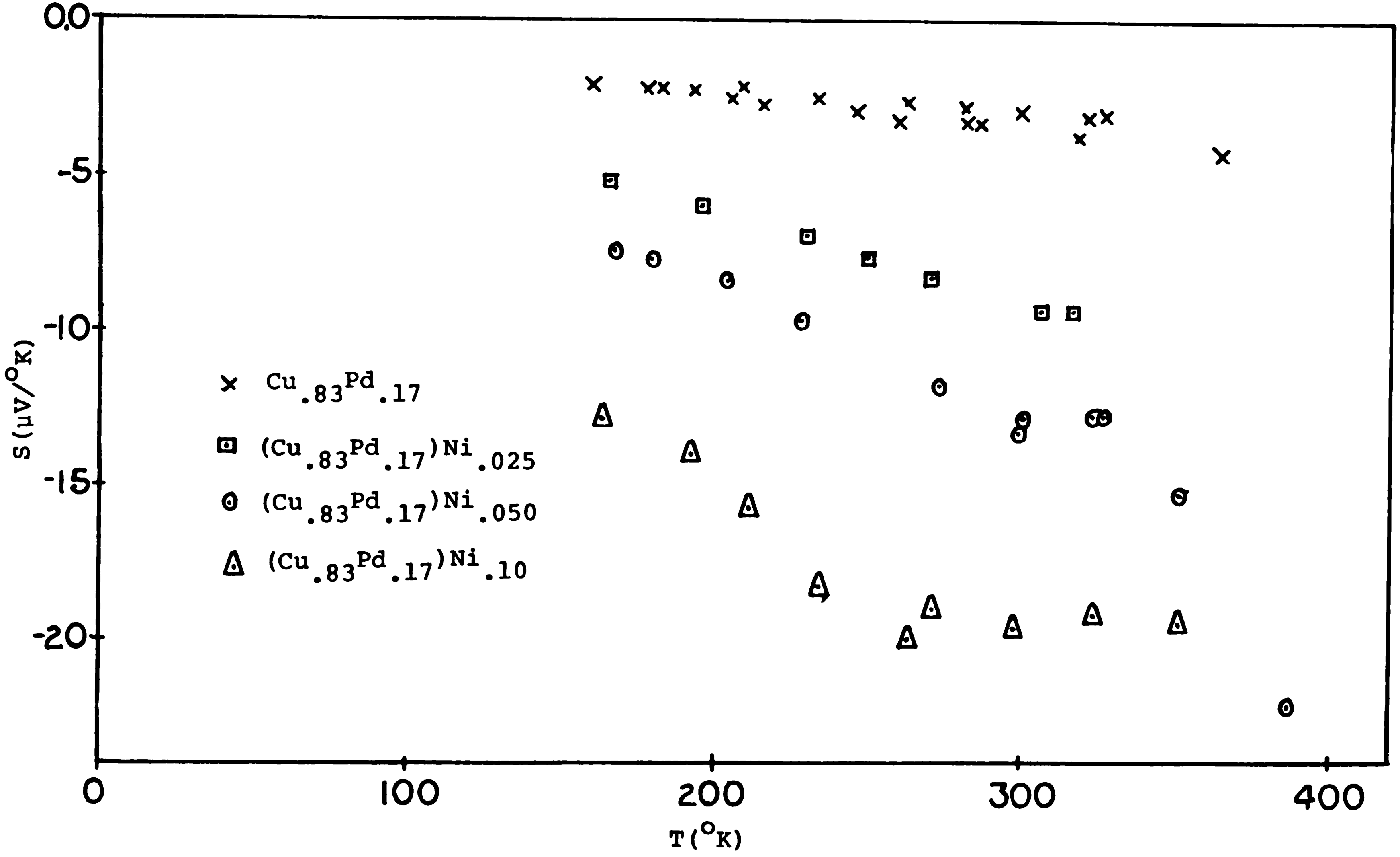

Figure 5 Thermopower, Effects of Ni Concentration

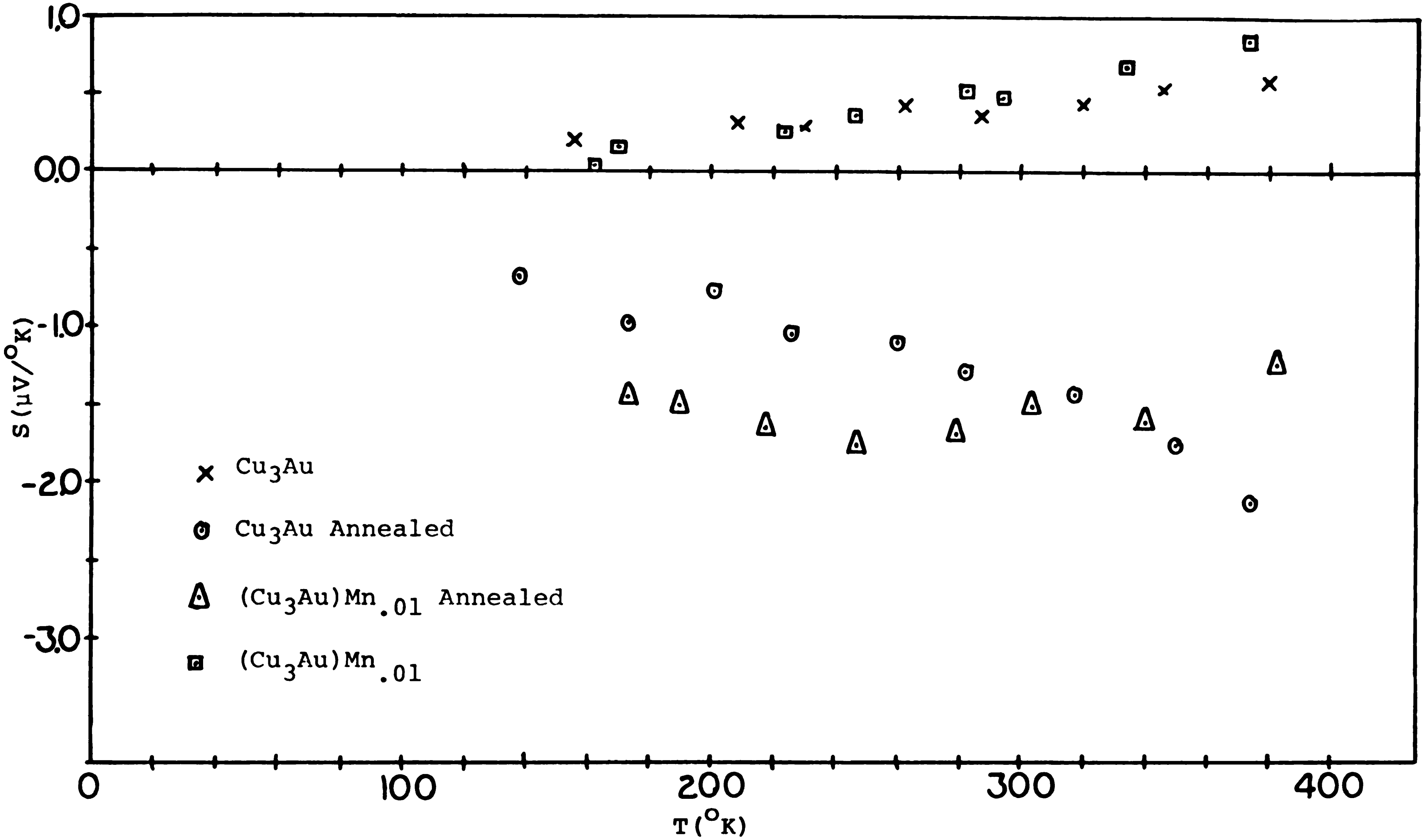

Figure 6 Thermopower, Effects of Atomic Ordering and Concentration of Mn

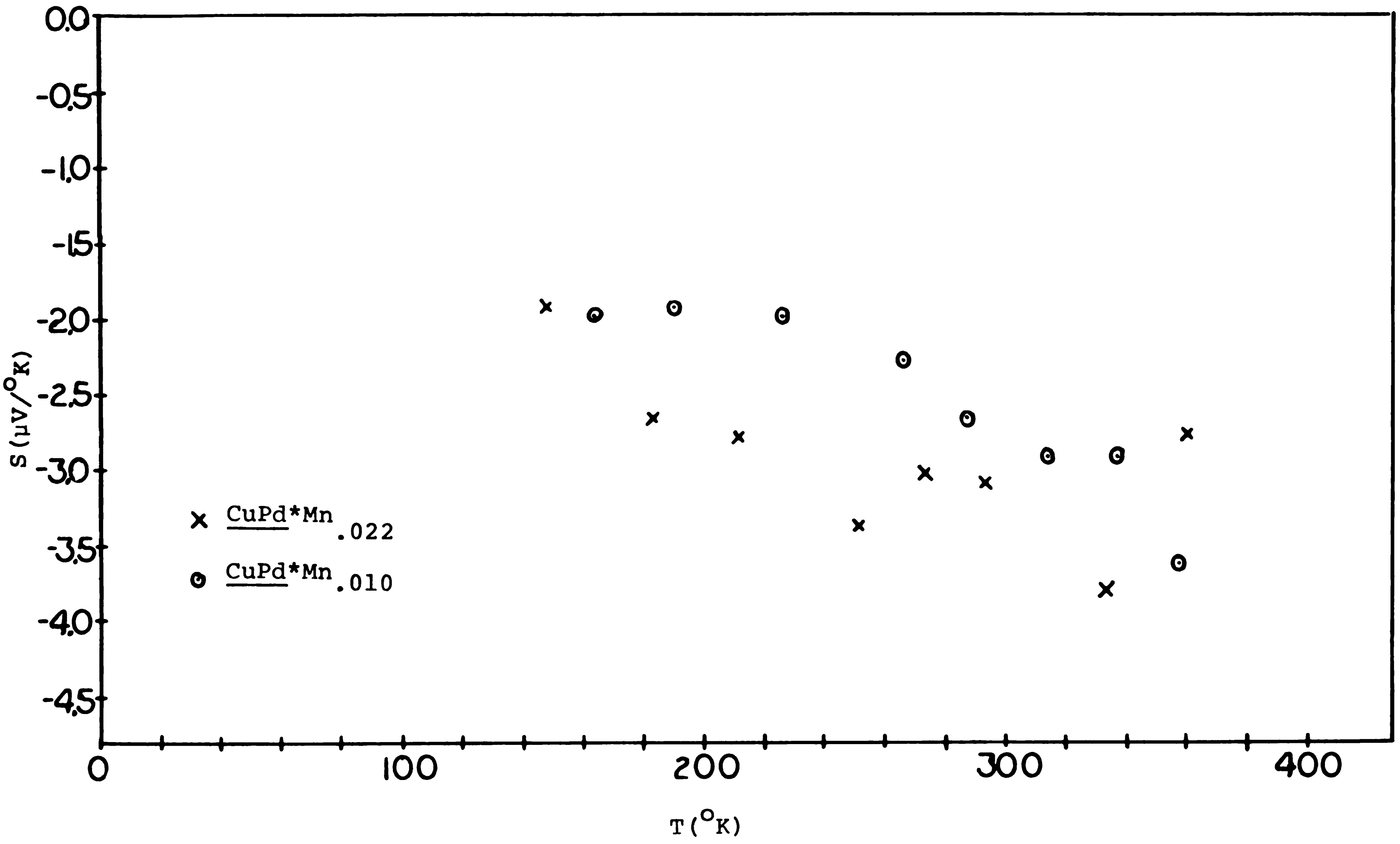

Figure 7 Thermopower, Effects of Impurity Concentration

## SECTION III

## CONCLUSION

There is an interest in measuring the Fermi surface of $Cu_{0.83}Pd_{0.17}$ both in the ordered and disordered states because there is a distinct change between these two states. $Cu_3Au$ also shows an order disorder transition like that of $Cu_{0.83}Pd_{0.17}$. Because of their similarities, and there are previous data available for $Cu_3Au$,(2) it shall also be discussed. Both of these materials have a low conductivity so that a direct measurement of the Fermi surface is not possible. Because a direct measurement is not possible, comparison of the transport and equilibrium properties is an indirect means of measuring the Fermi surface. Use of the preceding thermopower data will be considered now.

A comparison of $Cu_{0.83}Pd_{0.17}$ and $Cu_3Au$ can be made with Cu. In the disordered state $Cu_{0.83}Pd_{0.17}$ and $Cu_3Au$ have the same lattice structure as does Cu, face centered cubic. When $Cu_{0.83}Pd_{0.17}$ and $Cu_3Au$ order, they have a simple cubic lattice with a basis of four atoms. When $Cu_{0.83}Pd_{0.17}$ and $Cu_3Au$ are disordered properties similar to Cu would be expected because all three have the same lattice structure and nearly the same number of conduction electrons. In the ordered state $Cu_{0.83}Pd_{0.17}$ and $Cu_3Au$ have the simple cubic structure, new Brillouin zones are drawn and a change in the thermopower would be expected.

Figure 8 verifies this prediction for it shows the $Cu_3Au$ results in the disordered state to be similar to Cu and shows a change in the thermopower for the ordered state. The disordered state of $Cu_3Au$ has a thermopower similar in sign, both positive, and magnitude to Cu. When $Cu_3Au$ is ordered the sign of the thermopower reverses. Notice, however, that the disordered state of $Cu_{0.83}Pd_{0.17}$ has a negative thermopower and the ordered state has a positive thermopower, Figure 9. The results for $Cu_{0.83}Pd_{0.17}$ would have been expected to be similar to those of $Cu_3Au$ instead of the mirror image as seen in Figure 9. The discrepancy in the signs of the thermopower may result from an insufficiency in the preceding analysis.

To resolve or confirm this discrepancy the total thermopower of a sample must be interpreted as the weighted sum of its components(4)

$$W_t S_t = \sum_k W_k S_k \qquad (1)$$

$S_t$ is the total thermopower of the sample, $W_t$ is the total thermal resistivity of the sample, and $W_k S_k$ are the thermopower and thermal resistivity of the various scattering processes. The sum is over k, all possible processes. In this analysis two terms of the series were used, a lattice vibration contribution and a residual scattering contribution. The electrical resistivity for the samples was available therefore, by the Wiedemann-Franz relation, the electrical resistivity can be substituted for the thermal resistivity in Equation (1). With these substitutions Equation (1) now becomes

$$\rho_t S_t = \rho_1 S_1 + \rho_r S_r \quad . \qquad (2)$$

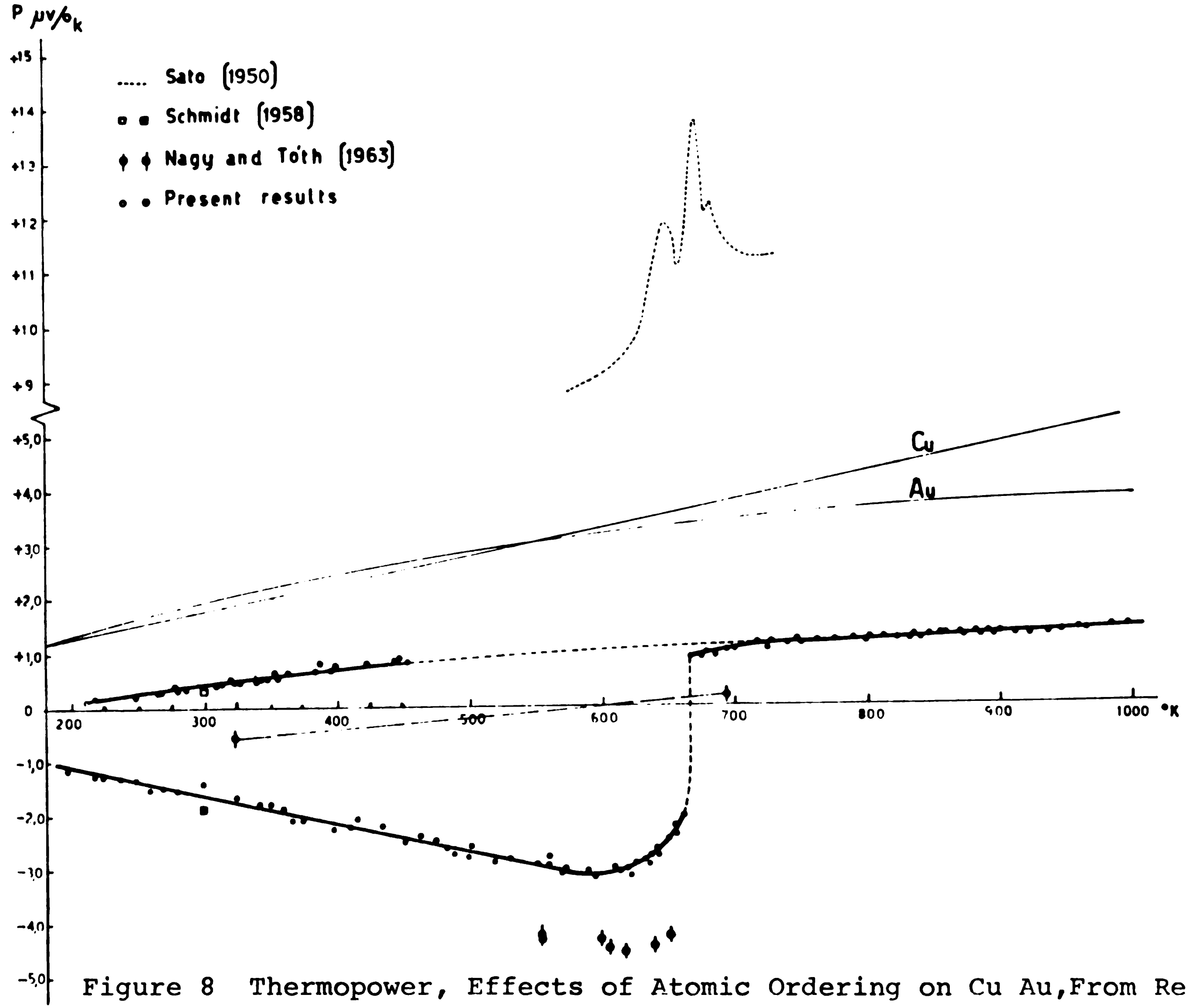

Figure 8 Thermopower, Effects of Atomic Ordering on Cu Au,From Reference #2

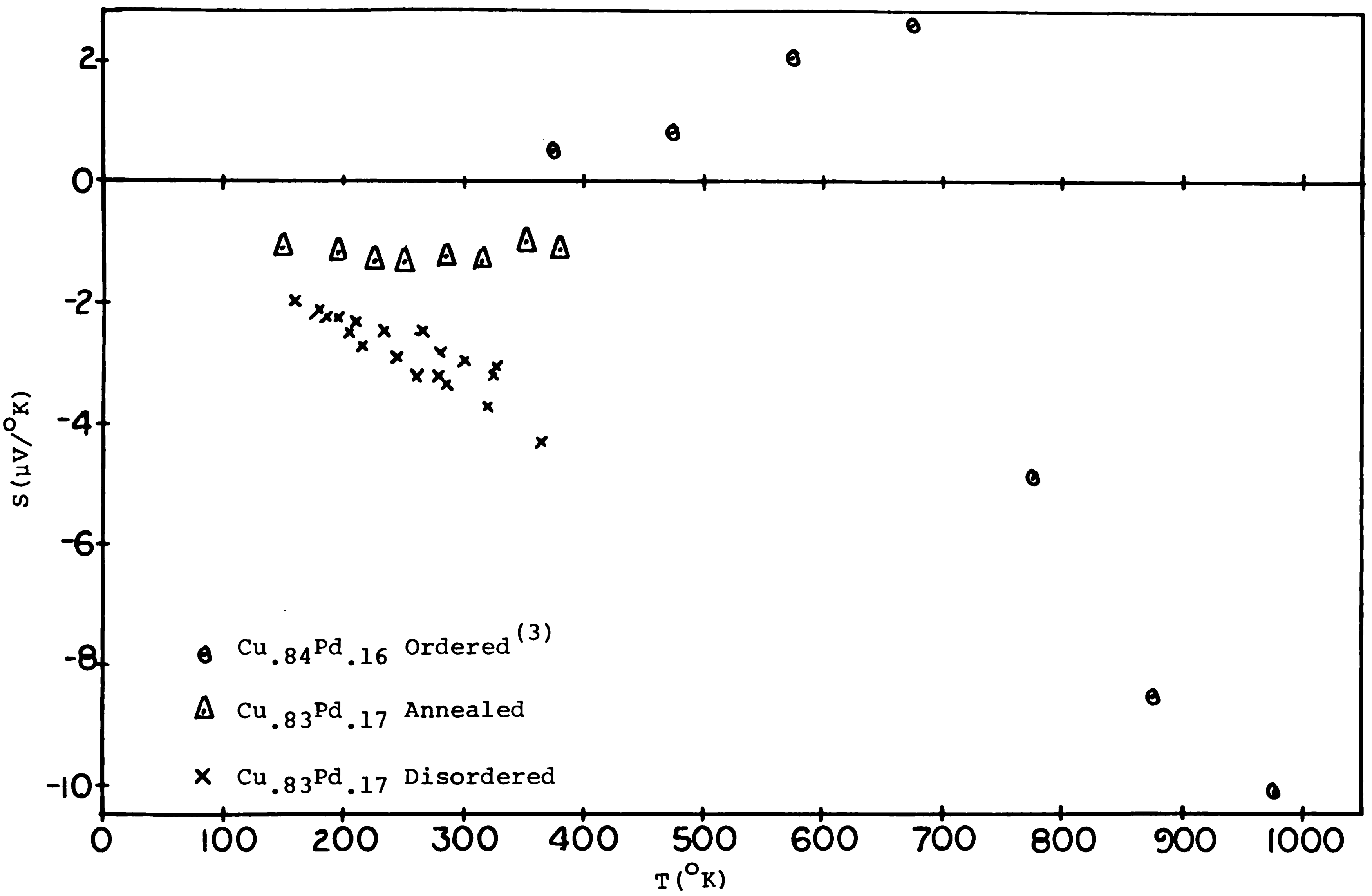

Figure 9 Thermopower, Effects of Atomic Ordering on $Cu_{.83}Pd_{.17}$

In this way the individual contributions to the thermopower can be identified and studied.

To obtain the individual thermopower contributions from a set of terms that are temperature dependent, assumptions need to be made about this dependence. General experience has shown that an adequate first order model assumes that $\rho_1$, $S_1$ and $S_r$ are proportional to temperature and that $\rho_r$ is a constant$^{(5)}$. If these relations are substituted into Equation (2) and the equation is divided by temperature, T, the following relation is obtained

$$\frac{\rho_t S_t}{T} = AT + B \quad . \qquad (3)$$

In equation (3), A and B are proportionality constants where A is related to the lattice vibration contribution and B to the residual scattering contribution. This result can then be analyzed with the data presented earlier in this paper.

If the assumptions leading to Equation (3) have been valid, then the graph of $\rho_t S_t/T$ vs T should give a straight line. Figure 10 shows such a plot for $Cu_{0.83}Pd_{0.17}$ in both ordered and disordered states. These plots do appear linear. (Similar linear plots occur for $Cu_3Au$ and Cu.) The results of analyzing the slopes and intercepts of plots for $Cu_3Au$, both ordered and disordered, $Cu_{0.83}Pd_{0.17}$, ordered, disordered, and annealed, and Cu are given in Table 7. With the exception of the values of Cu, all the $S_r/T$ values are good to about ±10% and all $S_\ell/T$ values are good to about ±20%. The results for Cu are obtained both by the method described in this paper and, in parenthesis, by conventional means. For $Cu_3Au$, the total

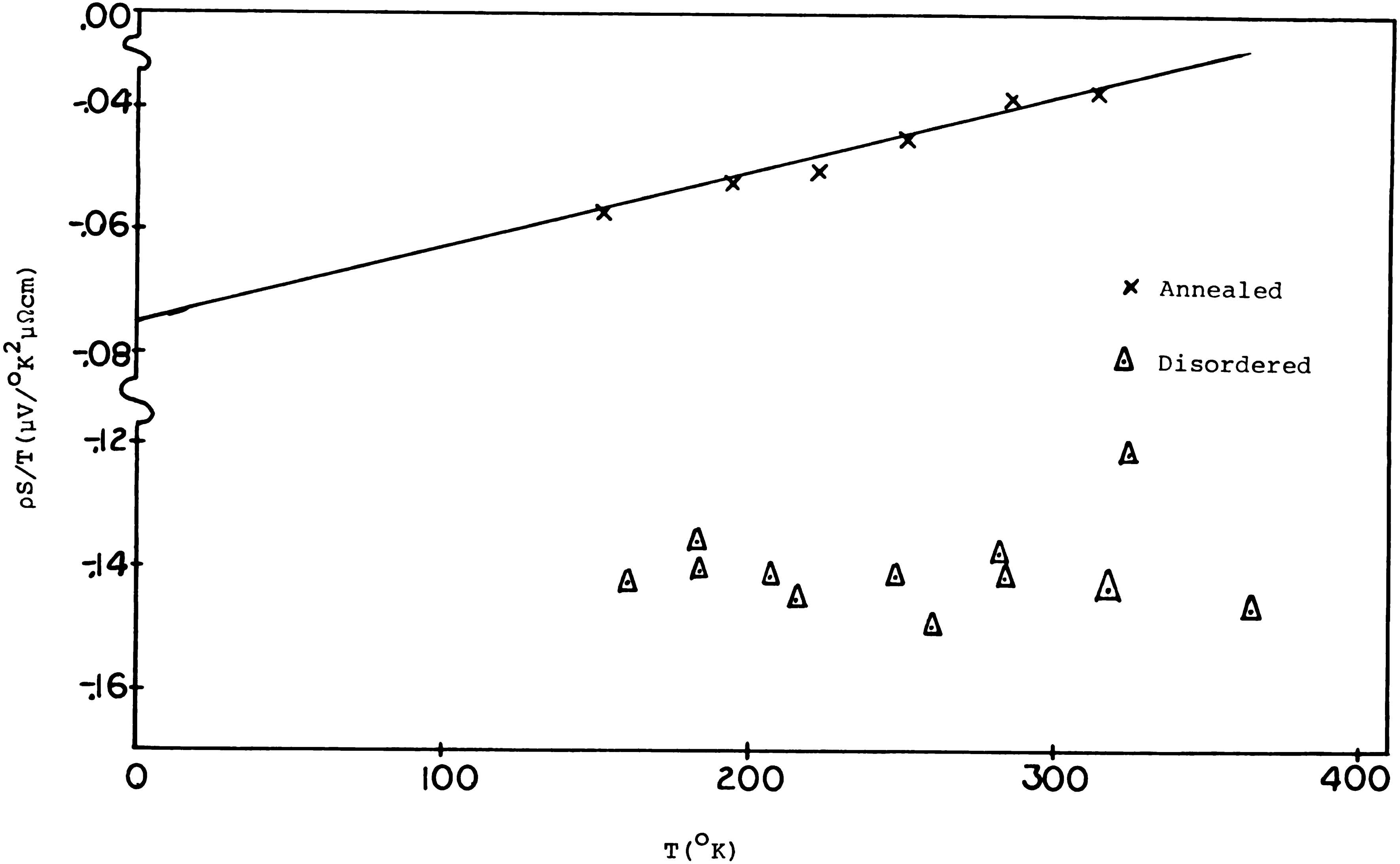

Figure 10 Separation of the Total Thermopower of $Cu_{.83}Pd_{.17}$

Table 7. Results of the separation of thermopower

| Sample | | $S_e/T$ nV/°$K^2$ | $S_n/T$ nV/°$K^2$ |
|---|---|---|---|
| $Cu_3Au$ | Ordered | 2.8 | -8.5 |
| | Disordered | 14 | -2.1 |
| $Cu_{0.83}Pd_{0.17}$ | Ordered | 6 | 8 |
| | Annealed | 14 | -11 |
| | Disordered | 0 | -13 |
| Cu | | 6.3<br>(5.5) | -0.7 to -6.0 |

thermopower for the disordered is positive and goes negative when ordered. During this change the lattice vibration contribution, $S_\ell$ decreases but remains positive while the residual contribution becomes more negative. When $Cu_{0.83}Pd_{0.17}$ makes the transition from disordered to ordered the thermopower goes from negative to positive. The lattice vibration contribution increases, while always remaining positive in making the transition from the disordered state to the ordered state. The residual contribution becomes less negative for the transition from disordered to ordered state.

Thus in both $Cu_3Au$ and $Cu_{0.83}Pd_{0.17}$, the lattice vibration contribution to the thermopower remains positive, and similar to the value of Cu, even though the sign of the total thermopower changed. Therefore, the fact that the total thermopowers of the $Cu_3Au$ and $Cu_{0.83}Pd_{0.17}$ systems are mirror images of each other is not associated with a sign change in the lattice vibration contribution but is the result of competition between different terms.

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