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Anomalies in the low-temperature thermal and electrical resistivities of silver

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Abstract. Very accurate measurements of the thermal conductivity of pure silver (RRR = 1912) reveal abrupt changes in the temperature dependence of the inelastic electron-phonon scattering component of the thermal resistivity W_v at about 5, 7 and 12.8 K. Similar changes in the temperature dependence of the electrical resistivity of silver are found at about 4, 7 and 12 K. These results are consistent with previous results found in the electrical and thermal resistivities of Cu and Au and they remain unexplained.

1. Introduction

The electrical resistivity of a pure metal sample may be represented by the equation

$$\rho = \rho_0 + \rho_i + \rho_d + \rho_m \tag{1}$$

where ρ is the measured value of the electrical resistivity, ρ_0 is the residual resistivity, $\rho_{\rm i}$ is the ideal resistivity, $\rho_{\rm d}$ represents deviations from Matthiessen's rule and $\rho_{\rm m}$ is due to the scattering of electrons by magnetic impurities (the Kondo effect). In experiments carried out to obtain an improved understanding of the transport of electrons in metals, the electrical resistivity of eight samples of high-purity Ag with varying defect concentrations (the residual resistivity ratio varied from 2.1×10^3 to 1.4×10^2) were measured (Kos 1973a). Since the Kondo temperature of Ag is believed to be so low that $\rho_{\rm m}$ is essentially zero for measurements above 1 K, it was not included in the analysis of data, and indeed no evidence of a resistance minimum appeared in the results. An analysis of the data revealed that, at any given temperature within the range of measurements (1.5–300 K), ρ_d could be represented approximately as a linear function of ρ_0 , permitting the construction of linear graphs of $\rho_i + \rho_d = \rho - \rho_0$ as a function of ρ_0 which, extrapolated to $\rho_0 = 0$, yielded approximate values of $\rho_i(T)$ and $\rho_d(\rho_0, T)$. At that time it was surprising that $\rho_i(T)$ did not vary as T^5 , as predicted by the Bloch theory, and even more surprising that a graph of $\ln \rho_i$ versus $\ln T$ indicated a kink at around 11 K. Below 9 K, ρ_i varied as $T^{4.56\pm0.06}$ and, above 12 K, ρ_i varied as $T^{4.88\pm0.02}$. If this result were real, one could expect to find a corresponding discontinuity in the'thermal conductivity data for Ag.

Evidence of a discontinuity was found (Kos 1973b) in the thermal conductivity data of Van Baarle *et al* (1966) and in the data of Fenton *et al* (1963). Independent measurements by Ehrlich and Schriempf (1974) also indicated a discontinuity at about 11 K in both the electrical resistivity and the thermal conductivity of Ag. Subsequent

accurate measurements of the electrical resistivity of copper (Moussouros and Kos 1977) and of gold (Kos and Barton 1979) revealed several such discontinuities in both at different temperatures and showed that the T^5 law did not apply in any region above 2 K. Excellent experimental work by numerous workers (Rumbo 1973, 1976, Ford *et al* 1974, Barber and Caplin 1975, Barnard and Caplin 1977, Barnard *et al* 1981, 1982, Rowlands and Woods 1975) as well as by others confirmed the breakdown of the T^5 law but none observed discontinuities.

Astute theoretical work (Rowlands and Woods 1975, Bergmann *et al* 1982a, b, Kaveh and Wiser 1984, 1986) and others (references in the above-listed publications) reconciled most of the experimental results with accepted theoretical ideas; however, the observed discontinuities remained unexplained.

Accurate measurements of the thermal conductivity of gold (Kos 1985a) and of copper (Kos 1985b) on pieces of the same material used in the electrical resistivity measurements revealed the existence of kinks and regions which corresponded accurately to those observed in the electrical resistivity. These were important confirmations of the former work in that the effects observed in the resistivity might have been attributed to artifacts of the apparatus. A miscalibration of thermometers was highly unlikely because two thermometers were used for all the resistivity measurements. One was a platinum resistance secondary reference thermometer calibrated at the National Research Council of Canada, the other was a Cryo Cal germanium resistance thermometer calibrated by the manufacturer and their intercalibration was within expected tolerances. The appearance of corresponding anomalies in the thermal conductivity data obtained by using an entirely different apparatus of very different design and different thermometers provided compelling evidence for the existence of the observed discontinuities.

The analysis of the thermal conductivity data is guided by standard theory (Klemens and Jackson 1964) which relates the low-temperature thermal resistivity of a monovalent metal to the electrical resistivity through the expression

$$W = \rho_0 / L_0 T + \rho_i / L_0 T + \alpha T^2$$
(2)

where the first two terms derive from the Wiedemann–Franz law and represent 'horizontal processes', due to elastic electron–phonon scattering, whereas

$$W_{\rm v} = \alpha T^2 = (64.0/497.6)(\Theta_{\rm D}/T)^2(\rho_{\rm n}/L_0T)$$
(3)

represents 'vertical processes' in which inelastic electron-phonon scattering changes the energy of the scattered electron at the Fermi surface, W is the measured thermal resistivity, L_0 is the Sommerfeld value of the Lorenz number, Θ_D is the Debye temperature and ρ_n is the component of the ideal resistivity due to normal electron-phonon scattering, whereas ρ_i includes umklapp processes. Using a geometric model to take into account the fact that the Fermi surface touches the zone boundaries in Cu, Ag and Au, Klemens and Jackson (1964) have shown that $\rho_n \approx \frac{1}{3}\rho_i$. To include deviations from Matthiessen's rule, we may add the term ρ_d/L_0T to equation (2) to obtain

$$W = \rho_0 / L_0 T + \rho_i / L_0 T + \rho_d / L_0 T + \alpha T^2 = W_0 + W_i + W_d + W_v.$$
(4)

 W_0 and W_v are the dominant terms at low temperatures and the other two are often neglected; however, in Ag they contribute about 0.5% of the value of W at 4 K, 5% at 8 K, 10% at 11 K and 20% at 15 K and therefore should always be included in accurate work.



Figure 1. Diagram showing how leads L are connected to the sample S and how the effective length l of the sample is measured.

Theoretical work on the thermal conductivity of the noble metals proceeded slowly. Despite the inclusion of umklapp processes in their calculations, Klemens and Jackson (1964) obtained values of α which still disagreed with experiment. Geîlikman and Orlov (1976) took into account the non-equilibrium nature of the phonon distribution and attributed the deviation of ρ_i from a T⁵-dependence and the deviation of W from a T²dependence to normal processes on the necks of the Fermi surfaces of the noble metals. They predicted that $\rho_i \propto T^m$ where m < 5 and that $W \propto T^n$ where n > 2. Calculations for Cu and Ag by Borchi et al (1979) using a variational method for two different pseudopotentials showed that W does not obey a well defined power law. They suggested that, if one assumes a simple relation $W \propto T^n$ then, between 2 and 15 K, using the Moriarity pseudopotential, *n* ranges smoothly from n = 2.3 to n = 1.66 for Cu and from n = 2.05 to n = 1.67 for Ag and, using the Nand *et al* pseudopotential, *n* ranges smoothly from n = 2.16 to n = 1.79 for Cu and n = 2.03 to n = 1.69 for Ag. Suslov (1981), retaining Bloch's hypothesis, obtained an exact expression for the electronic thermal conductivity of Cu, Ag and Au which in the pure and low-temperature limits predicted that W_{y} = αT^2 with α (Cu) = 1.3 × 10⁻⁷ m W⁻¹ K⁻¹, α (Ag) = 2.7 × 10⁻⁷ m W⁻¹ K⁻¹ and α (Au) = $2.1 \times 10^{-7} \text{ m W}^{-1} \text{ K}^{-1}$.

In this paper we present the results of accurate measurements of the thermal conductivity of Ag and in view of these data we review our previous measurements of the electrical resistivity of Ag.

2. Experimental procedure

Very accurate measurements of the thermal conductivity of a silver wire with an effective length l of 16.818 cm and a diameter of 5.036×10^{-2} cm were taken using the standard steady-state linear heat flow method. The silver sample was a piece of the wire designated as sample A2 for which electrical resistivity measurements were previously reported (Kos 1973a). Its total impurity content was listed as 1.4 ppm. Its effective length was measured to about one part in 10^4 using a travelling microscope. Its cross sectional area (hence its diameter) was determined by weighing the sample and using the value of 10.50 for the specific gravity of Ag at 20 °C (*Handbook of Chemistry and Physics* 1975). The uncertainty in density therefore limits the absolute accuracy of our data to about one part in 10^3 .

The thermal (and electrical) leads were soldered to hair-thin slivers cut from (but still attached to) the sample with a scalpel under the microscope (figure 1, where S and

L refer to sample and leads respectively). These very small connections to the sample permitted an accurate measurement of its effective length. The leads to the thermometers were made of annealed high-purity silver wire 0.25 mm in diameter taken from the same batch as used in our samples. These wires were wrapped around and soldered to a copper thermometer holder. The germanium thermometers were inserted into copper holders and their leads were attached to insulated copper wires which were wrapped around and firmly cemented to the copper holder. The other ends of the copper wires were soldered to leads made of manganin 290 wire 2.5×10^{-2} mm in diameter coated with a superconducting layer of 60–40 tin–lead about 10^{-3} mm thick and then insulated with H-Formvar. These leads were then passed to the top of the sample chamber where they were connected to number 40 insulated copper wires which were thermally anchored and passed out of the cryostat to the measuring system through feed-through seals. The copper thermometer holders were suspended within the sample chamber from nylon threads about 6 cm long and 0.15 mm thick.

The ends of the silver sample were soldered to a heat source and heat sink. The heat source was a copper cylinder about 3.0 cm in length and 2.0 cm in diameter. It was suspended from the sample chamber by the same nylon threads used to suspend the thermometer holders. A measured heat input into the heat source was provided by passing a manually selected current either through a 7.3 k Ω wire-wound disk heater or through a 100 k Ω film resistor (both of which were firmly attached to the heat source using indium contacts). The current leads to these heaters were made of 30 cm lengths of niobium–titanium (T48B) superconducting wire 12.5×10^{-2} mm in diameter coated with a copper layer 2×10^{-2} mm thick and insulated with Formvar. The voltage leads were made of the same tin–lead-coated manganin as used for the thermometer leads. The current through the heater was measured by passing it through a 2500 Ω standard resistor (at room temperature) and measuring the voltage across the selected heater (in four-terminal configuration) within 0.5 s. The calculations of power input was carried out on line by the same computer which controlled the Datron voltmeter.

The heat sink was a copper cylinder about 4 cm long and 5 cm in diameter. It was suspended by a stainless steel tube 1 in in diameter from the main top flange of the vacuum chamber which was immersed in liquid helium.

Thermal links could be provided by attaching pieces of high-purity copper wire 1 mm in diameter and/or pieces of high-purity indium wire 1 mm in diameter between the flange and the heat sink. In addition, a mechanical thermal switch operated from outside the cryostat could be used to adjust the heat flow between the flange and the heat sink. The heat sink also contained a germanium thermometer and a wire-wound disk heater. Its temperature could be accurately controlled by adjusting the power input into the disk heater, either manually or by computer control of an Electronic Development Corporation programmable DC calibrator.

Temperatures were measured using Cryocal 1000 germanium thermometers selected by the manufacturer to have very similar resistance versus temperature characteristics and calibrated by the manufacturer. Their resistances were measured in four-terminal configuration using a computer-controlled Datron Autocal 1071 digital (seven-digit) multimeter with an effective input resistance greater than $10^{10} \Omega$ and a read rate of about 8 s on the 0.1 V range. Inputs from the various thermometers, heater, etc, were switched into the Datron voltmeter using a home-made multiplexer (Ramadan and Kos 1990). Calibration and intercalibration data were programmed into the computer which then calculated temperatures on line as data were input from the Datron voltmeter. Although the absolute temperature scale is reliable to somewhat less than 1 mK, in the range of interest, temperatures were read to 0.01 mK and with proper intercalibration corrections obtained during each helium run, reliable *temperature differences* could be measured to about 0.1 mK. The temperature stability so essential to the steadystate method could be achieved to better than 0.1 mK per reading. Once stability at a selected temperature was indicated a series of readings (generally nine to 25) were taken on line under computer control. The time required for the series varied between 12 and 30 min and the temperature drift of the mid-point of the sample, during this time, was generally less than 1 mK but often only 0.2–0.4 mK.

Values of the thermal conductivity were calculated for each reading and after the series was completed graphs of K versus T were printed and a linear least-mean-squares calculation was carried out by the computer to the plotted points. Points (usually two or three out of 16) which were more than about two standard deviations from the calculated line were removed from the data and a new graph with a new least-mean-squares fit was again printed out. If the temperature drift was higher than expected at a given temperature, the temperature was restabilised and another series of readings was taken.

Intercalibration data were obtained by heating the heat sink and therefore the entire sample chamber to the desired temperature and reading ten to 20 values of the indicated temperature differences δ . The large number of values were taken to ensure that temperature stability had been attained and to allow calculation of a good average value of δ at each intercalibration temperature. A graphical interpolation between the intercalibrated points was used to obtain values of δ at the temperatures at which the thermal conductivity values were obtained. This procedure is justified because the values of δ are small—generally only a few tenths of a millikelvin.

A detailed description of the apparatus including on-line computer control and data acquisition techniques is being prepared for publication.

3. Results and analysis of data

Measured values of the temperature T, the thermal conductivity K, the temperature gradient ΔT and the thermometry correction factor δ were used to generate corrected values K_c of K as a function of T. The lowest temperature values of K_c were plotted on a graph and those below 3.25 K (which fell on a straight line) were used in a linear least-mean-squares fit to the equation

$$K_{\rm c} = (L_0/\rho_0)T$$

to determine L_0/ρ_0 and to obtain $\rho_0 = 5.9316 \times 10^{-12} \,\Omega$ m where $L_0 = 2.4430 \times 10^{-8} \,\mathrm{W} \,\Omega \,\mathrm{K}^{-2}$.

Previously published data (Kos 1973a) for $\rho_i(T)$ and $\rho_d(T, \rho_0)$ (appropriate to $\rho_0 = 5.9316 \times 10^{-12} \Omega$ m) were used to calculate $\rho_i + \rho_d$. In the above publication a plot of ln ρ_i as a function of ln T indicated only one discontinuity at about 12 K. However, a better graphical indication of the variation in ρ_i as a function of temperature is obtained in a plot of ρ_i/T^4 as a function of T on log–log paper, which is very similar to the graph in figure 2 where $(\rho_i + \rho_d)/T^4$ is plotted as a function of T and shows discontinuities at about 4.2, 7.5 and 12 K. In each region

$$\rho_{\rm i} + \rho_{\rm d} = JT^k$$

with different values for J and k as given in table 1.



Figure 2. Graphs of W_v/T^2 and $(\rho_i + \rho_d)/T^4$ as functions of temperature on a log-log scale. (The circled points were removed from the data used to carry out the least-mean-squares fit.)

Temperature range (K)	$J imes 10^{18}$ (SI units)	k	$lpha imes 10^8$ (SI units)	n	$3C = (J/\alpha) \times 10^{11}$ (st units)	q = k - n
1.95–4.15 3.73–4.96	39.6 ± 2.7	4.68 ± 0.05	3.07 ± 0.28	3.39 ± 0.06	129	1.29
5.67–7.31 5.67–7.02	60.3 ± 11	4.36 ± 0.09	25.7 ± 0.9	2.10 ± 0.02	23.5	2.26
7.31–10.9 7.02–12.9	95.2 ± 11	4.12 ± 0.04	40.7 ± 0.4	1.864 ± 0.005	23.4	2.26
12.01–21.66 13.60–23.61	20.9 ± 0.8	4.73 ± 0.01	21.3 ± 0.3	2.115 ± 0.005	9.81	2.61

Table 1. Values of $J, k, \alpha, n, 3C$ and q for Ag as a function of the temperature range in which they are determined.

Values of W_v as a function of T were obtained from

$$W_{\rm v}(T) = W(T) - \rho_0 / L_0 T - (\rho_{\rm i} + \rho_{\rm d}) / L_0 T$$

where $W(T) = 1/K_c(T)$ and were plotted as a graph of $W_v T^2$ versus T as shown in figure 2. This graph also clearly indicates the existence of four separate regions in which $W_v = \alpha T^n$ with different values of α and n for each region as given in table 2. This behaviour

Metal	Temperature range (K)	Observed $(J/\alpha) \times 10^{11}$ (SI units)	Calculated from equation (5) $(J/\alpha) \times 10^{11}$ (SI units)	Observed $q = k - n$
Cu Ag Au			0.560 1.30 2.17	
Cu Ag Au	3.7–4.2 3.6–4.0	129 1.7×10^5		1.29 4.2
Cu Ag Au	4.5–7.0 5.7–7.0 4.5–5.6	124 23.5 ← →14.0		1.08 2.26 ← →2.62
Cu Ag Au	7.0–10.0 7.0–10.9 7.0–17.7	21.3 23.4 ↓ →13.9		1.98 2.26 ← → 2.64
Cu Ag Au	11–18 13.6–21.7	5.83 9.81		→2.55 2.61
Cu	20–30	L→ 6.05		→2.55

Table 2. A comparison of the values of J/α and q as obtained for Cu, Ag and Au and as calculated from equation (5).

of W_v cannot be attributed solely to the use of $\rho_i + \rho_d$ data in the calculation of W_v . Values of W'_v obtained from

$$W'_{\rm v} = W(T) - \rho_0 / L_0 T$$

provide a graph similar to that shown in figure 2 (with four separate regions), although the temperature variation of W'_v is of course somewhat different in each corresponding region. The same considerations also applied in the analysis of data for Cu and Au.

In those cases, however, the discontinuities in $\rho_i + \rho_d$ occurred at approximately the same temperatures as the discontinuities in W_v . In the present results the discontinuities in $\rho_i + \rho_d$ are displaced by as much as 0.8 K from those in W_v . We suspect that these differences are due to the relatively lower quality of the $\rho_i + \rho_d$ data. The corresponding data for Cu and Au were obtained several years later with an improved apparatus and consisted of a greater number of points in the region of interest.

Equation (3) may be written as the ratio

$$\rho_n / W_v = CT^q = (497.6/64.0)(L_0 / \Theta_D^2)T^3$$

With $\rho_i \simeq \frac{1}{3}\rho_n$ (to take into account umklapp processes (Klemens and Jackson 1964)) we have

$$\rho_{\rm i}/W_{\rm v} \simeq (\rho_{\rm i} + \rho_{\rm d})/W_{\rm v} \simeq 3CT^{q} = 3(497.6/64.0)(L_{0}/\Theta_{\rm D}^{2})T^{3}$$
(5)

In this form, equation (5) is independent of umklapp processes, of any dispersion of phonon velocity at high frequencies and of any possible variation in the electron-phonon coupling constant (if the same constant applies to both horizontal and vertical processes).

Metal	Temperature range (K)	$\alpha \times 10^8$ calculated by Suslov (m W ⁻¹ K ⁻¹)	$\alpha \times 10^8$ observed (s1 units)	n observed	n calculated by Borchi <i>et al</i>
Cu	4.5-7.0 7.0-10 11-18 20-30 Low 2-15 (Moriarity) 2-15 (Nand)	13	$\begin{array}{c} 4.99 \pm 0.24 \\ 18.1 \pm 1.1 \\ 32.3 \pm 1.0 \\ 7.07 \pm 0.25 \end{array}$	$\begin{array}{c} 2.67 \pm 0.03 \\ 2.00 \pm 0.03 \\ 1.74 \pm 0.01 \\ 2.26 \pm 0.01 \end{array}$	2.3–1.66 2.16–1.79
Ag	3.7–5.0 5.7–7.0 7.0–12.9 13.6–23.6 Low 2–15 (Moriarity) 2–15 (Nand)	27	3.07 ± 0.28 25.7 ± 0.9 40.7 ± 0.4 21.3 ± 0.3	$\begin{array}{l} 3.39 \pm 0.06 \\ 2.10 \pm 0.02 \\ 1.864 \pm 0.005 \\ 2.115 \pm 0.005 \end{array}$	2.05–1.67 2.0–1.69
Au	3.6–4.0 4.3–5.8 6.5–17.8 Low	21	$(8.7 \pm 33) \times 10^{-5}$ 14.8 ± 2.5 158 ± 2	$11.7 \pm 1.1 \\ 3.05 \pm 0.1 \\ 1.737 \pm 0.006$	

Table 3. A comparison of the values of α and n as obtained for Cu, Ag and Au and α as calculated by Suslov and n as calculated by Borchi *et al.*

Indeed table 1 shows that $3C = J/\alpha$ and q = k - n have the same value in the range 5.6–7 K and in the range 7–11 K despite the fact that J, k, α and n are different in these two ranges. However, the value of the q is 2.26 rather than 3 as predicted by equation (5) and the value of 3C is 23.5×10^{-11} (and 23.4×10^{-11}) in st units, rather than 1.30×10^{-11} as predicted by equation (5) (where we have used the low-temperature value $\Theta_D = 209$ for Ag). Surprisingly, the value of 3C increases to 129×10^{-11} st units and q decreases to 1.29 below 5 K, while 3C decreases to 9.81×10^{-11} st units and q increases to 2.61 in the region 13-22 K.

The values of 3C and q for Cu, Ag and Au are listed as a function of temperature range in table 2 (where typographical errors in the data for Cu (Kos 1985b) have been corrected). They indicate that for Cu and Au there also exist two regions in which J, α , k and n change but in which J/α and k - n do not. In all cases the observed values of J/α are very different from values calculated using equation (5). Moreover, equation (5) predicts that the values of J/α should be in proportion to Θ_{D}^{-2} as one passes from Cu to Ag to Au, and this proportion is not observed in any region. Finally equation (5) predicts that q = 3, whereas the observed values vary from 1.08 to 4.2.

In table 3 we compare our experimentally determined values of α and *n* for Cu, Ag and Au with theoretically calculated values.

4. Discussion

The theoretical model employed by Suslov provides values of α which correspond reasonably well to experimental values in the narrow temperature ranges 7.0–10 K, 4.7– 7.0 K and 4.3-5.8 K for Cu, Ag and Au respectively. For Cu and Ag these are also the ranges in which $n \approx 2$ (in which the Bloch hypothesis appears to be valid).

The calculations by Borchi et al appear to contain some essential elements which allow them to predict with good success the variation in n between 4.5 and 18 K for Cu and the variation in n between 5.7 and 12.9 K for Ag. In both cases they do not attempt to account for the variation in n at higher temperatures where we observe subsequent increases in n. Their calculation for Ag fails to predict the increase in n below 5 K which we observe. Moreover, their calculation predicts a smooth variation in n with temperature while we observe that n takes on four discrete values for Cu and Ag and three discrete values for Au.

Our observed discontinuities fall into two categories. In the first, J, α , k and n change from one region to the other but the ratio J/α and the difference k - n do not. This implies that the coupling constants in the electron-phonon interaction appear to have the same value (or to change in the same way) for elastic scattering as for inelastic scattering. In the second category, J/α and k - n do not remain constant as J, α, k and *n* change from one region to another. In the latter case it appears that it is necessary to consider different coupling constants for elastic and inelastic scattering, that one or both change from one region to another and that, if both change, they do not change in the same way. For example, a certain phonon mode may freeze out as the temperature is lowered. If this mode is far more effective in scattering electrons elastically than in scattering them inelastically, a discontinuity of the second kind might occur. If, on the other hand, the effectiveness of that phonon mode for elastic and inelastic scattering were in the same ratio as the effectiveness of all the remaining phonon modes for elastic and inelastic scattering, its removal might give rise to a discontinuity of the first kind. However, the fact that a given mode (or modes) freeze out, while others do not, implies that it is (they are) fundamentally different from the remaining modes and therefore, it is highly unlikely that its (their) ratio of effectiveness of elastic to inelastic scattering is identical with that of the remaining modes. The probability is further reduced when the same process must occur in three different metals.

It is perhaps better to take a general view of the observed discontinuities as being the result of phase changes in the electron-phonon system. In this case, two distinct types of phase change appear to occur and their relevant mechanisms are still to be determined.

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