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Low-Temperature Specific Heat of Ruthenium

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Measurements of the specific heat of Ru have been performed in the temperature range 2–11.5 K. Values for the Sommerfeld γ and the Debye Θ of 3.10 ± 0.02 mJ/mole K² and 555 ± 6 K were obtained. Deviations of Θ from its limiting value were observed above 9 K. This value of Θ is in good agreement with the acoustically determined value, thus removing the discrepancy between calorimetric determinations of Θ and the acoustic results.

There is considerable disagreement on the values of the parameters which characterize the low-temperature heat capacity of Ru, the Sommerfeld γ , and the Debye Θ , between those experiments which allow determination of these parameters.^{1–5} The existing situation is summarized in Table I and discussed briefly below. This study, the measurement of the heat capacity of Ru between 2 and 11.5 K, was prompted by the disagreement between the acoustically determined value of Θ ⁵ and that obtained in the analysis of the calorimetric data of Clusius and Piesbergen.² The Clusius and Piesbergen data seemed consistent with the acoustic data, provided that Θ increased slowly with decreasing temperature throughout the range 23–11 K rather than remaining constant below 23 K, as was assumed. If this is the case, a slightly larger value for γ than was obtained in the original analysis will be obtained. The results of this experiment were to find that this was the case. The limiting value of Θ was reached only below 9 K. The limiting value was in good agreement with the acoustic value. Little can be said of Wolcott's results,¹ which were obtained at

temperatures as low as 1.2 K, since no results other than values of γ and Θ and the value of the heat capacity at 20 K were given. The values of γ ob-

TABLE I. The parameters which characterize the low-temperature heat capacity of Ru according to various experiments.

Calorimetric determinations	γ (mJ/mole K ²)	Θ (K)
Wolcott (Ref. 1)	3.35	600
Clusius and Piesbergen (Ref. 2)	2.6	505
This work	$3.10 \pm .02$	555 ± 6
Acoustic determinations		
Fisher and Dever (Ref. 5)		554
Critical field analysis		
Hulm and Goodman (Ref. 3)	1.2	
Finemore and Mapother (Ref. 4)	2.4	

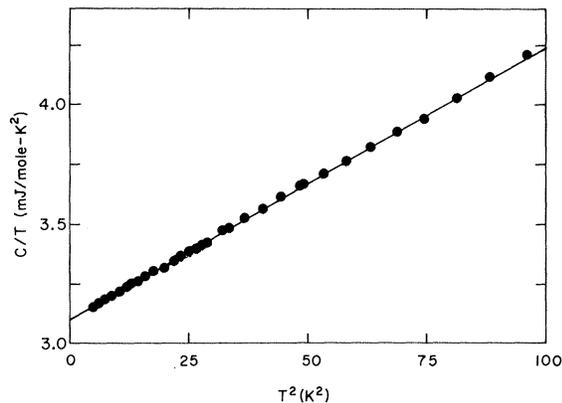


FIG. 1. The heat capacity of Ru below 10 K shown in the normal presentation of C/T versus T^2 . The straight line is a fit to the data below 9 K and yields the parameters given in Table I.

tained by analysis of the critical-field curve of Ru in the superconducting state^{3,4} can perhaps be ignored, because in one case the investigations were limited to the region near the transition temperature⁴ and in the other case the critical-field data were of poor quality.³

Ru represents an unfavorable case for an accurate determination of Θ , since Θ is relatively high. Thus the low-temperature specific heat is largely electronic in origin. For example, at 3 K the electronic contribution is 97% of the total. This feature, together with the suspected deviation of Θ from its limiting value at relatively low temperatures, makes the temperature range within which meaningful determinations of Θ can be made a rather restricted one: from about 3 to 9 K as proved to be the case.

The measurements were conducted using an adiabatic calorimeter described elsewhere.⁶ Measurements were conducted using temperature steps of approximately 5% of the initial temperature. The thermometer employed was a gold-doped germanium resistor which has been calibrated against a gas-bulb thermometer above 4 K and against the vapor pressure of helium⁷ below 4.2 K. The calibration above 4 K was checked by determining the heat capacity of a copper sample from 4 to 20 K. The result of this check showed that the copper measurements differed nowhere from the copper reference equation⁸ by more than 0.5%. Thus it is believed that the underlying temperature scale has an accuracy of 0.5%. The resistance-temperature calibration data were fit in the form of $\ln T$ as a power series in $\ln R$ using up to eight coefficients. Corrections were then applied to account for systematic deviations of the calibration data from the

formulas employed.

Measurements were conducted on the same sample on which the acoustic measurements were made, a zone-refined single crystal supplied by International Nickel Co., Ltd. The crystal, which had a total mass of 22.72 g, was divided into two pieces, of nearly equal mass, using the spark-cutting technique. The experiment was conducted in two series, first using the full Ru sample and second using only half the Ru sample. A total of 60 heat capacity determinations were made in the first series and 48 during the second. The addenda contribution to the measured heat capacity could be determined from these measurements and showed that above 4 K the addenda contributed about one-third of the total, although pains had been taken to minimize this contribution. Below 4 K the addenda contribution to the total heat capacity was a smaller fraction than above 4 K.

The results of this experiment are shown in Fig. 1. The data are presented in the usual manner by plotting C/T against T .² The line shown is a fit to the data below 9 K and yields the values of γ and Θ shown in Table I. The most notable feature of the data is the rather good agreement between the calorimetric and acoustic values of Θ . This removes the previously existing discrepancy. The current measurements of the heat capacity agree with the previous measurements of Clusius and Piesbergen to within the scatter of the latter measurements in the very restricted region of overlap. This comparison is best made in Fig. 2 where Θ , as deter-

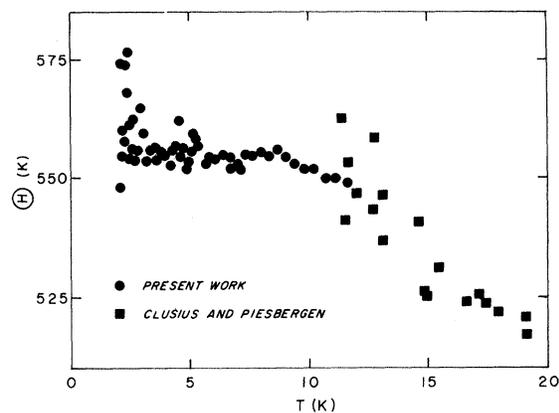


FIG. 2. The Debye temperature of Ru as determined calorimetrically assuming that $\gamma = 3.104$ mJ/mole K^2 as a function of temperature. The average value between 3 and 9 K is 555 K, which compares very favorably with the acoustically determined value, 554 K. The greater scatter observed below 3 K is a consequence of the small fraction of the total heat capacity contributed by the lattice in this temperature region.

mined calorimetrically assuming that $\gamma = 3.104 \text{ mJ/mole K}^2$, is shown as a function of T . This shows that Θ decreases from its low-temperature value for temperatures above 9 K. The tendency toward slightly higher Θ values below 3 K than above this temperature is felt to be spurious and to result from the extreme sensitivity of the calorimetric

value of Θ to small errors in the heat-capacity measurement when the lattice contribution is as small, as it is below 3 K. Although departures of Θ from the limiting value take place at slightly lower values of T/Θ than is typical of many metals, cases where deviations are found at even lower values of T/Θ are known to occur.⁹

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Electron Mean Free Path in Potassium*

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The magnetoacoustic effect was measured in potassium at temperatures of 1.2–8.5°K. A quantitative comparison with the free-electron theory indicates that the mean free path at higher magnetic fields is effectively 1.1 to 1.25 times larger than at low fields. It is suggested that this deviation is due to the failure of the relaxation-time approximation to give an adequate description of the electron scattering. The temperature dependence of the mean free path was found to be in agreement with resistivity measurements.

INTRODUCTION

The purpose of this paper is to report measurements of the magnetoacoustic effect in potassium at temperatures ranging from 1.2 to 8.5°K (above which the effect disappears). A quantitative comparison of the data with the free-electron theory of Cohen, Harrison, and Harrison¹ (CHH) is made. This comparison is used to determine the electronic mean free path (MFP) and its temperature dependence. A consistent deviation of the theoretical attenuation from the data is observed at higher magnetic fields at all temperatures. A qualitative argument indicates that this deviation could be due to the incorrectness of the relaxation-time assumption in the CHH theory.

EXPERIMENTAL PROCEDURE

The potassium samples were cylindrical disks $\frac{7}{16}$ in. in diameter and approximately $\frac{1}{4}$ in. long. These were spark cut from single-crystal boules grown from zone-refined stock by a method described by Foster *et al.*² The residual resistance

ratios of the samples varied from 6000 to 8500. A single coaxially plated x-cut quartz transducer was bonded to each sample with Dow Corning 7 compound. Longitudinal waves were propagated along or near the [110]-growth direction. The samples were mounted in such a way that the propagation vector \vec{q} was always perpendicular to the magnetic field \vec{H} , while the latter could be rotated through 360°. A pulse generator-receiver system was operated at 45 MHz to produce and detect the ultrasonic signals. A Matec model 1235 attenuation recorder was used so that the relative attenuation could be plotted directly as a function of magnetic field.

To obtain temperatures above the boiling point of liquid helium, heat was supplied by a resistance coil wrapped around the copper sample holder. The sample holder was insulated from the helium bath by a double-walled stainless-steel jacket, leaving a small residual coupling via the stainless-steel support tube. This coupling was necessary to maintain thermal equilibrium for a given heat input. Temperatures were measured with a germanium