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ELECTRICAL RESISTIVITY OF PLUTONIUM METAL BETWEEN 1.73°K AND 298°K

by

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ABSTRACT

A brief description of apparatus and methods used in measuring the electrical resistivity of plutonium metal down to 1.73°K is given. Figures showing the electrical resistivity variations between 1.73°K and 55°K, from 50°K to 295°K, and from 130°K to 298°K are included. Interpretation of the data reported offers confirmation for the major peaks observed in low temperature heat capacity data of plutonium.

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1. INTRODUCTION

The electrical resistivity of plutonium metal has been previously reported for the temperature region 25.8°K to 787°K by Sandenaw and Gibney.[1]

In the low temperature range, the specimen was cooled by helium exchange gas, with the outer brass vacuum jacket (or container) being in direct contact with either liquid nitrogen or liquid hydrogen. Cooling to the lowest constant temperature took only a few hours, and then warming of the specimen commenced. The normal procedure was to warm the specimen from ~26°K to 298°K in a matter of 6 to 10 hours. The reproducibility of readings was 0.17 per cent or better on warming back to 273.15°K.[1] From these results there was little reason to believe that any hysteresis existed in the physical properties of plutonium at low temperature or that any anomalous behavior could be expected.

When measurements of heat capacity[2] were started in the low temperature region, an irreproducible number of heat capacity spikes were noticed between 15°K and 170°K depending upon specimen processing. Resistivity measurements were again made over the attainable temperature region below room temperature in order to get confirmatory evidence, if any, for the specific heat spikes.

2. EXPERIMENTAL DETAILS

2.1 Apparatus

As has been done in previous instances, the current contacts and potential drop probes were spring loaded against the plutonium for ease of assembly and disassembly. The bridge circuit used for measuring resistivity was described in Reference 1.

The temperature was measured between 3.96°K and 298°K with thermocouples made from 2.11 atomic per cent cobalt in gold vs copper. The calibration of thermocouples has been described elsewhere.[3] The vapor pressure of liquid helium was used in measuring the temperature below 3.96°K.

2.2 Procedure for Electrical Resistivity Measurements

The specimen, in its holder, was placed in a double dewar cryostat. After the inner dewar, which contained the specimen holder, was leak tested, helium was admitted to a pressure of 15 mm. Liquid nitrogen was added to the outer dewar and the specimen was sometimes allowed to cool slowly to approximately 85°K by long standing. At other times nitrogen was transferred directly on the specimen. The liquid nitrogen was boiled away in the latter case before liquid helium was added. Resistivity measurements were made while the helium was pumped to the lowest temperature attainable. Resistivity readings were again made while the specimen warmed back to a temperature very close to 3.96°K (boiling point at Los Alamos normal pressure). Measurements above 3.96°K were made by boiling away the remaining liquid helium and letting the self heating of the specimen and heat leaks along wires warm the specimen and holder slowly. Added heat was sometimes supplied above 20°K.

Liquid nitrogen can be pumped to a temperature of ~48°K. Certain limits of the resistivity curve were determined by varying the cooling cycle, the holding time at ~48°K, and the warming rate up to room temperature.

2.3 Specimen Description

The three specimens examined in the electrical resistivity apparatus were made from normal-purity material; specimen No. 1 was of the best quality available at the time. Specimens 1 and 3 were cylindrical rods of length 2-1/4 in. and diameter 11/64 in. Specimen No. 2 was also a cylindrical rod, but shorter; its dimensions were: length, 1-5/8 in. and diameter, 7/32 in.

The major impurities in specimens 1 and 2, as reported from spectrographic and chemical analyses, are given in Table I.

TABLE I.

Impurity Elements in Plutonium Specimens (ppm)

Specimen	Mg	Ca	Al	Si	Ni	Fe	Cr	C	O_2
No. 1	70	< 5	50	80	100	50	30	80	25
No. 2	10	15	20	85	50	350	< 20	50	130

3. EXPERIMENTAL RESULTS

3.1 Liquid Helium Runs

The results of electrical resistivity measurements made between 1.73°K and 55°K are shown in Figure 1. Curves No. 1 and No. 2 correspond to the specimens designated by these numbers.

Curve No. 1 is an average of three determinations. Reproducibility was extremely good with No. 1. Curves obtained were apparently independent of cooling rate from 298°K to 3.96°K and also independent of holding time in the liquid helium. This specimen was processed for 1 day before cooling to 3.96°K; it was cooled directly from 76°K to 3.96°K as rapidly as possible; it was then cycled for 2 days between ~76°K and ~48°K before cooling again to 3.96°K.

Specimen No. 1 was held at 3.96°K or below for 1 hour before the first set of measurements, 10 minutes before the second run, and 3 minutes before the third run. On warming after these three described processing and holding cycles, the maximum deviation of any one point was ± 1 per cent on the scale as shown, and very few points had this deviation. This specimen was high density, but was not quite what could be classed as high purity. Note the slight offset at 31.5°K.

Two curves were obtained with specimen No. 2. The point of offset was at ~27°K. One curve merged with that of specimen No. 1 at ~38°K and followed the same path upward as No. 1. The other curve remained consistently lower than any other curve previously obtained, having a peak

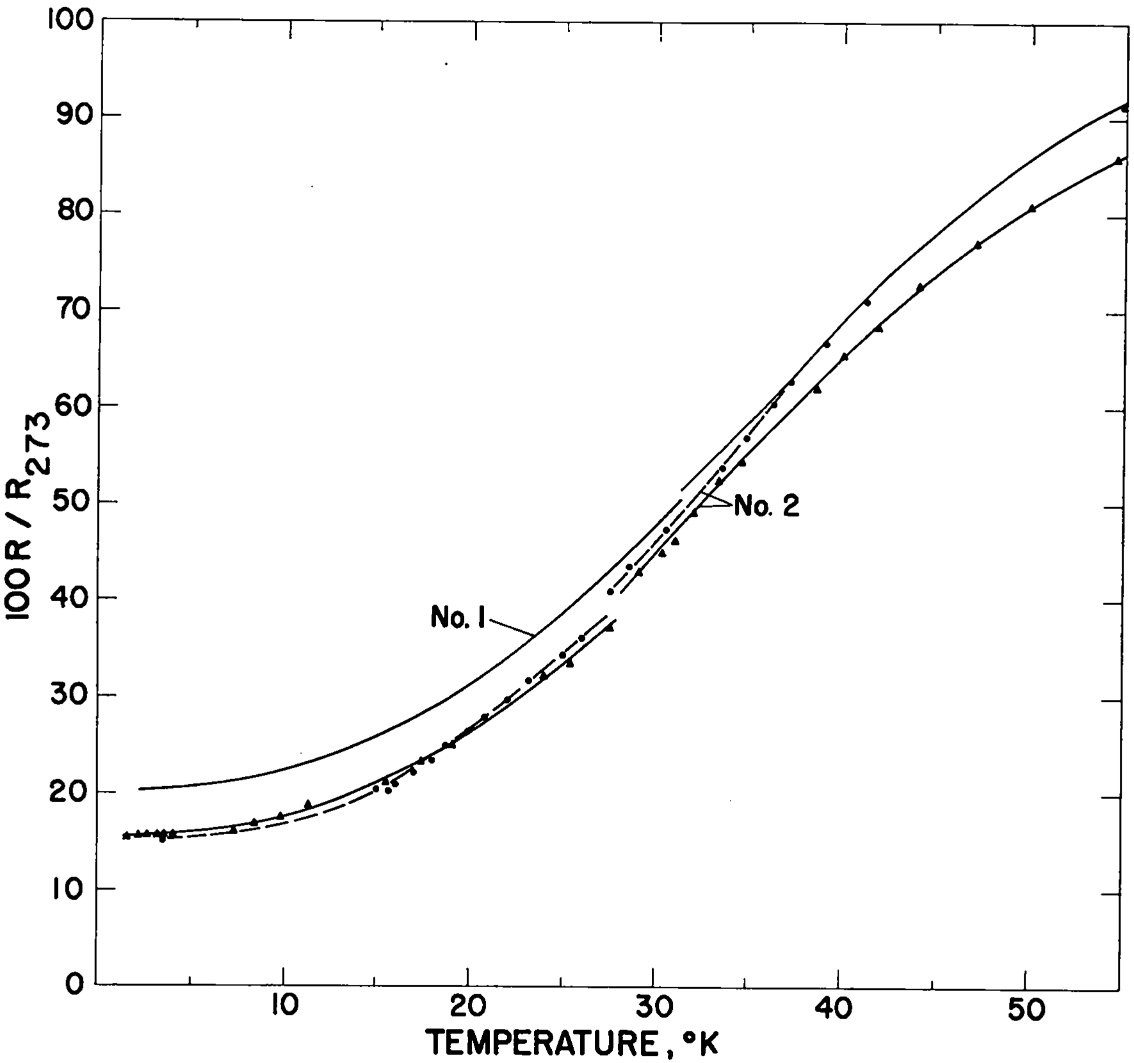

Fig. 1. Electrical resistivity curves for plutonium metal as a function of temperature between 1.73°K and 55°K. (No. 1 represents an average of three runs with specimen No. 1; No. 2 shows branching observed with specimen No. 2.)

resistivity of 1.05 R_{273} at 110°K. On slow warming, the resistivity returned to its original value at room temperature.

The higher of the two curves for specimen No. 2 resulted after rapid cooling from room temperature and holding in liquid helium for slightly over 3 hours. The lower of the two curves found for this specimen resulted after 2-1/2 days of processing in the temperature range of 101°K to 104°K, followed by 2-1/2 hours in liquid helium. The times taken in covering the temperature range of 3.96°K to 55°K (Fig. 1) were also different in the two sets of measurements taken with specimen No. 2. The time taken in determination of the upper curve was 3-1/4 hours, while time taken in determination of the lower curve was 2-1/2 hours.

The offset in resistivity at ~27°K was more pronounced in specimen No. 2 than with specimen No. 1 at 31.5°K. (The quality of material making up the two specimens was different as shown in Table I.)

3.2 Pumped Liquid Nitrogen Runs

Limits of the resistivity curve were determined after varying both the length of the holding time at ~48°K and the warming rate up to room temperature. These limits and the normal curve obtained on cooling from 298°K to ~48°K in a few hours and warming back again to 298°K in a few more hours are shown in Fig. 2. These curves were obtained with specimen No. 3 from runs not involving liquid helium. The upper limit near room temperature was obtained by repeated cycling between 63°K and ~48°K and then rapid warming to room temperature. The return to the normal

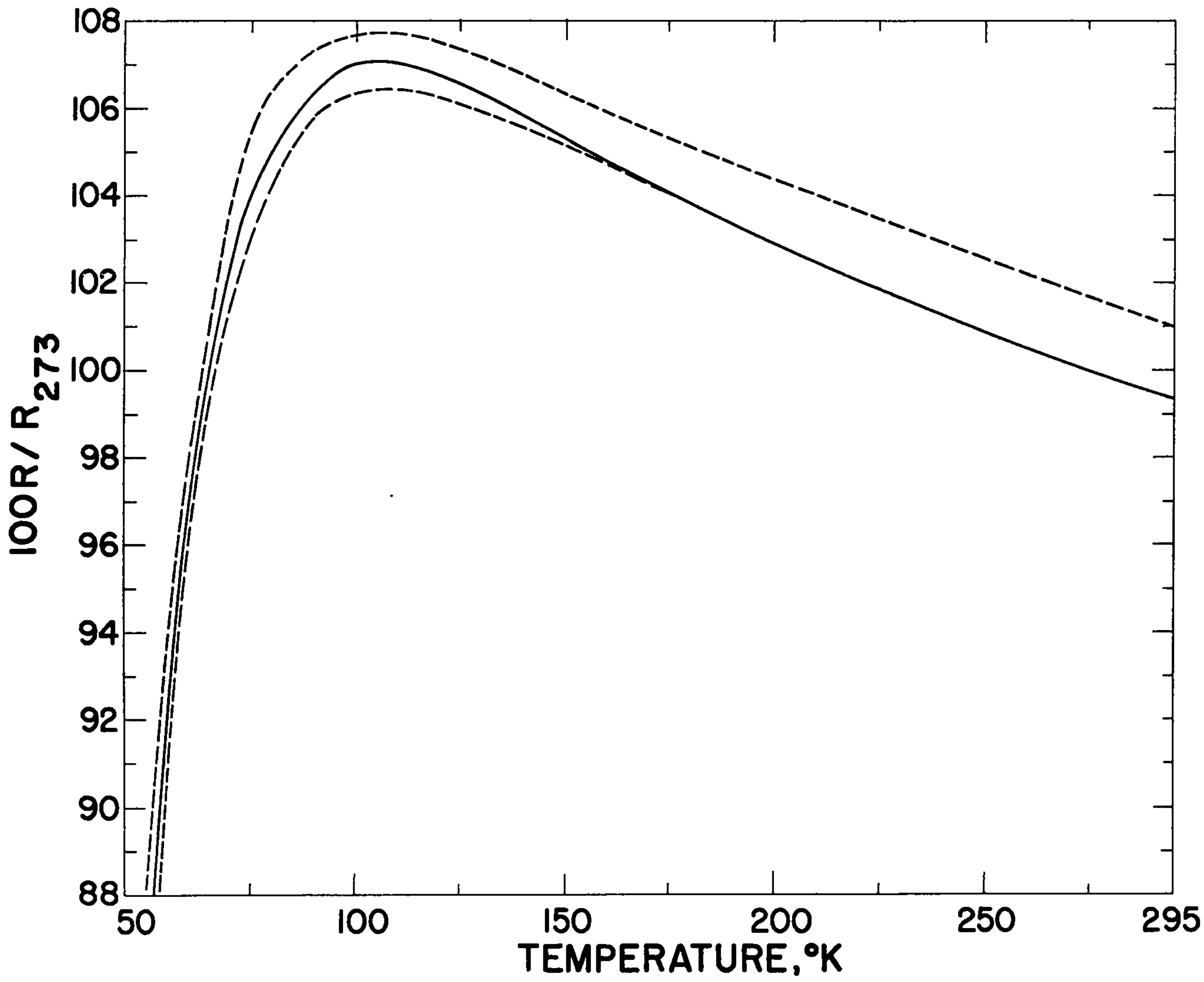

Fig. 2. Electrical resistivity of plutonium metal between 50°K and 295°K. (Dashed line represents observed limits; solid line represents normal curve.)

resistivity (found repeatedly at room temperature) in this case required several days. This indicated that some low temperature properties had been retained which were very sluggish in their transformation.

3.3 Results from Slow Cycling between 298°K and 130°K

The branched curve obtained on cycling specimen No. 1 between 298°K and 130°K is shown in Fig. 3. The points as shown on this figure represent two complete cycles over the given temperature range. Cooling from 298°K to 130°K took 13 hours in one case and 14 hours in the other. The warming times were over 2 days in each case. Warming was accomplished by letting the liquid nitrogen in the outer dewar boil away normally.

Note that branching in the R vs T curve seems to occur at ~288°K on cooling and that the branching ends at ~150°K. There is a definite change in slope of the cooling curve at ~225°K. The slope of the warming curve between 185°K and 238°K seems to be the same as between 250°K and 270°K. An offset is apparent between 240°K and 250°K, and a peculiar dip and rise is also noticeable in the warming curve between 270°K and 288°K.

The data of Fig. 3 were plotted as a function of R/R_{298} because of the evident branching noted at about 288°K.

4. DISCUSSION OF RESULTS

4.1 Correlation of Resistivity Results with Heat Capacity Data

The electrical resistivity data appear to offer confirmatory evidence for all of the major heat capacity peaks observed by Sandenaw,

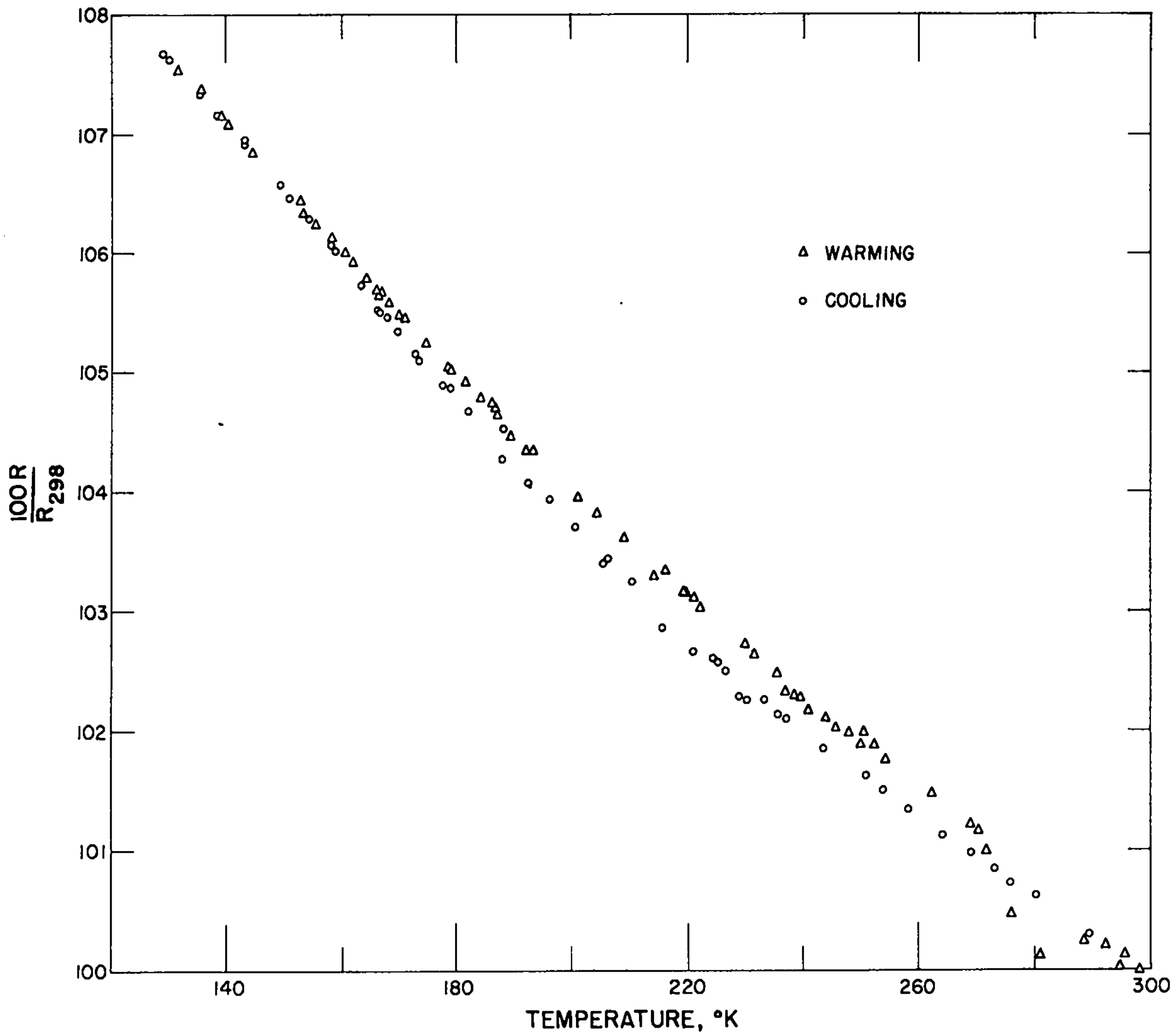

Fig. 3. Electrical resistivity of plutonium metal between 130°K and 298°K. Circles represent slow cooling data and triangles indicate data taken on slow warming.

Olsen, and Gibney[2] in the C_p vs T curve of plutonium metal. The offset in the resistivity curve, which occurred at 31.5°K with one specimen and 27°K with the other, is very close to the temperature observed for the first heat capacity peak, i.e., 31°K.

The resistivity behavior, noted between the lowest temperature attainable with pumped nitrogen (~48°K) and 295°K (shown in Fig. 2), can be explained as being due to the factor causing the second heat capacity peak observed at 47°K. The minimum temperature reached by pumping liquid nitrogen and the length of time the specimen could be held (or was held) at the minimum temperature could determine whether or not any conversion to another state took place and the amount of conversion.

It has been suggested[1] that the peak in electrical resistivity at 105°K, as shown in Fig. 2, is due to spin disorder, and it has been assumed that the ordering of spin is antiferromagnetic. The heat capacity peak at 123°K could then be tied in with this electrical resistivity behavior at ~105°K, and one set of measurements could be confirmation of the other set. The lower temperature limit of branching in the resistivity curve as shown in Fig. 3 is at ~152°K. This is very close to the temperature of the minimum in heat capacity which follows the 123°K peak.[2]

The heat capacity curve of high-purity plutonium also showed a dip at 240°K accompanied by resumption of a slow continuous rise in heat capacity beginning at 250°K. Apparent verification of this heat capacity behavior is shown by the offset in the warming curve of resistivity in this same temperature span.

The peculiar dip and rise in the R vs T curve of Fig. 3 between 270°K and 288°K has a possible explanation in annealing out of radiation damage caused by α-particle bombardment. Such a dip and rise has been seen in the resistivity curve of gold subjected to deuteron bombardment, as reported by Seitz and Koehler.[4] A small fluctuation in the heat capacity curve of plutonium[2] was seen between 270°K and 275°K, but it was disregarded in drawing the curve.

A different observable effect occurred in electrical resistivity in the temperature range of each of the heat capacity peaks noted. A different mechanism appears to be the cause of each peak.

4.2 Effect of Impurities on Physical Properties

Attention is called to the flatness of the resistivity curve between 1.73°K and 9.0°K and the high residual resistivity. Also one should notice that the lowest residual resistivity was found with the specimen containing the greater quantity of ferromagnetic elements as impurities. The interaction of conduction electrons with the localized spin of the ferromagnetic impurities seemingly lowers the residual resistivity.

If spin disorder was the cause of the major specific heat peak in plutonium at 123°K and of the high electrical resistivity which peaks between 100°K and 110°K, then impurity elements which restrained ordering of spin should be evident by a lowering of peak resistivity and absence (or near absence) of the heat capacity peak. Of the impurity elements present in plutonium, only nickel, iron and cobalt (ferromagnetic elements) logically appear to be ones which could interfere with the antiparallel ordering of spins postulated for antiferromagnetism.

The change in the slope of the resistivity curve found on cooling at ~225°K (Fig. 3) occurs at the same temperature as the commencement of a heat capacity peak in normal-purity plutonium having an iron and nickel content of 360 ppm. The change in slope of the linear expansion curve of plutonium having an iron content of 600 ppm[5] is at ~193°K. It is thus suggested that these different physical measurements discussed for the temperature range 193°K to 250°K represent the same phenomenon as influenced by ferromagnetic impurities. It should be pointed out that no matter how high-purity or low-purity specimens were processed in the temperature range of 1.73°K to 298°K, the original room temperature density was always found when checked at 298°K, after completion of the individual experiment.

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