Transverse Magnetoresistivity of α and β Plutonium*

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Transverse magnetoresistivity measurements have been made on pure, polycrystalline α and β plutonium. The magnetoresistivity of both phases is negative at 5°K and low applied fields, and becomes positive at fields greater than 2000 G. The negative effect decreases with increasing temperature and is not dependent on sample texture. The higher-field results follow the H^2 dependence found in "normal" metals. It is concluded that the negative effect is due to magnetic ordering, which results from s-f exchange, and the Néel temperatures are near 20–30°K for both phases. The high-field data are consistent with earlier models in which the electrical conductivity is dominated by a small number of holes in the $7s$ band.

I. INTRODUCTION

EXISTENCE of maxima in the resistivity-versus-temperature plots for α,[1] β,[2,3] and δ[4] plutonium (see Fig. 1) led to the proposal of low-temperature antiferromagnetism in these materials.[1,5–7] Direct confirmation of this hypothesis has not been obtained from magnetic-susceptibility,[4,8–10] neutron-diffraction,[11] or specific-heat[12] measurements. Those property measurements which have indicated a magnetic transition are radiation damage,[13–16] Hall effect,[3,17] thermal expansion,[18,19] and thermoelectric power.[6,19] The latter set of properties give indirect indications of magnetism by means of similar behavior for known magnetic metals, e.g., α manganese and the rare earths. The general conclusion has been that the possibility of magnetic transitions does exist, but the magnitude of the magnetic moment must be very small.[20]

The present study was made to see if the magnetoresistivity of pure, polycrystalline α and β plutonium would indicate the presence of magnetic order. The results were found to give the strongest evidence, to date, of magnetic transitions in these allotropes. Some general conclusions about the band structure of plutonium have also been drawn from the data.

II. EXPERIMENTAL

The equipment and techniques used here were described previously.[3] Most of the potential across the resistivity leads was bucked-out with a Honeywell 6-dial potentiometer, and the remaining signal was amplified with a Keithley model-149 millimicrovoltmeter and displayed on a Leeds and Northrup Speedomax W recorder. The amplifier was adjusted to give the greatest sensitivity consistent with the signal noise generated by vibrations of the glovebox system. Most measurements were made with the recorder full-scale equivalent to 1 or 3 μV. Data were taken in increasing and decreasing magnetic fields, with the sample current and field in each of their two possible directions. The potentials were found to reverse with current but not with field, and were therefore due to the usual magnetoresistivity.

The plutonium had been purified by fused-salt electrorefining.[21] Total impurity content, exclusive of americium, was 140 parts per million (ppm) by weight. Samples containing 400 and 600 ppm of americium were used, and no observable differences were found. Sheet samples of α plutonium (monoclinic structure) were prepared by cross-rolling or by reverse-rolling. The latter procedure is known to introduce a large amount of preferred orientation in the samples,[22] and the resistivities were measured parallel and perpendicular to

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the rolling direction. β plutonium (body-centered monoclinic) was retained at low temperatures by quenching from the equilibrium temperature range of 390–480°K into liquid nitrogen. The resistivity-temperature behavior of the quenched sample established that the β phase had been retained.[2,3]

The major sources of uncertainty in the measurements were the large temperature dependence of resistivity and the increase of resistivity with time due to the self-radiation damage. Effects of the former were minimized by making all measurements with the samples immersed in a suitable cryogen. The liquids used were helium, hydrogen, neon, and nitrogen. Even with this approach, some drift was observed in the data beyond that due to the self-damage. This is not surprising since the slopes of the resistivity-temperature curves between 10 and 20°K are as high as 3.5 and 8.0 $\mu\Omega$ cm/°K for α and β plutonium, respectively. These correspond to changes of 40–80 μV/°K in this study.

At 5°K, the self-damage of α plutonium caused an increase of about 0.01 μV/min, which may be compared with a 1 μV change due to an applied field of 19 kG. Since 30 min were required for a full field cycle, the data were corrected to a reference time. A hysteresis was observed in many of the field cycles, but it was probably not from a magnetic hysteresis, because the sign of this effect seemed to be random. It is likely that the hysteresis is due to an accumulation of systematic errors from the corrections for self-damage at the 20–30 fields used during each cycle.

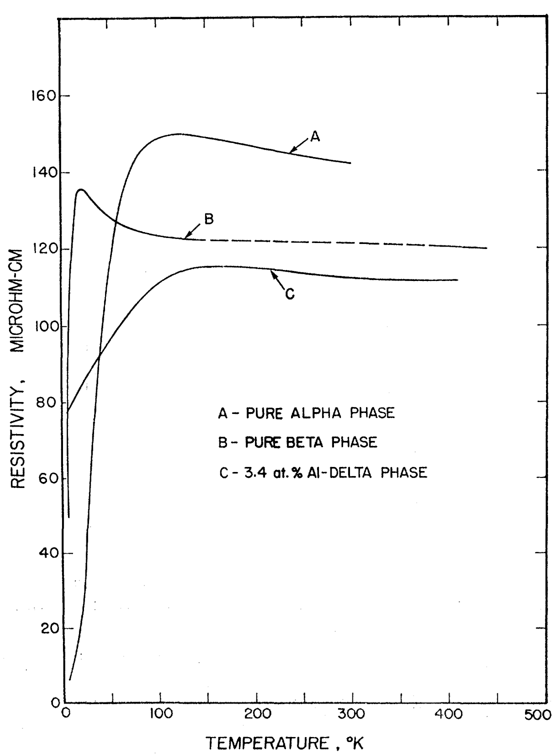

FIG. 1. Electrical resistivity of α, β, and δ plutonium (Ref. 3).

III. RESULTS

A. α Plutonium

Representative data are given in Figs. 2–4. It is seen that the effect of small applied fields is to reduce the resistivity. After the field has been increased above several thousand gauss, the magnetoresistivity ($\Delta\rho/\rho_0 = \Delta E/E_0$) becomes positive. The data are interpreted to be due to a negative magnetoresistivity at low fields, caused by magnetic effects in the sample (see Sec. IV), which is over-ridden at higher fields by the usual positive magnetoresistivity. The negative effect at 5°K is larger than the errors discussed above, and is shown in Fig. 2 to be definitely present despite the hysteresis with field. Figure 2 also shows that little difference is found with the field applied in either of the two directions. There is little effect of preferred orientation on the results, and it may be concluded that the negative magnetoresistivity is not the result of a special field-current-lattice direction combination. The results for cross-rolled samples were intermediate to those of the reverse-rolled metal. The negative effect was found to decrease with temperature, and the highest temperature at which it was observed was 27°K. The next highest temperature studied was 53°K, and if the negative effect exists at 53°K, it could be no larger than one part in 10^5.

The solid curves in Figs. 3 and 4 have been drawn through the higher-field data with an assumed positive dependence of magnetoresistivity on the square of applied field. The higher-field data are seen to follow this usual dependence. Plots of $\Delta E/E_0$ versus H/E_0 at various temperatures for data above 3 kG are shown in Fig. 5. "Normal" magnetoresistivity data for a metal are expected to fall along a single curve in such a Kohler plot.[23] This seems to be the case in Fig. 5. A plot of the "reduced Kohler diagram" show the data for α plutonium to lie near those of antimony, and the slope is close to the expected value of 2.0.

B. β Plutonium

Figure 6 shows the data for the metastable β phase at 5°K, where the solid curve shows the expected behavior with H^2. These data are very similar to those for α plutonium, i.e., a negative magnetoresistivity at low fields, and a positive magnetoresistivity at higher fields, which increases as H^2. The greater variation of resistivity with temperature for β plutonium made measurements between 5 and 22°K (the temperature corresponding to the maximum in the resistivity-temperature curve) very difficult. The magnetoresistivity at 21°K seems to be negative, with $\Delta\rho/\rho_0 = -4\times 10^{-5}$. Data taken at 52 and 78°K showed no negative effects.

A Kohler plot of the positive magnetoresistivity data at 5°K was similar to those for α plutonium, but it was

[23] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, London, 1960), pp. 490ff.

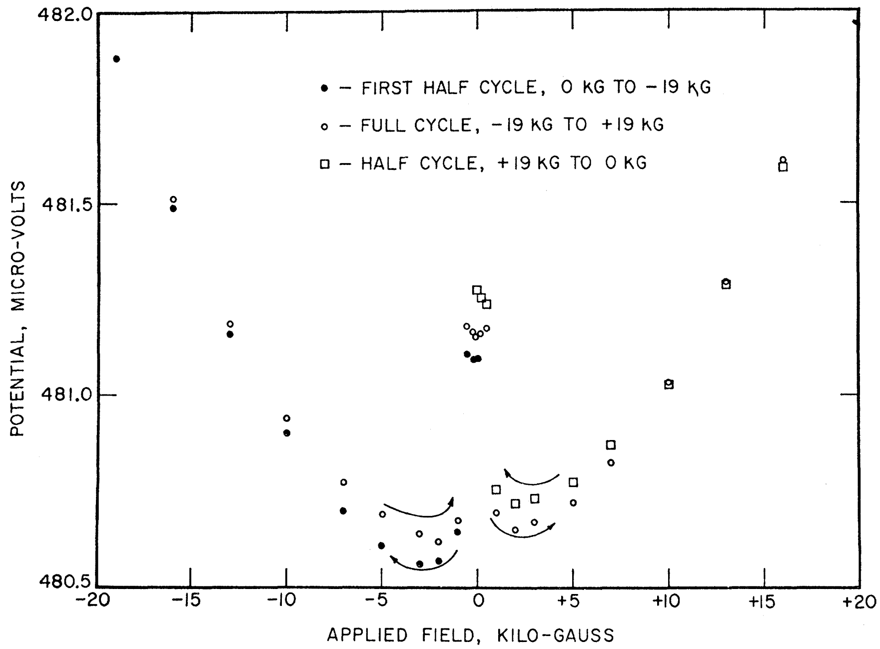

FIG. 2. Resistive potential of α plutonium versus magnetic field at 5.0°K (current perpendicular to the rolling direction).

not possible to obtain curves at different temperatures. The reduced Kohler plot for β plutonium is about the same as for arsenic.[23] This normal behavior of the higher-field data for α and β plutonium supports the separation of the results into abnormal, negative, and normal, positive contributions.

IV. DISCUSSION

Magnetic Transition

The negative magnetoresistivities of α and β plutonium are the strongest evidence for a magnetic transition found thus far for these materials. Negative magnetoresistivities have been found only in magnetic metals[24–26] or ultrapure samples having electronic mean free paths comparable to sample dimensions.[27] The high residual resistivities of the samples used here, none less than 16 $\mu\Omega$ cm, rules out the latter possibility. The negative effects have been explained by *s-d* or *s-f*

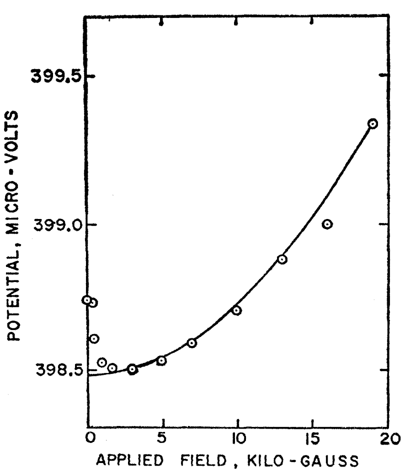

FIG. 3. Resistive potential of α plutonium versus magnetic field at 5.0°K (current parallel to the rolling direction).

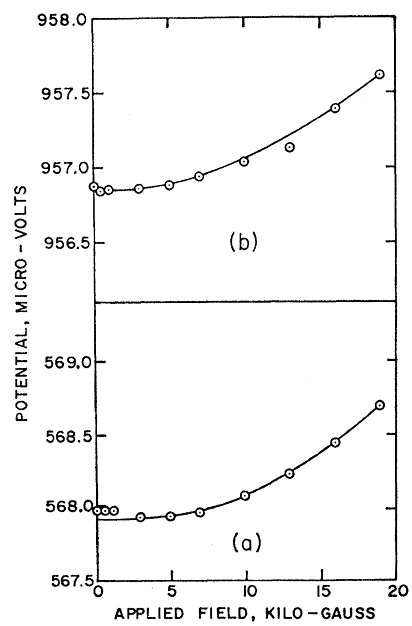

FIG. 4. Resistive potential of α plutonium versus magnetic field (current perpendicular to the rolling direction): (a) 12.7°K; (b) 20.9°K.

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[25] M. P. Sarachik, Phys. Rev. **137**, A659 (1965).

[26] A. Isin and R. V. Coleman, Phys. Rev. **142**, 372 (1966).

[27] F. J. Blatt, A. Burmester, and B. LaRoy, Phys. Rev. **155**, 611 (1967).

exchange interactions in dilute magnetic alloys and in rare-earth metals.[28,29] Rocher considered the possibility of 5f magnetic electrons in virtual bond states in plutonium, which are in resonance with the conduction (s-d) electrons.[7,30] He was able to account for the large magnitude of the resistivity above the magnetic transition with only a small difference in the occupation of the spin-up and spin-down virtual states. The small difference between $n\uparrow$ and $n\downarrow$ explains the lack of confirming evidence in the magnetic susceptibility. Although Rocher's method of determining the spin-disorder contribution to the resistivity is slightly arbitrary, his general conclusions are probably valid.

The present work does not establish the exact Néel temperatures for the two phases. In view of the rapid drop of the negative part of the magnetoresistivity with temperature, it is likely that the Néel temperature cannot lie far above 27°K for α and 21°K for β plutonium. This conclusion is in agreement with the earlier choice of Néel temperatures based on the temperatures of a maximum or minimum in the Hall effect.[3] Thus, as before, it is concluded that the resistivity maximum for α plutonium at 100°K is not due to the antiferromagnetic transition and that Smoluchowski's treatment of phonon-assisted, interband scattering must be considered.[31] The Néel temperature for β plutonium occurs above the temperature at which the interband scattering starts to saturate, and thus the magnetic transition is seen as a maximum in the resistivity-temperature curve. A break is not seen in the resistivity-temperature curve for α plutonium at its Néel temperature because the rate of change due to the interband scatter is too great at that temperature.

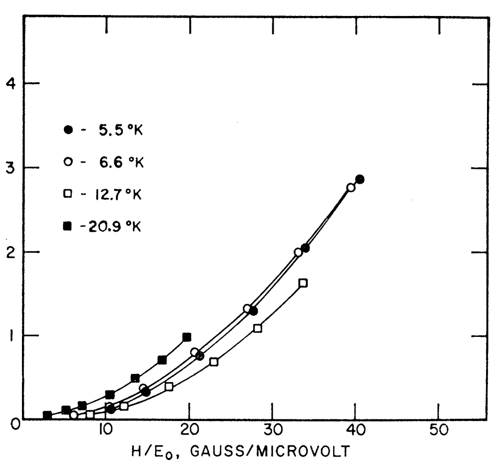

FIG. 5. Kohler plot of "positive" magnetoresistivity for α plutonium.

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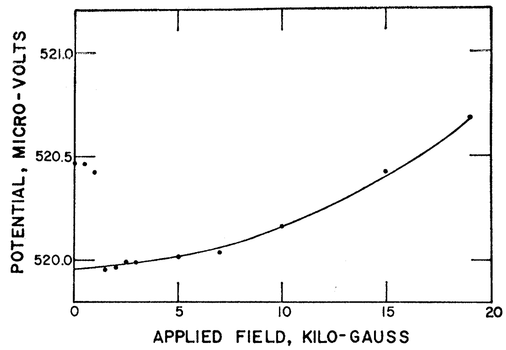

FIG. 6. Resistive potential of β plutonium at 5°K.

No attempt has been made to calculate exact, two-band parameters from the combined Hall and magnetoresistivity results because of the limited applicability of such an approach even for simple metals.[23] Qualitatively, however, the fractional numbers of electron holes calculated for the $7s$ band from Hall data[3] are consistent with the magnetoresistivity results, which group α and β plutonium with other high-resistivity metals, such as arsenic and antimony. Recent tight-binding and augmented-plane-wave calculations for δ plutonium (fcc) show that $5f^67s^2$ or $5f^{6-y}6d^y7s^2$ configurations can account for the observed magnetic susceptibility and electronic specific-heat term.[32] These models are similar to the $5f^{6+x}7s^{2-x}$ or $(5f-6d)^{6+x}7s^{2-x}$ configurations deduced from the Hall data. Measurements of posititron annihilation in uranium[33] yield a $(5f-6d)^{4.1}7s^{1.9}$ band structure, which agrees with the structure for uranium obtained from the Hall-coefficient data. Despite the assumptions used in the two-band model of conductivity, the model appears to be useful qualitatively for the actinide metals.

It is concluded that although the detailed band structures of α and β plutonium are not known, a unified picture is evolving, with a high density of states at the Fermi surface; a nearly full $7s$ band; a very narrow split $5f$ band due to spin-orbit coupling; gross similarities in electron-transport properties in various allotropes, and therefore, little change in properties from Brillouin-zone boundary effects; and general similarities of band structure among the actinide elements.

ACKNOWLEDGMENT

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