There appears to be no simple relation between j and J, but it is interesting that j must be positive because of the square in (3). It is clear that in the limit of large transfer energies  $(q_1^{BB} \rightarrow \infty)$  the  $S^6$  contribution to j will vanish, an expression of our earlier result that the ionic term contributes first to j terms of order  $S^8$ .

Anderson estimated that  $j \sim b^4/U^3$  where b was a transfer integral and U a transfer energy. Our result is in agreement with his if we make the reduction associated with the neglect of terms in the Hamiltonian containing S explicitly. We find  $j = 8b^2(J' + J'')^2/(\Delta E)^3$  where J' and J'' are the Anderson and Anderson-Hasegawa exchange integrals, respectively,  $\Delta E$  is the energy difference between excited and ground configurations, and b, with the neglect of some Coulombic terms, is Anderson's transfer integral.

We have used Keffer and Oguchi's values appropriate to MnO for the various overlap integrals appearing in (3) and have estimated the transfer energy  $q_1^{BB}$  in a manner similar to Kondo.<sup>10</sup> We find  $q_1^{BB} \sim 14$  eV and, using this value, find the results shown in the following table:

$$J(^{\circ}K) = j(^{\circ}K) = j/J$$

Calc. (MnO)	43.5	0.70	0.016
Exp. (MnO,	11		0.01 - 0.02
reference 2)			
Calc. (Mn <sup>++</sup> in MgO)	60.9	1.37	0.022
Exp. (Mn <sup>++</sup> in MgO,	14.6	0.73	$0.05 \pm 0.03$
reference 1)			

The theoretical results for MnO are in reasonable agreement with the results of Rodbell et al.<sup>2</sup> considering the approximate nature of the overlap integral computation. In particular, we expect the ratio of j/J to be rather more accurate than either our result for j or J alone. Indeed, it is seen that our value of j/J lies within the experimental range of error. Our MnO results are not in very close agreement with the pair spectrum

values, but this is to be expected because of the difference in lattice constant between MnO and MgO. We have applied Harris and Owen's empirical factor of 1.4 for J and  $(1.4)^2$  for j in arriving at the results presented in the third line of the above table.

We are in the process of computing the overlap integrals more carefully for  $Mn^{++}$  ions in both MnO and MgO using Hartree-Fock wave functions. We hope this will improve the accuracy of our results and will warrant similar calculations for other salts. In any case, we have certainly shown here that the usual superexchange mechanism is fully capable of explaining the origin and magnitude of the biquadratic exchange interaction.

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## EFFECT OF PREFERRED ORIENTATION ON THE ELECTRICAL RESISTIVITY OF ALPHA PLUTONIUM AT LOW TEMPERATURES\*

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The electrical resistivity of monoclinic alphaphase plutonium is known to rise rapidly between  $4^{\circ}$ K and  $105^{\circ}$ K, and then to decrease slowly to  $388^{\circ}$ K (at the  $\alpha - \beta$  transition). We have now found this behavior to be strongly affected by preferred orientation of the polycrystalline grain structure.

Oriented specimens were prepared by heating plutonium to a temperature high in the beta-phase region, where a compressive load of 60 000 psi



Fig. 1. Resistivity data vs temperature for highly oriented and randomly oriented alpha plutonium.

was applied and held until the metal had cooled and transformed to alpha phase. The resulting microstructure consisted of columnar, rodshaped grains, whose axes were parallel to the pressing direction. X-ray diffractometer traces of prepared surfaces revealed that the (020) planes were aligned perpendicular to the pressing direction. It was also determined that no secondary alignment was present, i.e., orientation was random in all directions orthogonal to the pressing direction. Details of this procedure used in preparing oriented specimens will be published elsewhere.<sup>1</sup>

The resistivity specimens were rods 0.6 cm in diameter and 2.54 cm long, machined so that the (020) planes were preferentially aligned either per-

Orientation of specimen	Residual Resistivity (μΩ-cm)	Temp at <sup>P</sup> max (°K)	$\rho_{max}$ ( $\mu\Omega$ -cm)	$\rho_{\rm max}^{-\rho} 300^{\circ} {\rm K}$ ( $\mu\Omega$ -cm)
(020) planes ⊥ to direction of current	6.2	125-170 <sup>a</sup>	137	1
Randomly oriented	• • •	105	163.5	14
(020) planes    to direction of current	7.5	92	171.3	20.7

Table I. Comparison of resistivity data for highly oriented and randomly oriented alpha plutonium.

<sup>a</sup>Resistivity maximum is flat in this region.

pendicular or parallel to the long axis of the rod and, consequently, to the flow of the electrical current. The randomly oriented specimen was made by casting the metal in vacuo in an induction furnace. High-purity electrorefined plutonium was used for all specimens. The densities of the oriented and cast specimens were 19.80 and 19.67 g/cc, respectively.

The resistivity curves,  $\rho$  vs T and  $\rho/T$  vs T, are shown in Fig. 1 and the data are summarized in Table I. It can be seen that the electrical behavior of alpha plutonium is highly anisotropic. The oriented and randomly oriented specimens show certain similarities at low temperatures, but the absolute magnitudes and the temperature dependences of their resistivities are very different. In plutonium with its (020) planes aligned parallel to the direction of current flow, the resistivity maximum is much greater and occurs at a lower temperature than in the randomly oriented plutonium. On the other hand, in plutonium with its (020) planes aligned perpendicular to the direction of current flow, the resistivity maximum is much smaller and occurs at a higher temperature. The negative temperature coefficient of resistivity between room temperature and the temperature at  $\rho_{max}$  is greatest for the parallel alignment and least for the perpendicular alignment. Resistivity curves for plutonium with these two preferential orientations combine to give a net curve approximately equivalent to the one obtained for randomly oriented plutonium. Residual resistivities were about the same magnitude for both oriented specimens, and are the lowest that have been reported for alpha plutonium.

Plots of the ratio ho/T vs temperature have a pronounced peak near 50°K for both oriented and randomly oriented plutonium (see Fig. 1). This peak corresponds in temperature with anomalies observed in the thermal expansion, theormoelectric power, Young's modulus,<sup>2</sup> and specific heat.<sup>3</sup> It has been noted,<sup>4</sup> however, that although a number of specific heat "anomalies" have been reported at low temperatures, none have been found to be consistently and reliably reproducible. The indicated transition at 50°K does not now appear to be associated with changes in crystal structure or crystal symmetry. It can be seen from the figure that the peak is more pronounced for the parallel orientation and less pronounced for the perpendicular and random orientations.

The sharp drop in resistivity below  $105^{\circ}$ K has been discussed by various workers as being due to (1) atomic order-disorder changes, (2) interband scattering, and (3) magnetic ordering,<sup>4</sup> but the true nature of the electrical conduction mechanism in plutonium remains to be established.

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