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#### TENTH QUARTERLY TECHNICAL REPORT

"Growth and Characterization of Single Crystals of Rare Earth Compounds" Contract AF 49(638)-1245

### Synopsis

Magnetization measurements were made on the Gd-Nd alloys to ascertain the mode of the spin coupling between a heavy and a light rare earth atom. The magnetic data have substantiated the ferromagnetic spin coupling assumed in the Dekker theory of electrical resistivity of the rare earth alloys.

It was further disclosed in a scientific paper that the Dekker theory is inapplicable to approximately half of the possible intra-rare earth alloys. Its failure reflects a lack of understanding of the light rare earth ions. A remedy of the theory necessitates a major change of the analysis basing on the actual energy-level structure of these ions, which is not known at the present time.

Low-temperature resistivities were determined of several Pd alloys containing solute atoms of Gd, Rh, an Fe. The electrical data have reconfirmed the absence of the abnormally large spinpolarization effect of the localized moments of the Fe atoms in Pd.

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Experimental efforts were made to explore the possible occurrence of resistance minimum in the Pd-Gd alloys.

The effect of local modes at the impurity centers was examined upon the scattering of electrons in aluminum between  $4.2^{\circ}$  and  $300^{\circ}$ K.

### (I) Exchange Scattering of the Conduction Electrons in Rare Earth Alloys

A scientific paper was prepared to disclose the serious nature of the deficiency of the Dekker theory of electrical resistivity of the rare earth alloys (Phys. Stat. Sol. 7, 241 (1964)). Prior to the present work, Dekker's analysis was hailed as the best current theory in the framework of the first Born approximation.

Since large discrepancies have been found between the theoretical predictions and the resistivity data for alloys of Gd and Dy containing light rare earth solutes, it is inferred that Dekker's theory is inapplicable to roughly half of the possible intra-rare earth alloys.

Among the assumptions involved in the theory, the mode of the spin coupling between the parent and foreign atoms has a direct effect on the outcome of the calculation of the impurity-induced resistivity  $\Delta \rho$  at 0°K. A ferromagnetic spin coupling was assumed in the original analysis, disregarding the atomic species of the solute. For alloys

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containing only heavy lanthanides, this assumption is readily justified by virtue of the superb agreement between the theoretical and the experimental results and because  $\underline{J} = \underline{L} + \underline{S}$ , where  $\underline{L}$ ,  $\underline{S}$ , and  $\underline{J}$  are the orbital, spin, and total angular momenta of an atom. For alloys containing light rare earth solutes, however, there exists another possible mode of spin coupling. Therefore, the situation deserves a careful examination.

As  $\underline{J} = \underline{L} - \underline{S}$  for Ce-Sm, the ferromagnetic coupling may occur between the magnetic moments, instead of the spins, of a heavy and a light rare earth atom. In this alternative scheme, the spin coupling between the two atoms becomes antiferromagnetic. It is then easy to show that a reversal of the spin coupling from ferromagnetic to antiferromagnetic would cause a corresponding change of the sign in the original term leading to the component of  $\Delta \rho$  due to exchange scattering. To test this possibility, we have plotted the deduced values of  $\Delta \rho$ from the 4.2°K resistivity data against  $|S_a - (\frac{J_b}{J_b+1})S_b|^2$  where S and J are the quantum numbers of <u>S</u> and <u>J</u>, the subscripts a and b refer to the solvent and solute atoms respectively, and a plus sign is inserted as a result of the antiferromagnetic spin coupling. For both Gd and Dy alloys, an excellent linear relationship emerges in the plot for three

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(Sm, Nd, and Pr) of the five solutes under consideration. Meanwhile, such a plot leaves the experimental points for La and Ce to be unreconciled, and also leads to an unreasonable conclusion concerning the role played by the electrostatic scattering. In view of the conflicting evidence furnished by the resistance data, we have subsequently measured the magnetizations of a series of Gd-Nd alloys at and above 77°K. The magnetic data strongly favor the ferromagnetic spin coupling assumed in the Dekker theory.

Obviously, the deficiency of the Dekker theory is originated in the choice of the quantum number representation of the ground state, from which the non-vanishing matrix element of the exchange-scattering perturbation is derived. The Russell-Saunders coupling and consequent, the (L, S, J, and  $M_J$ ) representation are effective for the heavy rare earth ions, but become faulty for the light ions. A correct representation for the latter ions may invoke the j-j coupling, an intermediate coupling, or an admixed ground state, depending on the actual energylevel structure of the ions concerned. The failure of the Dekker theory is therefore a manifestation of the inadequacy of our current understanding of the light rare earth atoms.

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#### (II) Low-Temperature Electrical Resistivities of Palladium Alloys

Resistance measurements were made on Pd alloys containing solute atoms of Fe, Rh, and Gd between 4.2° and 300°K. All these solute atoms have been shown to carry localized moments consistent with their electronic configurations of the incomplete filled magnetic shells. The alloys are considered to be suitable for a study of the correlation between the magnetic and electrical properties.

Preliminary results of the measurements at 4.2°K gave the following values for the increase in the residual resistivity of Pd caused by 1 at. % of solute: 1.3, 2.0 and 3.4 microhm-cm for Rh, Fe, and Gd respectively. The relatively small value for Fe implies that, even though Pd and Pt belong to the same column in the periodic chart, the pronounced spin polarization of the parent lattice caused by the localized moments of the Fe atoms does not occur in Pc as in Pt.

Experimental efforts were made to determine the temperature dependence of resistivities of the Pd-Gd alloys between  $1.8^{\circ}$  and  $25^{\circ}$ K. According to Crangle (Phys. Rev. Letters <u>13</u>, 569 (1964)), these alloys become ferromagnetic at very low temperature. For example, the Curie temperature for the Pd-0.5% Gd alloy is  $2.5^{\circ}$ K. The low Curie temperature coupled with the localized moment being carried by atoms of a

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rare earth element makes the alloys especially interesting in the exploration of the onset of resistance minimum relative to the new theory recently proposed by Kondo (Prog. Theo. Phys. <u>32</u>, 37 (1964)).

## (III) Effect of Local Modes on the Electrical Resistivity of Dilute Aluminum Alloys

Work was continued on a search for the effect of local modes at the impurity centers upon scattering of the conduction electrons. Attempts to prepare dilute aluminum alloys containing Gd or Lu as little as 0.05 at. % were proved to be futile. In fact, the addition of either lanthanide to Al led to a considerable decrease of the resistivity of Al at 4.2°K, indicating that these rare earth elements probably acted as oxygen getters to exert a purifying effect on Al. Experimental studies have since been limited to the Al-Ag and Al-Mg alloys.

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