in Fe and Co the Anderson $s-d$ mixing effect is strong enough to produce a negative polarization in the conduction electrons. These calculations refer to quite different materials, of course, in particular to ferromagnets rather than paramagnets, but it seems likely that the same mechanism could be invoked to explain the negative polarization your experiments indicate.

C. KITTEL: It would appear possible to determine the sign of the interaction from the positive and negative of the Faraday rotation associated with the exchange frequency resonance. The g-values of the s and f electrons being quite different (except with Gd), the exchange frequency resonance should be quite intense.

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Magnetic Properties of Rare Earth Aluminum Compounds with $MgCu_2$ Structure*

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The magnetic moments of some RAl_2 (R=rare earth element) cubic Laves phase compounds have been measured at temperatures from 1.4°K to 300°K. The measurements indicate that the spin moments of the rare earth ions are coupled ferromagnetically. The Curie points of the RAl_2 compounds are found to be uniformly higher than the corresponding Laves compounds, ROs_2, RIr_2 and RRu_2. Solid solutions of some of the compounds were also investigated. For example, in the $Gd_xPr_{(1-x)}Al_2$ compounds, the magnetic moments of the Gd ions are antiparallel to those of the Pr ions because J is antiparallel to S in the ground state of the Pr ion. Compensation points were observed in this system.

Introduction

Recently Jaccarino *et al*[1]., have determined the magnitude and sign of the conduction electron polarization in a series of rare earth aluminum compounds having the cubic Laves structure, RAl_2 (R = rare earth element)[2,3]. Their measurements indicate that there is a negative (*i. e.*, antiferromagnetic) exchange interaction between the localized f electrons and conduction electrons. It is possible that the predominant rare earth exchange interaction occurs through the conduction electrons. It is therefore of interest to determine whether the coupling between the rare earths is ferromagnetic or antiferromagnetic. This paper contains the results of such a study. The structure of these compounds is shown in Fig. 1.

In the present investigation we have measured the magnetic moments and Curie temperatures of these compounds and of some solid solutions between these compounds[4]. The measurements were made with a pendulum magnetometer[5] at temperatures ranging from 1.4° to 300°K and with applied fields up to 14,000 oersteds.

Magnetic Moments of the Compounds

The magnetic moments per formula unit for the compounds $CeAl_2$, $PrAl_2$, $NdAl_2$, $SmAl_2$, $GdAl_2$, $TbAl_2$, $DyAl_2$, $HoAl_2$, $ErAl_2$, $TmAl_2$ and $YbAl_2$ are shown in Fig. 2. The shape of this curve is similar to a curve of the moments of the free trivalent ions but the measured values are always

* Read by V. Jaccarino.

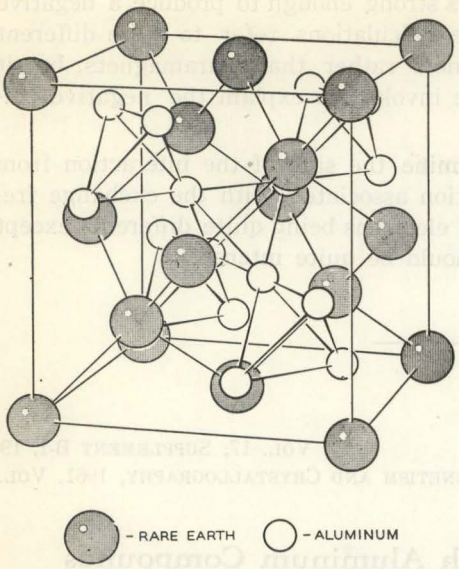

Fig. 1. Structure of the RAl_2 (R=rare earth element) compounds.

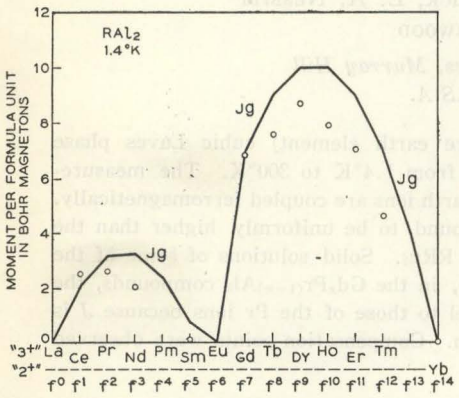

Fig. 2. Measured moments of the RAl_2 compounds compared with the theoretical values (solid line).

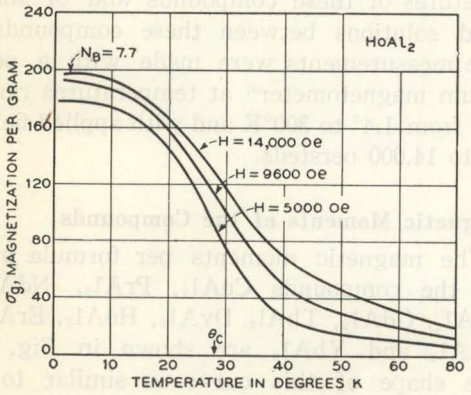

Fig. 3. A typical set of magnetization vs temperature curves.

lower. For the compound $GdAl_2$, a value of 6.83 Bohr magnetons was obtained which is close to the value of 7.0 for gadolinium. For the Compound $HoAl_2$, a value of 7.86 Bohr magnetons was obtained which differs from the value of 10.0 for holmium by approximately 21 per cent. These values and all other values reported were obtained by extrapolating σ vs $1/H$ from a field of 14000 oersteds to infinite field. In the case of $YbAl_2$ the zero moment is attributed to Yb being divalent and therefore having the $4f$ shell filled. Haszko[3] has shown that the lattice constants for $YbAl_2$ and $EuAl_2$ are larger than one would expect from the extrapolation of a plot of lattice constant vs rare earth atomic number for this series of compounds[2,3]. This indicates that both Yb and Eu are divalent in these compounds at room temperature whereas the other rare earth ions are trivalent.

A set of magnetization vs temperature curves for $HoAl_2$ is shown in Fig. 3. This set is typical of most of the compounds tested. The shape of the curves indicates that the compound is ferromagnetic while the spread between the curves for the three different field strengths show that the compound is difficult to saturate.

Curie Temperatures of the Compounds

The ferromagnetic Curie temperatures are plotted in Fig. 4. In general, the values

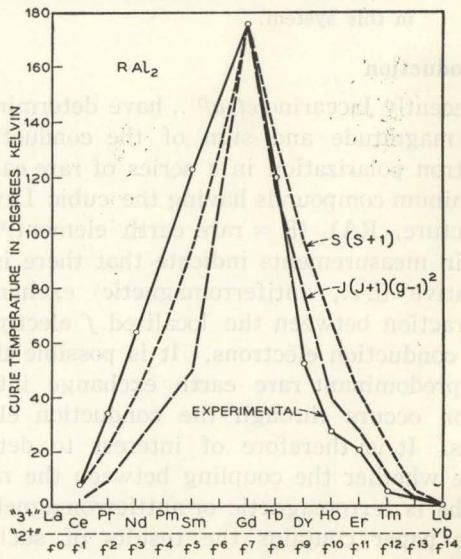

Fig. 4. Experimental values of the Curie temperatures compared with theoretical values.

were obtained by plotting H/σ vs σ^2 isotherms[6]. The values obtained for some of these compounds are surprisingly high e.g., the Curie points of $SmAl_2$ and $NdAl_2$ are 122° and 68°K respectively. Published data for Sm and Nd indicates that these metals are antiferromagnetic with Néel points of 15° and 7.5°K respectively. The Curie points of these Al_2 compounds are uniformly higher than those obtained on the corresponding Laves phase compounds of ROS_2, RIr_2 and RRu_2 previously measured by Bozorth *et al*[7]. For example, they obtained a Curie point of 88°K for $GdIr_2$ whereas we obtained a Curie point of 176°K for $GdAl_2$. This difference in the Curie points of the two systems is probably related to the nature of the polarization of the conduction electrons. The shape of the curve for the Curie temperatures vs number of f electrons is compared with the functions $S(S+1)$ and $J(J+1)\ (g-1)^2$ as was done previously[7].

Solid Solution Compounds of Two Rare Earth Elements with Magnetic Moments Antiparallel

A continuous series of solid solutions having the cubic Laves structure occur in the $Gd_xPr_{(1-x)}Al_2$ system[4]. Fig. 5 shows magnetization vs temperature curves for a number of these solid solution compounds. Gd_x .314 Pr.686 Al_2 and $Gd_{.2}Pr_{.8}Al_2$ compounds have compensation points at approximately 1.4° and 45°K respectively, indicating that the magnetic moments of Gd and Pr are opposing. This situation arises from the fact that L and S of the Pr atom are in opposition. The spins of the rare earth atoms couple parallel (ferromagnetically) but the magnetic moment of the compound is proportional to the difference in the moments of the two rare earths. Fig. 6 shows the magnetic moment per formula unit at 1.4°K as a function of x. The experimental values obtained are in good agreement with the curve calculated for the moments of the Pr atoms opposing those of the Gd atoms.

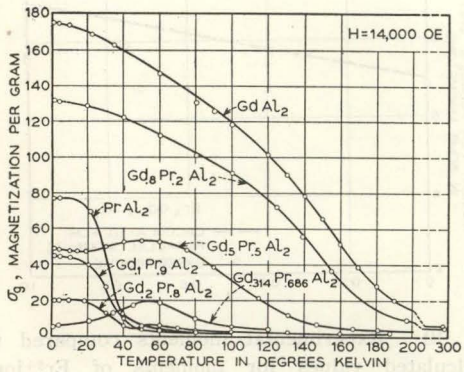

Fig. 5. Magnetization vs temperature curves for the $Gd_xPr_{(1-x)}$ Al_2 system.

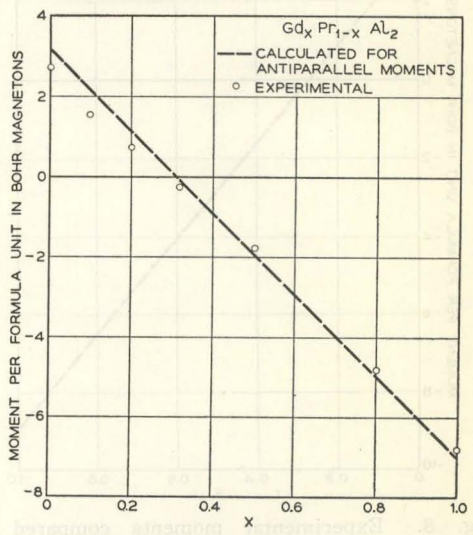

Fig. 6. Experimental moments compared to calculated values for moments of the Gd^{3+} ions antiparallel to moments of Pr^{3+}ions in the Gd_x $Pr_{(1-x)}$ Al_2 system.

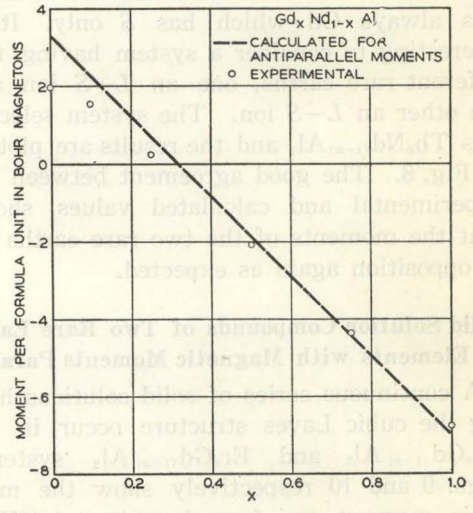

Fig. 7. Experimental moments compared to calculated values for moments of Gd^{3+}ions antiparallel to moments of Nd^{3+}ions in the Gd_x $Nd_{(1-x)}$ Al_2 system.

The solid solution compounds of $Gd_xNd_{(1-x)}Al_2$ behave in a similar manner since the Nd atoms, like the Pr atoms, have L and S in opposition. The data are shown in Fig. 7. The deviation from the theoretical curve is probably due to a lack of saturation.

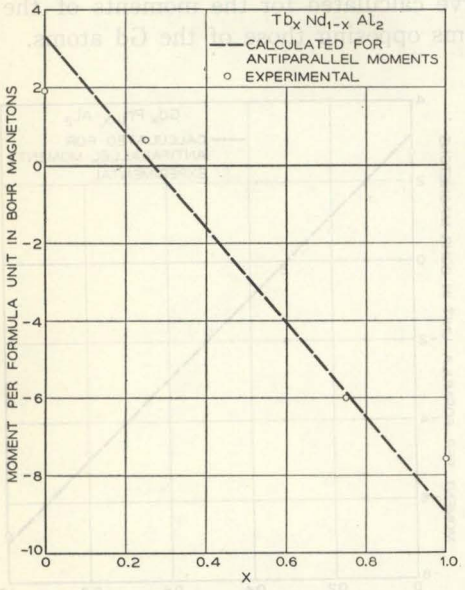

Fig. 8. Experimental moments compared to calculated values for moments of Tb^{3+}ions antiparallel to moments of Nd^{3+}ions in the $Tb_xNd_{(1-x)}Al_2$ system.

The two previous systems involved solid solutions of two rare earths but one of these was always Gd which has S only. It is interesting to consider a system having two different rare earths, one an $L+S$ ion and the other an $L-S$ ion. The system selected was $Tb_xNd_{(1-x)}Al_2$ and the results are plotted in Fig. 8. The good agreement between the experimental and calculated values shows that the moments of the two rare earths are in opposition again as expected.

Solid Solution Compounds of Two Rare Earth Elements with Magnetic Moments Parallel

A continuous series of solid solutions having the cubic Laves structure occur in the $Dy_xGd_{(1-x)}Al_2$ and $Er_xGd_{(1-x)}Al_2$ systems. Figs. 9 and 10 respectively show the magnetic moment per formula unit at 1.4°K as a function of x for both these systems. The experimental values obtained are in reasonable agreement with the curves calculated for the moments of the Dy–Gd and for the moments of the Er–Gd to add. This situation arises from the fact that both Dy and Er are $L+S$ ions and their moment adds to that of Gd which is an S ion.

It is of interest in substantiating the idea that the spins always couple parallel (ferromagnetically) to investigate both a solid solution compound having two $L-S$ ions and one having two $L+S$ ions. The compound having two $L-S$ ions was $Pr._5Nd._5Al_2$ which when measured had 2.39 Bohr magnetons per formula unit indicating that the moments of Pr and Nd have added. This result is in good agreement with the average of the measured values for $PrAl_2$ and $NdAl_2$ of 2.33 Bohr magnetons. The compound having two $L+S$ ions was $Tb._5Dy._5Al_2$ which gave an experimental value of 8.72 Bohr magnetons per formula unit. This value is somewhat

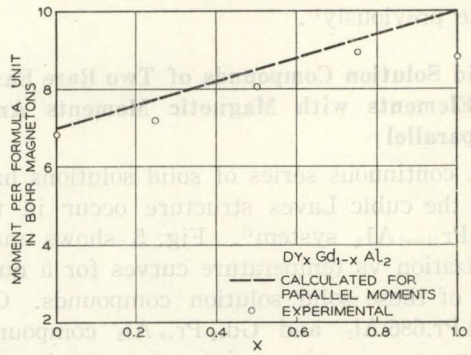

Fig. 9. Experimental moments compared to calculated values for moments of Dy^{3+}ions parallel to moments of Gd^{3+}ions in the $Dy_xGd_{(1-x)}Al_2$ system.

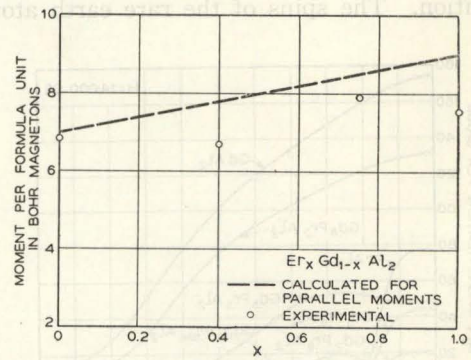

Fig. 10. Experimental moments compared to calculated values for moments of Er^{3+}ions parallel to moments of Gd^{3+}ions in the $Er_xGd_{(1-x)}Al_2$ system.

higher than 8.21 Bohr magnetons the average of the measured values for $TbAl_2$ and $DyAl_2$.

Conclusions

This investigation shows that the spin moments of the rare earth ions couple ferromagnetically in the RAl_2 (R=rare earth element) compounds and solid solution compounds containing two different rare earth ions. The NMR results of Jaccarino *et al.* indicate that the rare earth spins, S, polarize the conduction electron spins, s, in such a manner that the spins of the rare earth ion and the conduction electrons are antiferromagnetically aligned. However, if the interaction between the rare earth spins proceeds via the conduction electrons the net exchange $JS_i \cdot S_j$ will be *ferromagnetic* irrespective of the sign of the rare earth conduction electron interaction. Consequently our results are consistent with the NMR results.

Solid solution compounds containing two ions of similar type such as two $L-S$ ions or two $L+S$ ions including Gd (S only) with the latter have a magnetic moment proportional to the sum of the moments of the two sets of rare earth ions. Solid solution compounds containing two ions of dissimilar type have a moment proportional to the difference of the moments of the two sets of rare earth ions. Some of the solid solution compounds have compensation points of the σ vs T curves.

The Curie temperatures of the RAl_2 compounds were found to be uniformly higher than those of the corresponding Laves compounds of ROs_2, RIr_2 and RRu_2.

Acknowledgments

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