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STRUCTURAL AND MAGNETIC STUDIES OF THE PSEUDO-BINARY COMPOUNDS \((\text{Gd}_{1-x}\text{Nd}_x)\text{Co}_2\)

by G. J. PRIMAVESI (*), K. N. R. TAYLOR (*) and I. R. HARRIS (**) 

Résumé. — Nous avons effectué des mesures d’aimantation et de paramètre du réseau pour les composés \(\text{Gd}_{1-x}\text{Nd}_x\text{Co}_2\). On interprète le comportement magnétique en termes de moment bloqué du néodyme dont l’amplitude varie avec la température et la composition. On peut mettre en relations les variations de paramètre du réseau et les données magnétiques.

Abstract. — Measurements have been made of the magnetization and lattice parameters of the compounds \(\text{Gd}_{1-x}\text{Nd}_x\text{Co}_2\). The magnetic behaviour has been interpreted in terms of a quenched neodymium moment whose magnitude varies with temperature and composition. The changes in lattice parameter may be correlated with the magnetic data.

Introduction. — In the course of a series of investigations into the magnetic properties of compounds and alloys formed with the heavy and light rare earth metals, and of the effects of magnetic order on the lattice parameters of Laves phase compounds, we have examined the pseudobinary series \(\text{Nd}_x\text{Gd}_{1-x}\text{Co}_2\).

Experimental. — The techniques used for specimen preparation, the determination of lattice parameters and magnetization measurements have been described previously [I]. Figure 1 shows the variation with neodymium concentration of the room temperature lattice parameters and saturation magnetization at 4.2 °K for \(\text{Gd}_{1-x}\text{Nd}_x\text{Co}_2\).

The lattice parameter variation across the series shows a change in slope at about \(x = 0.33\) and a sudden rise at \(x = 0.57\).

Discussion. — Previous workers [2, 3] have shown that the observed molecular moments of the \(\text{AB}_2\) compounds can be understood in terms of an exchange interaction which causes the spins of the rare earth and transition metal ions to be anti-parallel. With the heavy rare earths this leads to a ferrimagnetic structure and with the light rare earths to ferromagnetism. Use of this model leads to values of the ionic cobalt moment close to 1.0 \(\mu_B\) for \(\text{GdCo}_2\) and 0.8 \(\mu_B\) for \(\text{NdCo}_2\). In addition it is frequently necessary to invoke a crystal field quenched rare earth

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Fig. 1. — Composition dependance of the room temperature lattice parameters and saturation magnetization at 4.2 °K for \(\text{Gd}_{1-x}\text{Nd}_x\text{Co}_2\).

Fig. 2. — Temperature dependance of the remanent magnetization and magnetization at 4 kOe applied field for composition \(x = 0.57\).

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moment to give a favourable comparison with the experimental magnetization data. In this way \( \mu_{Nd} \) in NdCo$_2$ is found to be 2.6 \( \mu_B \). The molecular moment data shown in figure 1 may be examined in terms of these values. For \( x < 0.7 \) the observed results agree well with a total moment for the compound calculated on the basis of \( \mu_{Gd} = 7.0 \) \( \mu_B \), \( \mu_{Nd} = 2.1 \) \( \mu_B \), \( \mu_{Co} \) varying linearly from 1.0 to 0.8 \( \mu_B \), and an antiparallel alignment of the gadolinium moments with respect to the neodymium and cobalt moments. These values, however, predict a NdCo$_2$ moment of approximately 3.7 \( \mu_B \) which is appreciably less than that observed. The neodymium rich specimens \( (x > 0.75) \) are better fitted by \( \mu_{Nd} = 2.6 \) \( \mu_B \), and it would appear that the rapid rise in the total moment in the vicinity of \( x = 0.72 \) originates from this change in the neodymium moment.

The reduced moments of the neodymium ions almost certainly arise because of the effects of crystal field splitting of the \( J = 9/2 \) level of this ion; this has been discussed in the past by Bleaney [4]. In these compounds the exchange field is comparable to the crystal field, and no quantitative calculations of the combined effects of both these fields on the \( J = 9/2 \) manifold have been made.

By computing the energy level diagram for various crystal and exchange field strengths we have been able to account for the behaviour described above. Using the Hamiltonian:

\[
\mathcal{K} = \mathcal{K}_{CF} + \mathcal{K}_{ex}
\]

agreement with experiment is only possible provided the easy direction of magnetization is parallel to the 111 direction. This Hamiltonian has been evaluated for exchange fields up to 2 MOe. The ground state level at \( H_{ex} = 500 \) kOe has a moment of about 2.3 \( \mu_B \), the first excited state lying some tens of degrees above this and having a moment in excess of 3 \( \mu_B \). By allowing a continuous variation of the exchange and crystal field parameters the degree of admixing of the lower levels changes and in NdCo$_2$ can give an increase in the neodymium groundstate moment of 0.5 \( \mu_B \). Although the absolute magnitudes of these moments are higher than the experimental values it is encouraging that moment increases of the type observed can be predicted. Excitation into the excited states is also thought to be responsible for the anomalous results of figure 2. The \( x = 0.57 \) specimen is predicted to have zero magnetization at 0 °K from the measurements made at the gadolinium rich end of the series. This is found to be the case, and with increasing temperature the increase in remanence arises from the detailed differences between the magnetizations of the different sublattices with temperature. This would not normally become zero again until the Curie temperature is reached. However, due to thermal excitation into the excited states of the \( J = 9/2 \) energy level system the neodymium moment is increased and upsets the magnetization balance in the system. Assuming that at low temperatures the net magnetization is parallel to that of the gadolinium sublattice, then the increase in \( \mu_{Nd} \) causes the molecular moment to decrease to zero and subsequently to increase with the net magnetization parallel to the neodymium and cobalt sublattices. We estimate that the change in \( \mu_{Nd} \) is of order 0.5 \( \mu_B \) to account for the total change in remanence between the turning points. Having attained the higher neodymium moment the individual sublattice magnetizations no longer give a compensation point at 0 °K, and it is possible that the second reversal at 205 °K may be the compensation point corresponding to the new moment values.

The magnetization of this specimen increase rapidly for \( T > 100 \) °K, reaching a maximum of 0.34 \( \mu_B \) at 200 °K, which is of the correct order for a neodymium moment change of 0.5 \( \mu_B \).

The change in slope of variation of the lattice parameter with composition at \( x = 0.33 \) coincides with the composition at which the Curie temperature becomes comparable to room temperature. Consequently the gadolinium rich specimens are magnetically ordered at the measuring temperature, while those for \( x > 0.33 \) are not. Changes of this type have been observed previously in other AB$_2$ compounds [1] and may be attributed to magnetostriction effects. The discontinuous change in lattice parameter near to the equilibrium composition may be associated with the changes in the 4f state caused by both composition and temperature.

References