EXCHANGE INTERACTION IN DILUTE YTTRIUM-RARE EARTH ALLOYS*

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ABSTRACT

Dilute alloys of the heavy rare earths Gd, Tb, Dy, and Ho in yttrium all exhibit the identical type of helical long-range magnetic order at low temperature. This fact suggests that the magnetic order in these alloys is determined predominantly by the RKKY exchange interaction $J(q)$ which is proportional to the susceptibility $\chi(q)$ for the conduction electrons of yttrium. In order to obtain information about $J(q)$, the magnetic excitations in the c-direction of Y(10%Tb) and Y(10%Ho) have been studied at 4.7°K by means of neutron inelastic scattering techniques. In spite of the low concentration of magnetic atoms well-defined propagating excitations were observed for both alloys. The data have been analyzed on the basis of a model in which the equations of motion for spins are averaged over randomly distributed sites of magnetic atoms with the assumption that $J(q)$, properly normalized, is common to both alloys. The results show this to be a reasonably good assumption, and the resulting $J(q)$, which may be interpreted to be characteristic of Y, shows a peak at the wave vector of the helical structure which is much larger than those measured in the pure rare earth metals.

INTRODUCTION

The principal mechanism responsible for the complicated magnetic order exhibited by the heavy rare earth metals is believed to be an indirect isotropic exchange interaction which takes place through the conduction electrons. Of central importance is the response of the conduction electrons to the exchange interaction I between the localized 4f spin moments and the conduction electron spins. This response is described by the susceptibility $\chi(q)$ which is related to the energy bands and Fermi surface of the conduction electrons. The Fourier transform of the exchange interaction $j(q)$ between rare-earth ions is given approximately by¹

$$j(q) = I^2(q)\chi(q) . \quad (1)$$

Yttrium while having no 4f electrons, does have an outer electron configuration and electronic energy band structure quite similar to that of the heavy rare earth metals. Thus although

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yttrium is itself non-magnetic, the conduction electron susceptibility provides a mechanism for the existence of a long range exchange interaction between rare earth ion impurities. Indeed rather dilute (e.g., $\sim 2\%$ Tb) quantities of rare earth ions in Y show a long range ordering of their magnetic moments at low temperatures in spite of their random locations in the yttrium structure. Furthermore, the magnetic order of dilute alloys is essentially independent of the particular rare earth impurity in that all such alloys possess oscillatory antiferromagnetic structures which are described by the same periodicity or wave-vector q_0 .^{2,3} Thus the susceptibilities for these alloys are probably very similar and perhaps essentially the same as that of pure yttrium.

As in the case of the pure rare-earth metals, information about $\chi(q)$ for yttrium-rare earth alloys may be inferred from the exchange interaction $j(q)$ which can normally be obtained from neutron inelastic scattering measurements of the spin-wave dispersion relations.⁴ Of course the interpretation of the magnetic excitations in an alloy is itself a non-trivial problem. The substitution of impurities into magnetic insulators⁵ and metals⁴ can strongly perturb the spin waves, resulting in distortions of the host spin-wave dispersion relations as well as producing localized magnetic excitations. In the present case as a starting approximation, we shall ignore such problems. Although the host is non-magnetic and the concentration of magnetic impurities is rather low ($\sim 10\%$) in the samples studied, a good description of the measurements is obtained with a mean lattice model. Since the exchange interaction is long range, the effects of the variations of the local environment around different magnetic ions may be small. Thus, in the magnetic Hamiltonian which we assume to be

$$H = B \sum_i^{\text{mag}} S_{i\zeta}^2 - \frac{1}{2} \sum_{i,j}^{\text{mag}} J_{ij} \vec{S}_i \cdot \vec{S}_j, \quad (2)$$

the summations over the sites of the magnetic ions are replaced by summations over all atomic sites by merely taking into account the probability, c , that a site is occupied by a magnetic ion, viz,

$$H = cB \sum_i^{\text{all}} S_{i\zeta}^2 - c^2 \frac{1}{2} \sum_{i,j}^{\text{all}} J_{ij} \vec{S}_i \cdot \vec{S}_j. \quad (3)$$

Here J_{ij} is the exchange interaction, B defines the single-ion crystal field anisotropy, c is the concentration of magnetic ions, and ζ is in the c -direction of the (HCP) yttrium structure. The spin-wave energies along the c -direction are then given by

$$\hbar\omega(q) = S \left[\{cJ(q_0) - cJ(q) + 2B\} \{cJ(q_0) - \frac{1}{2}cJ(q_0+q) - \frac{1}{2}cJ(q_0-q)\} \right]^{1/2}, \quad (4)$$

where S is the total angular momentum so that $J(q) = (g-1)^2 j(q)$ and g is the Landé factor. This is the same expression usually used for the pure metals in their helical phases⁴ except the exchange interaction is reduced by the factor c .

EXPERIMENT

A triple axis neutron spectrometer at the Oak Ridge HFIR was used to measure the spin-wave energies at 4.7°K along the c -direction for two yttrium-rare earth alloys; one containing 9.8 at.% Tb and one containing 9.5% Ho. The Néel temperatures were observed to be approximately 54°K for Y(Tb) and 20°K for Y(Ho). The measurements were performed with the energy of the scattered neutrons fixed at 12.8 meV and with Be crystals as monochromator and analyzer. Graphite filters were placed between the sample and the analyzer.

In spite of the low concentration of magnetic ions, rather well defined excitations were observed, having energy widths of typically 0.5 meV. The spin-wave dispersion curves obtained from the measurements are shown in Figs. 1 and 2. Compared to the results for the helical magnetic phases of the pure rare earth metals,^{4,6} these curves have much larger initial slopes and much less dispersion at intermediate wave-vectors.

ANALYSIS AND DISCUSSION

In addition to our assumption that $\chi(q)$ for both alloys is nearly equal to that for pure Y, we also assumed that the interaction $I(q)$ in Eq. (1) is the same for both Tb and Ho. If these assumptions are valid, the spin-wave measurements for both alloys can be described by the same $j(q)$ function. Therefore, with $j(q)$ expressed in terms of interplanar exchange constants, j_ℓ , i.e.,

$$J(q) = (g-1)^2 j(q) = 2(g-1)^2 \sum_{\ell} j_{\ell} \cos(\pi \ell \tau) , \quad (5)$$

the measured dispersion curves have been fitted by a least squares procedure to eq. (4) with j_ℓ 's, taken to be common to both alloys, and the anisotropy constants B 's as the fitting parameters. The overall agreement between the experimental results and the calculations, shown as the full lines in Figs. 1 and 2, is quite good thereby substantiating our assumptions.

The anisotropy constants obtained in the fitting analysis are in fair agreement with those measured for the pure rare earth metals.^{4,6,7} Of particular interest, however, is the exchange function $j(q) = J(q)/(g-1)^2$ deduced from the measurements. This is shown in Fig. 3 together with earlier results for pure Ho,⁶ which possesses the largest peak $J(q_0) - J(0)$ of the pure metals. Such a peak is necessary for the existence of a helical magnetic structure. It is apparently much larger in the yttrium-rare-earth alloys, indicating that $\chi(q_0) - \chi(0)$ is much larger for Y than for the rare earths.

This conclusion is in good qualitative agreement with recent $\chi(q)$ calculations by Liu *et al.*,⁸ and it also appears to be consistent with other experimental results. For example, when the rare-earths are diluted with yttrium the tendency toward helical magnetic ordering increases strongly with increasing yttrium concentration.² The ferromagnetic structures of Tb and Dy, both of which also have helical structures, are completely suppressed at Y concentrations of 24% and 5%, respectively. In Gd which is ferromagnetic only, helical ordering occurs in Gd-Y alloys at an Y concentration of $\sim 20\%$, and ferromagnetism is completely suppressed at ~ 60 at.% Y.³ It is tempting to explain such structure properties in terms of a concentration dependent $J(q)$, or $\chi(q)$, for which $\chi(q_0) - \chi(0)$ grows with yttrium concentration. Such a model would qualitatively explain also the results of Belov *et al.*⁹ who find that although the ordering temperatures for Tb_cY_{1-c} alloys decrease as expected with decreasing c , the magnetic field which is required to transform their helical structures to ferromagnetic structures increases strongly with decreasing c for $0.36 \leq c \leq 1.0$.

One consequence of the mean lattice assumption employed here is that single-ion interactions such as the crystal field, are proportional to c , whereas two ion interactions such as the exchange are proportional to c^2 . In the limit of small c the crystal field contribution to the magnetic Hamiltonian can become comparable to the exchange interaction so that the conventional spin-wave theory we used may not be justified. To investigate this aspect of the data analysis, the crystal field energy levels for a point charge model in a mean field approximation were calculated for both Y(Tb) and Y(Ho) alloys at various temperatures and for various values of $J(q_0)$ and B which gave approximately the observed Néel temperatures. The ground states at 0°K were found to have essentially the maximum S_z for both alloys, ~ 6 for Tb and ~ 8 for Ho, thereby justifying the procedures used in the present analysis.

SUMMARY

Neutron inelastic scattering measurements on dilute ($\sim 10\%$) alloys of the heavy rare-earths Tb and Ho in yttrium metal show the existence of rather well defined propagating spin-waves. The resulting dispersion relations are satisfactorily described by a mean lattice model with the assumption that the exchange interaction, $j(q)$, is essentially proportional to the conduction electron susceptibility $\chi(q)$ of yttrium and, hence, is common to both alloys. The $j(q)$ function deduced from the data possesses a peak at the wave vector of the helical magnetic structures of the alloys which is much larger than those measured for the pure rare earth metals. This result is consistent with theoretical calculations of $\chi(q)$ from the electronic energy bands for yttrium and with measurements of the magnetic structures of yttrium-rare earth alloys as a function of composition, temperature, and applied magnetic field.

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REFERENCES

1. W. E. Evensen and S. H. Liu, Phys. Rev. 178, 783 (1969).
2. H. R. Child, W. C. Koehler, E. O. Wollan, and J. W. Cable, Phys. Rev. 138, A1655 (1965).
3. H. R. Child and J. W. Cable, J. Appl. Phys. 40, 1003 (1969).
4. A. R. Mackintosh and H. Bjerrum Møller, chapter 5 in Magnetic Properties of Rare Earth Metals, edited by R. J. Elliott (Plenum Press, New York, 1972).
5. R. A. Cowley and W. J. L. Buyers, Rev. Mod. Phys. 44, 406 (1972).
6. R. M. Nicklow, J. Appl. Phys. 42, 1672 (1971).
7. J. J. Rhyne, chapter 4 in Magnetic Properties of Rare Earth Metals, edited by R. J. Elliott (Plenum Press, New York, 1972).
8. S. H. Liu, R. P. Gupta, and S. K. Sinha, Phys. Rev. B 4, 1100 (1971).
9. K. P. Belov, R. Z. Levitin, S. A. Nikitin, L. I. Solvtseva, Soviet Phys. JETP 27, 207 (1968).

FIGURE 1. SPIN-WAVE ENERGIES MEASURED FOR Y(9.8 AT.% Tb) ALONG c -DIRECTION. THE FULL CURVE IS A FIT TO THE DATA AS DESCRIBED IN THE TEXT.

FIGURE 2. SPIN-WAVE ENERGIES MEASURED FOR Y(9.5 AT.% Ho) ALONG c -DIRECTION. THE FULL CURVE IS A FIT TO THE DATA AS DESCRIBED IN THE TEXT.

FIGURE 3. THE EXCHANGE INTERACTION $J(q)/(g-1)^2$ FOR DILUTE Y(RE) ALLOYS AND FOR Ho METAL.