excitation of halide crystals if the exciting laser wavelength overlaps transitions of the metastable relaxed excitons.

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Spin Waves in the Cubic Ferrimagnet $Ho_{0.88}Tb_{0.12}Fe_2$ †

R. M. Nicklow
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
and
N. C. Koon, C. M. Williams, and J. B. Milstein
Naval Research Laboratory, Washington, D. C. 20390
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Using neutron spectrometry we have investigated the magnon dispersion relation for the ferrimagnetic cubic Laves compound $Ho_{0.88}Tb_{0.12}Fe_2$ at room temperature and 12 K. The results are described surprisingly well by a very simple near-neighbor exchange model which includes only iron-iron and iron–rare-earth interactions, crystal field anisotropy, and temperature renormalization that is derived from the sublattice magnetization.

The magnetic properties of the rare-earth–iron compounds RFe_2, both in the crystalline Laves phase ($MgCu_2$ structure) and in the amorphous form, are currently of considerable interest. Extensive investigations of the amorphous form of $TbFe_2$ have recently been carried out by magnetic-susceptibility,[1,2] neutron-scattering,[2-4] and magnetic-resonance[5] measurements. These studies may be particularly valuable for the understanding of amorphous magnetism because of the possibility of comparing properties of the crystalline and amorphous forms of the same compounds. While such comparisons have led to new theoretical models,[6] they have depended on somewhat uncertain assumptions and speculations about the nature and magnitude of the exchange and anisotropy interactions in these compounds. Similarly, interpretations of the unusual spin-orientation phase diagrams for crystalline ternary alloys of the type $(R^1)_x(R^2)_{1-x}Fe_2$, recently measured as a function of x and temperature by Mössbauer-effect[7] and torque-magnetometry[8] techniques, have been based on incomplete information about these magnetic interactions.

In this Letter we present the first measurements of the magnon dispersion relation in a rare-earth–iron compound—$Ho_{0.88}Tb_{0.12}Fe_2$. To date, there have been no other experimental results which could give direct information on the exchange interactions in these materials. The only previous data on the magnetic excitations were obtained from neutron scattering on polycrystalline and amorphous samples of $TbFe_2$.[2,4]

The experiment was carried out on a three-axis neutron spectrometer at the high-flux isotope reactor of the Oak Ridge National Laboratory, by using the standard techniques of coherent neutron inelastic scattering. The energy of

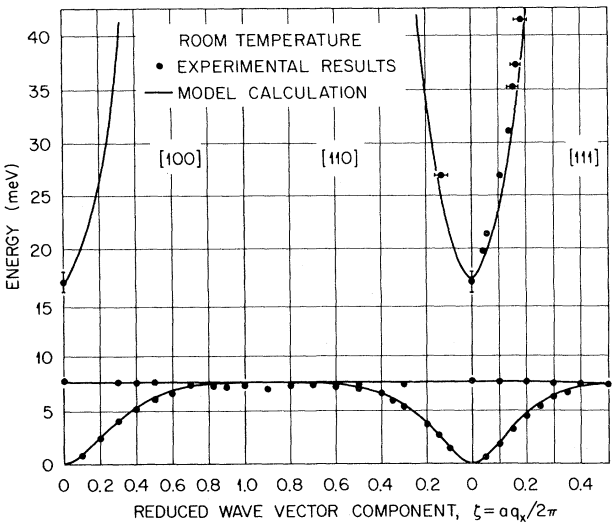

FIG. 1. Magnon dispersion relation for $Ho_{0.88}Tb_{0.12}Fe_2$ measured at room temperature.

the scattered beam was fixed at several values in the range 13–60 meV to optimize resolution and intensity for the different branches of the dispersion relation. Measurements were carried out at 12 K and room temperature. The sample was a single crystal (~0.6 cm^3) grown by a modified Czochralski method.[8,9]

In the cubic Laves phase there are six atoms, two RFe_2 units, in the primitive cell. The relative positions of the rare-earth and iron ions in the crystal lattice are identical to those of the so-called A and B sites, respectively, of the iron ions in Fe_3O_4.[10] For $HoFe_2$ the rare-earth and iron magnetic moments have magnitudes of approximately $10\mu_B$ and $1.7\mu_B$, respectively, at 4.2 K, and they are aligned antiparallel at all temperatures up to the Curie temperature (~610 K).[11] For the alloy studied here the easy magnetization direction changes from [110] to [100] at about 25 K as the temperature is raised above 4 K.

The nearly isotropic behavior of the alloy is evident in the magnon dispersion relation measured at room temperature which is shown in Fig. 1. In particular, the acoustic branch possesses a negligible energy gap (< 0.2 meV) at $|\vec{q}| = 0$, and for small $|\vec{q}|$ it is observed to be isotropic with nearly quadratic dispersion; i.e., $\hbar\omega \approx D|\vec{q}|^2$, and $D = 84$ meV Å^2.

Since there are six atoms in the unit cell, one expects six branches for the spin-wave dispersion relation, one acoustic and five optic. Qualitative considerations of the neutron scattering structure factors and theoretical calculations in-

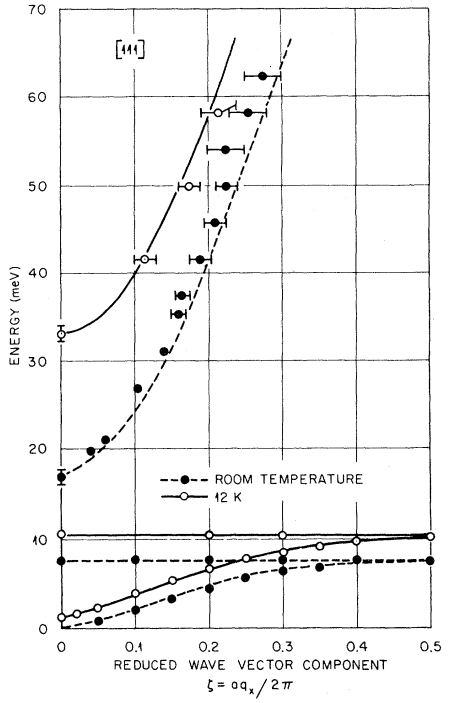

FIG. 2. Temperature dependence of the magnon dispersion relation in the [111] direction for $Ho_{0.88}Tb_{0.12}Fe_2$.

dicate that the optic branch at 7.5 meV in Fig. 1 represents the out-of-phase precession of the spins on the two rare-earth sublattices in the mean field of the iron sublattice. The nearly negligible q dependence of the branch shows immediately that the R-R exchange interaction is very small.

The second optic branch shows an extremely strong quadratic dispersion, with $D \simeq 290$ meV Å^2, very similar to that exhibited by the acoustic spin waves in pure iron metal[12] for which $D \simeq 280$ meV Å^2. This branch has been most extensively investigated in the [111] direction up to an energy of about 65 meV ($\zeta \simeq 0.28$, see Fig. 2). Because of the relatively small size of the sample (~0.60 cm^3), we have not been able to extend these measurements to the zone boundary. Therefore, we cannot yet determine whether this branch exists as a well-defined excitation throughout the zone or whether it eventually "vanishes" as it enters a region of a high density of Stoner excitations as in pure iron.[12] The three remaining optic branches are apparently even higher in energy and have not been measured.

In Fig. 2 the results obtained for the [111] direction at 12 K are compared to room-temperature data. The magnetic anisotropy increases as the temperature is lowered, giving rise to a

significant energy gap (~1.3 meV at 12 K) in the acoustic magnon branch. In addition, there is a strong temperature dependence of the two measured optic modes.

As a first step in the interpretation of these results, we have carried out conventional spin-wave calculations on the basis of a Hamiltonian with the form

$$\mathcal{H} = \sum_{n,m}\sum_{\alpha\beta} J(n\alpha; m\beta)\vec{S}_{n\alpha}\cdot\vec{S}_{m\beta} + \mathcal{H}_{cf}, \tag{1}$$

where $J(n\alpha; m\beta)$ is the isotropic exchange interaction between the αth ion in unit cell n, with spin $\vec{S}_{n\alpha}$, and the βth ion in unit cell m. For our purpose crystal field anisotropy, $\mathcal{H}_{cf}$, is appreciable only for the rare-earth ions ($\alpha = 1, 2$) and contributes to the spin-wave energies only at low temperatures where the magnetization is in the [110] direction. Thus we assume

$$\mathcal{H}_{cf} = -\sum_{n}\sum_{(\alpha=1,2)}\left[\tfrac{1}{4}B_4^{\,0}(O_4^{\,0} - 20O_4^{\,2} - 15O_4^{\,4}) + \tfrac{13}{8}B_6^{\,0}(O_6^{\,0} + \tfrac{105}{26}O_6^{\,2} + \tfrac{231}{26}O_6^{\,6})\right],$$

where the $O_l^{\,m}(\vec{S}_{n\alpha})$ are the usual spin-operator equivalents. The spin-wave energies have been calculated from the Fourier transformations of the equations of motion for the spin operators $S^{\pm} = S^x \pm iS^y$, as outlined by Goodings and Southern.[13] This requires the solution of six equations of the form

$$\hbar\omega(\vec{q})S_\alpha^{\,+}(\vec{q}) = -\sum_{\beta=1}^{6}\{S_\alpha^{\,z}J_{\alpha\beta}(\vec{q})S_\beta^{\,+}(\vec{q}) + [\Delta^+\delta_{\alpha,i} - S_\beta^{\,z}J_{\alpha\beta}(0)]S_\alpha^{\,+}(\vec{q})\} + \Delta^-\delta_{\alpha,i}S_\alpha^{\,-}(\vec{q});$$
$$\alpha = 1, 2, \ldots, 6; \quad i = 1, 2; \tag{2}$$

together with six equations having $S^{\pm} \to S^{\mp}$. The α and β subscripts label the six sublattices, and

$$J_{\alpha\beta}(\vec{q}) = \sum_{m\beta} J(n\alpha; m\beta)\exp[i\vec{q}\cdot(\vec{r}_{m\beta} - \vec{r}_{n\alpha})],$$

$$\Delta^+ = -20\langle B_4^{\,0}S(\tfrac{3}{2})\rangle - 546\langle B_6^{\,0}S(\tfrac{5}{2})\rangle, \tag{3}$$

and

$$\Delta^- = -60\langle B_4^{\,0}S(\tfrac{3}{2})\rangle + 210\langle B_6^{\,0}S(\tfrac{5}{2})\rangle,$$

where

$$S(n/2) = (S - \tfrac{1}{2})(S - 1)\cdots(S - n/2).$$

The $\langle \ldots \rangle$ are averages, over the concentration, of the corresponding quantities appropriate to the rare-earth ions Tb and Ho.

This theory is a slight extension of the near-neighbor theory developed by Glasser and Milford[10] for Fe_3O_4 to include crystal field anisotropy and exchange interactions to more distant neighbors. A surprisingly good description of the data, illustrated in Figs. 1 and 2, can be obtained with a very simple model which includes crystal field anisotropy and only first-neighbor iron-iron, J_{FeFe}, and iron–rare-earth, J_{RFe}, exchange interactions. For this model the locations and shapes of the calculated branches are very sensitive to the parameter values. For example, in the absence of anisotropy at room temperature the energies of the lower and upper optic branches at zero wave vector are given by $E_1^{\,op} = 12J_{RFe}S_{Fe}^{\,z}$ and $E_2^{\,op} = 6J_{RFe}(S_R^{\,z} - 2S_{Fe}^{\,z})$, respectively, where $S_{Fe}^{\,z}$ is the "z component" of the effective iron "spin" and $S_R^{\,z}$ is that for the total angular momentum for the rare-earth ions. The shape of $E_2^{\,op}(\vec{q})$ is determined primarily by J_{FeFe}.

The measured temperature variation of the acoustic and lower optic branches is almost entirely due to the strongly temperature-dependent contributions of the crystal field anisotropy to these branches. On the other hand, the strong temperature variation of the upper optic mode, $E_2^{\,op}(q)$, is more naturally explained in terms of the temperature dependence of $S_R^{\,z} - 2S_{Fe}^{\,z}$; the crystalline anisotropy contributes very little to this branch. Indeed, the observed temperature dependence of this branch is satisfactorily reproduced by the assumption that $S_R^{\,z}(T) = S_R^{\,z}(0)\times\sigma_R(T)$ and $S_{Fe}^{\,z}(T) = S_{Fe}^{\,z}(0)\sigma_{Fe}(T)$, where, for example, $\sigma_R(T)$ is the relative magnetization for the rare-earth sublattice.[11] One is also led to the conclusion that if $S_R^{\,z}(0) \simeq 8$ then $S_{Fe}^{\,z}(0) \simeq 0.81$. While this result is perhaps not too surprising, since it is consistent with the measured iron magnetic moment of $\sim 1.7\mu_B$,[11] it is interesting that both the static and the dynamic magnetic properties of the compound are described by the same effective nonintegral "spin" for the iron ions.

The values for the exchange and anisotropy interactions deduced from these data are given in Table I. A negative exchange parameter indicates ferromagnetic coupling. The value deduced for the iron-iron interaction is qualitatively consistent with that appropriate to pure iron metal. This interaction leads to the prediction that the

TABLE I. Model parameters used for the calculated magnon dispersion relations shown in Figs. 1 and 2.

Parameter	Value (meV)
J_{FeFe}	-46 ± 3
J_{RFe}	0.84 ± 0.06
J_{RR}	0.00 ± 0.01
Δ^-	-2.5
Δ^+	2.3

unobserved higher-energy optic branches for spin waves on the iron sublattice have energies in the 200–350-meV range. These conclusions related to the interactions between iron ions should be interpreted with caution since it is highly unlikely that this simple model of exchange interactions between localized moments is appropriate for describing the spin waves on the iron sublattice, in view of the probable importance of electron-band effects on the magnetic excitations (such as Stoner modes) as in the case of pure iron metal.[12]

The magnitude of the rare-earth–iron exchange interaction obtained from these measurements is in reasonable agreement with the value (0.67 meV) calculated from $12J_{R\mathrm{Fe}}S_{\mathrm{Fe}}{}^{z} = 2(g-1)\mu_B H_{\mathrm{exch}}$ and reported estimates of the exchange field, $\mu_B H_{\mathrm{exch}}$, deduced from Mössbauer-effect measurements.[7] The disagreement between these two results is not significant because the Mössbauer-effect measurements are not nearly as sensitive to the exact value of $\mu_B H_{\mathrm{exch}}$ as are the spin-wave energies.

The magnitude of the rare-earth–rare-earth exchange interaction is negligible compared to the other interactions. An upper limit of $J_{RR} \simeq 0.01$ meV has been estimated from the observed $\vec{q}$ dependence of the lower optic mode. This interaction is therefore considerably smaller than that found in the pure rare-earth metals or in the Laves phase $TbAl_2$. Of course, in none of these materials do the $4f$ electrons on different ions actually overlap. Apparently the conduction electrons are much more effective in providing an exchange interaction between the rare-earth and iron ions than between the rare earths themselves.

The results we obtained for the crystal field contributions to the spin-wave energies appear to be consistent with other estimates of these interactions. For example, if we express the $B_l{}^0$ constants in terms of the crystal field parameters A_l, e.g., $B_4{}^0 = A_4\langle r^4\rangle\beta$, which we assume to be the same for Tb and Ho, then we find $A_4 = 1.94$ meV$/a_0{}^{-4}$ and $A_6/A_4 = -0.045a_0{}^{-2}$. These compare favorably with the results, 1 meV $\lesssim A_4 a_0{}^4 \lesssim 4$ meV and $A_6/A_4 = -0.04a_0{}^{-2}$, obtained from the Mössbauer-effect experiments.[7]

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