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SPIN DYNAMICS OF THULIUM IONS IN TERBIUM

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Abstract. – The magnetic excitations of $Tb_{90}Tm_{10}$ have been studied by inelastic neutron scattering and calculated with a VCA-RPA model. Due to the difference between the interactions on the Tb and Tm ions, a low-lying excitation on the latter, characterized by strong fluctuations along the hexagonal axis, is observed.

The mutual solubility of metals of the rare earth series allows the fabrication of crystals with disparate and continuously variable magnetic properties. Previous studies of the spin dynamics of such systems, containing small quantities (typically 10 %) of solute rare earths in a host with different magnetic properties, could generally be interpreted in terms of an average-crystal model, with effective exchange and anisotropy parameters.

The case of Tm in Tb is however exceptional, since the Tm ions have a spin $S = 1$, much smaller than that of Tb ($S = 3$), and hence are relatively weakly coupled to their surroundings. Furthermore, due to the shapes of the respective 4f charge clouds, the axial anisotropy of the Tm ions is large and of opposite sign to that of Tb. As a result well-defined quasi-localized states may be excited on the Tm sites within the energy band of the Tb magnons. We have studied the magnetic excitations in a $Tb_{90}Tm_{10}$ single crystal at 4.2 K, by means of inelastic neutron scattering at the DR3 reactor at Risö, and the observed dispersion relations in the c-direction are shown in figure 1.

In order to interpret these rather complex results, we begin with the Hamiltonian

$$H = \sum_i \sum_{lm} B_l^m (i)\, O_l^m (\mathbf{J}_i) - -\frac{1}{2} \sum_{ij} \sum_{\alpha\beta} \mathcal{J}^{\alpha\beta} (ij)\, J_{\alpha i} J_{\beta j}. \quad (1)$$

The crystal-field parameters of Touborg [1], deduced from measurements on dilute alloys, are used for the Tm ions, while the effective single-ion anisotropy on the Tb ions is described by the parameters determined earlier from the study of the magnons in pure Tb [2]. The two-ion interactions between the Tb-ions, also taken from these magnon measurements, are projected on an anisotropic effective exchange coupling. The effective exchange between the different types of ions is then scalled according to

$$\mathcal{J}_{\mathrm{Tm-Tm}} (i,\, j) = \gamma \mathcal{J}_{\mathrm{Tm-Tb}} (i,\, j) = \gamma^2 \mathcal{J}_{\mathrm{Tb-Tb}} (i,\, j)\,. \quad (2)$$

For the conventional RKKY interaction between the spins, γ takes the value $(g-1)_{\mathrm{Tm}} / (g-1)_{\mathrm{Tb}} = \frac{1}{3}$, but this gives rise to a molecular-field excitation of the Tm ions at 4.4 meV, instead of the observed 3.1 meV. To eliminate the discrepancy we have taken γ as 0.24 rather than changing the crystal-field anisotropy of the Tm ions, as the first alternative leads to a more satisfactory fit of both the excitation energies and the scattering intensities. A departure of γ from the simple scaling with the spin is not particularly surprising, since there have previously been observed indications, especially in Tb [2], that the orbital moments are involved in the two-ion coupling in the rare earths, and these are very different for Tb ($L = 3$) and Tm ($L = 5$).

The dynamics of the magnetic system, and the associated neutron scattering cross-section, are determined from the generalized susceptibility tensor $\chi(\mathbf{q},\, \omega)$. In the random-phase approximation, and with the use of the virtual-crystal approximation for the alloy, the neutron scattering cross-section is proportional to

$$\begin{aligned} g^2 \chi (\mathbf{q},\, \omega) &= g_{\mathrm{Tb}}^2 (1-c)\, \chi_{\mathrm{Tb}}^0 (\omega) + g_{\mathrm{Tm}}^2 c \chi_{\mathrm{Tm}}^0 (\omega) \\ &+ \left\{ g_{\mathrm{Tb}} (1-c)\, \chi_{\mathrm{Tb}}^0 (\omega) + g_{\mathrm{Tm}} c \gamma \chi_{\mathrm{Tm}}^0 (\omega) \right\} \\ &\times \left[1 - (1-c)\, \mathcal{J}(\mathbf{q})\, \chi_{\mathrm{Tb}}^0 (\omega) - c\gamma^2 \mathcal{J}(\mathbf{q})\, \chi_{\mathrm{Tm}}^0 (\omega) \right]^{-1} \mathcal{J}(\mathbf{q}) \\ &\times \left\{ g_{\mathrm{Tb}} (1-c)\, \chi_{\mathrm{Tb}}^0 (\omega) + g_{\mathrm{Tm}} c \gamma \chi_{\mathrm{Tm}}^0 (\omega) \right\} \end{aligned} \quad (3)$$

where $\chi^0 (\omega)$ is the frequency-dependent single-ion susceptibility calculated in the molecular-field approximation, $\mathcal{J}(\mathbf{q})$ is the Fourier transform of the two-ion coupling, and $c = 0.1$ is the concentration of Tm ions. As may be seen from figure 1 there is a strong interaction between the magnetic excitations and the transverse phonons, and it is necessary to include this coupling in the calculations. This has been accomplished in accordance with the theory developed earlier for pure Tb [3].

The model accounts well for the experimentally observed excitation energies, as shown in figure 1, apart from a small discrepancy at small q-values in the optical magnon branch. The comparison between the

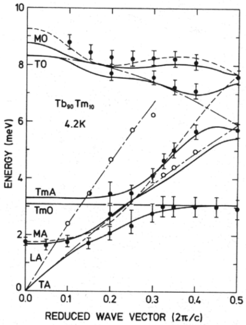

Fig. 1. – Excitations in the c-direction of $Tb_{90}Tm_{10}$ at 4.2 K. The Tb magnon modes, the crystal-field excitations on the Tm-ions, and the transverse phonons which are polarized parallel to the magnetization, mutually interfere to produce the dispersion relations shown by the thick lines, which are the result of the calculation described in the text. The dashed lines show the unpeterturbed Tb magnons and the dashed-dot lines the phonons. A and O signify acoustic and optical respectively. The open circles are experimentally observed pure phonons.

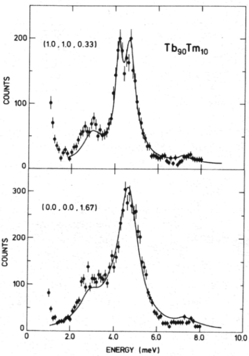

Fig. 2. – Experimental and calculated neutron scattering intensities for the indicated scattering vectors, which correspond to a reduced wave vector of 0.33 in figure 1. In the lower curve the scattering vector is in the c-direction, while it is close to the hexagonal plane in the upper. Adjustable energy resolutions have been included in the calculations. In the upper curve an unperturbed transverse phonon is observed, and the ratio of the impurity intensity of the magnon peak is roughly twice that in the lower curve.

measured and calculated intensities is illustrated in figure 2. The agreement is again generally satisfactory, although the intensity of the impurity mode at about 3 meV is somewhat underestimated. However, the results of other types of scan indicate that this discrepancy may be due to an energy-dependence of the instrumental resolution, which is not included in the calculations.

When the scattering vector is along the hexagonal c-axis the intensity is proportional to

$$I_{\parallel} = \mathrm{Im}\ \chi^{aa}(\mathbf{q},\omega) \tag{4}$$

whereas, because of the random domain structure, it is proportional to

$$I_{\perp} = \mathrm{Im}\ \chi^{cc}(\mathbf{q},\omega) + \frac{1}{2}\mathrm{Im}\ \chi^{aa}(\mathbf{q},\omega) \tag{5}$$

when the scattering vector is in the hexagonal plane. For studying the impurity modes it would be advantageous to produce a single-domain crystal, by the application of a magnetic field perpendicular to the scattering vector in the plane, which would remove the term due to Im $\chi^{aa}(\mathbf{q}, \omega)$ in (5). This would lead to a more clear distinction between the scattering intensities related to the two different components of the susceptibility tensor.

The difference between the interactions of the Tb and Tm ions in this alloy has a profound influence on the magnetic behaviour at the two types of site. The exchange forces the Tm moment to be in the plane at low concentrations but, according to our calculation, it is reduced from the saturation value of 7 μ_B to about 5.9 μ_B, whereas the Tb moment is very close to saturation. Furthermore the first excited state on the Tm ions is at a relatively low energy and the associated magnetic fluctuations are predominantly in the c-direction, reflecting an incipient realignment which actually occurs at higher concentrations [4]. The Tb fluctuations on the other hand are largely confined to the plane, with the result that the neutron scattering intensity stemming from the c-axis fluctuations is comparable for the two types of sites, even though only 10 % of the ions are Tm. The dynamics of the Tm moments could be investigated in greater detail by carrying out inelastic neutron scattering experiments in a magnetic field.

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