Journal of Magnetism and Magnetic Materials 76 & 77 (1988) 399–400
North-Holland, Amsterdam

IMPURITY MODE DYNAMICS OF DILUTE ERBIUM IN PRASEODYMIUM

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Inelastic neutron scattering measurements on a single crystal of $Pr_{0.95}Er_{0.05}$ show the presence of an impurity exciton mode, due to a crystal field excitation of the Er ion, which strongly perturbs the host excitons. A theoretical description, using the virtual crystal approximation is in good qualitative agreement with the data.

Praseodymium crystallises in the dhcp structure in which half the sites have local cubic symmetry and half hexagonal symmetry. Its magnetic properties show that both sites have singlet ground states in the crystal field, thereby tending to suppress magnetic order. The elementary excitations are magnetic excitons, whose dispersion has been extensively studied, yielding a wealth of detailed information [1]. In particular the excitations on the hexagonal sites ($|m_J = 0\rangle$ to $|m_J = \pm 1\rangle$) show evidence of a large degree of two-ion anisotropy which removes the two-fold degeneracy. Their large dispersion shows that the exchange is almost large enough to induce magnetic order. Indeed magnetic order is found at very low temperatures as a consequence of polarisation of the electronic system by the hyperfine interaction with the nuclei [2].

We have studied the magnetic excitations in a large single crystal of $Pr_{0.95}Er_{0.05}$ by inelastic neutron scattering, using the IN3 and IN12 triple axis spectrometers at ILL Grenoble. Elastic scans along the (η03) direction at 5 K revealed a broad diffuse peak, centred on $\eta = 0.15$, similar to that found in pure Pr [2]. At 1.3 K the same scan showed a magnetic Bragg peak centred at $\eta = 0.2$, arising from magnetic ordering on the hexagonal sites. The temperature variation of the Bragg peak intensity indicated a Néel temperature T_N of 2.4 K. We note that the modulation wavevector for the magnetic order is rather larger than that found in pure Pr where the magnetic Bragg peaks occur at $\eta = 0.14$. Since Er is a Kramers ion, it will have a doublet ground state in the crystal field. It is clear that the magnetic order is a consequence of the polarisation of the host electronic system by the magnetic moment of the impurity. The magnetic excitations, described below, give a detailed insight into this process.

We have concentrated attention on the excitations propagating on the hexagonal sites in the ΓA and ΓM directions at 4 K, just above T_N. These sites form an hcp sublattice, so there are two sets of modes corresponding to acoustic and optic excitations. The acoustic and optic branches were measured around the (004) and (003) reciprocal lattice points respectively. The measured neutron groups showed two, three or sometimes four resolved or partly resolved peaks at each wavevector. The peak positions were determined by fitting the appropriate number of overlapping Gaussians to the measured lineshape. The measured dispersion relations are shown in fig. 1. Comparison with the excitations of pure Pr show profound differences. In particular there is clear evidence of an impurity mode close to 2.4 meV which strongly perturbs the host excitons, especially where it crosses and mixes with the optic branches. We attribute this mode to a crystal field excitation from the ground state doublet of the Er impurity to an excited doublet. A second remarkable feature is the perturbation of the lowest optical branch in the ΓM direction. In pure Pr this branch, which displays some soft-mode features, has a minimum energy of ≈ 1.0 meV at a wavevector $\eta = 0.14$. Here there is a much deeper minimum ≈ 0.55 meV, at a larger wavevector $\eta = 0.20$. This is clear evidence that the impurity modifies the nature of the two-ion anisotropic coupling in the host. This result also accounts for the difference in the wavevector of the magnetic order in PrEr compared to pure Pr.

A preliminary calculation of the coupled host-impurity mode energies has been made using the

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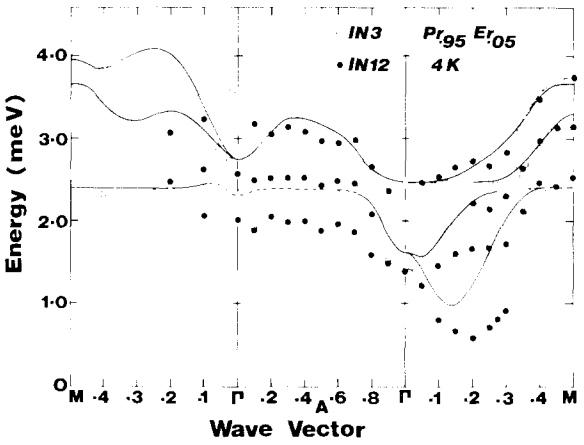

Fig. 1. Measured dispersion relations for $Pr_{0.95}Er_{0.05}$ single crystal. Open circles: IN3 data; filled circles: IN12 data. Solid lines are the predictions of the VCA calculation assuming host exchange interactions are unaltered by alloying. Dashed lines are guides to the eye indicating the trend of the optic mode energies.

virtual crystal approximation (VCA) [3]. For simplicity we have assumed that the exchange interactions of the host are not affected by the addition of Er. In this case the interactions between two Er ions and between an Er and a Pr ion are scaled versions of those between two Pr ions:

$$J_{\mathrm{ErEr}}(ij) = \gamma J_{\mathrm{PrEr}}(ij) = \gamma^2 J_{\mathrm{PrPr}}(ij) \qquad (1)$$

where $\gamma = (g-1)_{\mathrm{Er}}/(g-1)_{\mathrm{Pr}} = -1$. In the VCA the dynamical susceptibility of the alloy becomes (in units of $g^2\mu_B^2$):

$$g^2\chi(q,\omega) = (1-c)g_{\mathrm{Pr}}^2\chi_{\mathrm{Pr}}^0(\omega) + cg_{\mathrm{Er}}^2\chi_{\mathrm{Er}}^0(\omega) + \chi_1(\omega)T_c(q,\omega)\chi_1(\omega)$$

where c is the concentration of Er ions ($c = 0.05$), and

$$\chi_1(\omega) = (1-c)g_{\mathrm{Pr}}\chi_{\mathrm{Pr}}^0(\omega) + c\gamma g_{\mathrm{Er}}\chi_{\mathrm{Er}}^0(\omega).$$

In the simplest case the T-matrix of the pure material is given by $T(q,\omega) = J(q)/\{1 - \chi_{\mathrm{Pr}}^0(\omega)J(q))\}$; however for the case of Pr it is somewhat more complicated because of the two-ion anisotropy and the presence of two ions per unit cell. For the alloy the T-matrix $T_c(q,\omega)$ is derived from that for the pure crystal by replacing $\chi_{\mathrm{Pr}}^0(\omega)$ by

$$\chi^0(\omega) = (1-c)\chi_{\mathrm{Pr}}^0(\omega) + c\gamma^2\chi_{\mathrm{Er}}^0(\omega).$$

The only unknown quantity is $\chi_{\mathrm{Er}}^0(\omega)$. This is determined by the crystal field parameters for Er on the hexagonal sites of Pr. We have estimated these by scaling the B_{20} parameter of Pr by the Stevens α factor and taking B_{40}, B_{60} and B_{66} to be equal to those for Er in Y [4]. These parameters were then scaled by a factor 0.9 in order to give an excited doublet at 2.40 meV above the ground state. The next excited doublet lies at 4.35 meV.

The results of the calculation are shown as the solid lines in fig. 1. Overall these display the same qualitative features as the experimental dispersion relations. The model provides an excellent starting point for a more detailed analysis of the data, including the spectral weights derived from the neutron groups. The large discrepancies in the lower optical branch energies along ΓM show the remarkably large influence of only 5% of an impurity on the two-ion couplings of the host. The measured mode energy at the Γ point is lower than calculated. This suggests a shift in the Pr $|\pm 1\rangle$ crystal field level of 0.15 meV, perhaps through a modification of the c/a ratio. A further discrepancy is the occurence of an additional branch at 2.0 meV in the ΓA direction and a hint of another branch at 3.6 meV near the Γ point of the acoustic branches. These might arise from crystal field excitations of Er ions on the cubic sites. Further work is required to elucidate these features. Since the Er ions have doublet ground states, there will be a small quasielastic contribution to the basal plane component of the dynamical susceptibility. The transition to the ordered phase will be driven by a divergence in this response. This may have dynamical consequences far from zero frequency: the inclusion of a finite energy width into the quasielastic response produces a small decrease in the lowest optical mode energies.

References

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