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Magnetic excitations in praseodymium alloys

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Abstract

We have studied the effect of alloying on the magnetic excitations in Pr with the PRISMA spectrometer at ISIS. The dispersion of the excitations in single crystals of $Pr_{0.9}Tm_{0.1}$ and $Pr_{0.9}Ce_{0.1}$ along the a^*- and c^*-directions has been compared with that in pure Pr. We find that alloying Pr with Tm changes the excitation spectrum very significantly, due to strong interactions with the Tm impurity mode, whereas alloying with Ce produces more subtle changes.

1. Introduction

The magnetic excitations in the rare-earth metal praseodymium have been extensively studied, since it is a paradigm of a singlet-ground-state crystal-field system in which the exchange interaction is close to the critical value required to induce magnetic ordering [1]. The excitations may be described as magnetic excitons propagating on the hexagonal and cubic sites of the DHCP crystal structure, and in general there are four branches on the hexagonal and two on the cubic sites, respectively [1, 2].

It is interesting to investigate how alloying small concentrations of other rare-earth elements changes the magnetic properties of Pr. We have chosen to examine the magnetic excitations in Pr doped with 10% Ce and 10% Tm. Ce is of course the neighbouring ion to Pr with $J = 5/2$ and therefore with a Kramers doublet ground state. Thulium, the hole analogue of Pr has $J = 6$ and may have a singlet ground state. The excitations in pure Tm have been studied by us previously [3].

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2. Experimental details and results

Single crystals of Pr, $Pr_{0.9}Ce_{0.1}$ and $Pr_{0.9}Tm_{0.1}$ were grown by horizontal zone-melting techniques. Each sample, typically of mass 7 g, was mounted with a real space a-direction perpendicular to the [1, 0, 0]–[0, 0, 1] scattering plane in an "orange" cryostat. The temperature for all measurements was 1.8 K.

The experiments were performed using the PRISMA multi-analyser spectrometer at the ISIS Facility. Using the time-of-flight technique, this instrument covers a large area in (Q, ω) space in one experimental set-up and is therefore ideally suited for overview measurements. A detailed description of the PRISMA spectrometer and the measuring technique is given in Ref. [4]. We used the instrument with a set of 14 pyrolytic graphite analysers and 40′ collimation before and after the analysers.

In its present configuration the PRISMA spectrometer is subject to geometrical constraints which restrict the range of available analyser energies. We therefore chose to measure the excitation spectra (i) parallel to the a^*-axis through the (1, 0, 5) zone centre and (ii) parallel to the c^*-axis covering several Brillouin zones along the

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L–M zone boundaries from (1.5, 0, 3) to (1. 5, 0, 6). In this configuration the analyser energies varied from 17 meV (analyser 1) to 32 meV (analyser 14): the corresponding instrumental resolution varied from 1.2 to 2.74 meV.

Fig. 1 shows typical spectra (from detector 6) for the three samples, along the a^*-direction. The data have been fitted to a series of Gaussian peaks. The wave vector for the measurements at 1–4 meV in these spectra varies between (0.8, 0, 5) and (0.9, 0, 5), where the inelastic structure factor for the acoustic modes dominates over the optic modes. Nevertheless, the longitudinal optic mode at 1.2 meV can just be resolved, but the other three modes on the hexagonal sites in Pr cannot be separated properly. The cubic site modes in Pr are, as expected, almost degenerate at 7.8 meV. In $Pr_{0.9}Ce_{0.1}$ the centre of gravity of the scattering on the hexagonal sites has shifted downwards by about 0.6 meV: a smaller shift is also evident for the cubic site peak. In $Pr_{0.9}Tm_{0.1}$ we find that the scattering has shifted downwards noticeably and is now centred at 2.3 meV. We observe a new mode at 6.0 meV which can be resolved from the 8 meV cubic-site excitations.

Results for the c^*-direction exhibit similar trends. In pure Pr the hexagonal site modes now have energies from 3.2 to 4.0 meV [1, 2]. The cubic site excitations at 8 meV are clearly seen. The spectrum for $Pr_{0.9}Ce_{0.1}$ is very similar with only slight changes in the relative intensities. In contrast, in $Pr_{0.9}Tm_{0.1}$ the 3–4 meV modes are less clearly resolved from the scattering centred at zero energy, and there is clearly an additional peak at 6.4 meV, comparable in intensity with the 8 meV peak.

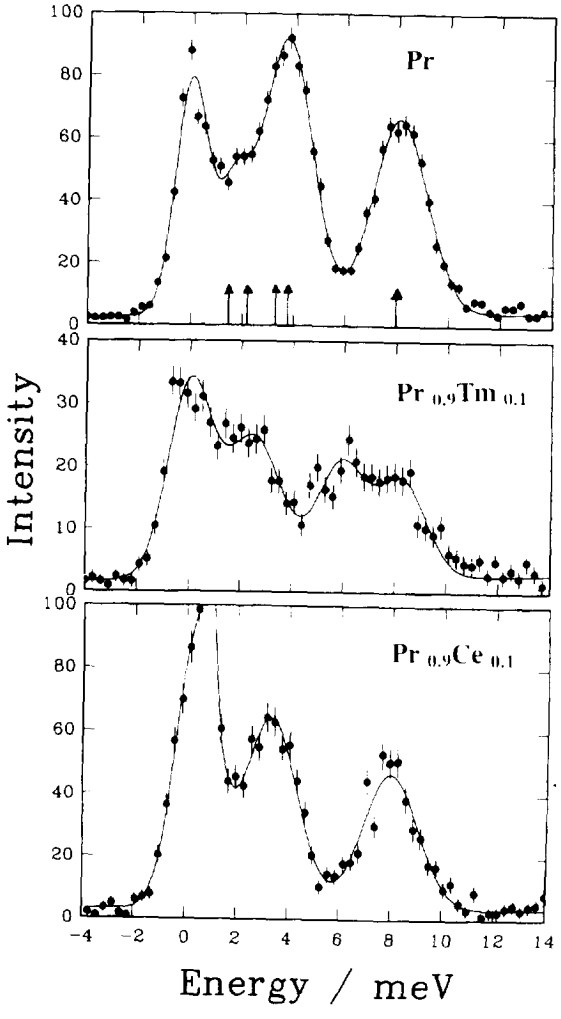

Fig. 1. Inelastic scattering spectra along the a^*-direction for Pr, $Pr_{0.9}Tm_{0.1}$ and $Pr_{0.9}Ce_{0.1}$ from detector 6.

3. Discussion

We interpret our experimental results in terms of the VCA (virtual crystal approximation) method [5] applied earlier to the case of $Pr_{0.95}Er_{0.05}$ [6]. The exchange interactions between Pr and the impurities (R = Tm or Ce) are scaled from the Pr–Pr interactions according to

$$J_{RR}(ij) = \gamma J_{RPr}(ij) = \gamma^2 J_{PrPr}(ij),$$

where $\gamma = (g-1)_R/(g-1)_{Pr}$. The dynamical susceptibility of the alloy may then be constructed from the dynamical susceptibility of the components. Since the γ-factors for Tm and Ce are numerically less than 1 ($-\frac{5}{6}$ and $\frac{5}{7}$, respectively), in both cases there is a dilution effect on the excitation spectrum so that the dispersion becomes less pronounced than in pure Pr.

We have calculated the response in the Γ–M direction (q along real space b-axis), considering only the excitations propagating on the hexagonal sites. For the Tm alloy, the Tm crystal-field parameters were taken from Ref. [3] which yield a dipolar excited state at about 8 meV (see Fig. 3 in Ref. [3]). This mode is sufficiently well separated from the Pr hexagonal modes that there is negligible coupling between them. Hence, for the lower energy part of the spectrum only the dilution effect is important. Fig. 2 shows the calculated dispersion along the Γ–M direction for $Pr_{0.9}Tm_{0.1}$. The most pronounced effect is the increased energy of the optic modes around the minimum in the dispersion curve.

In the case of $Pr_{0.9}Ce_{0.1}$, the dilution effect is slightly greater since $\gamma = \frac{5}{7}$ here. We have deduced the Ce hexagonal site crystal field levels from a simple Stevens factor scaling of the crystal-field parameters of Pr, which yields $B_{20} = 0.5$ meV and $B_{40} = 0.0025$ meV, leading to excited doublets at about 2.25 and 8.85 meV. The low-lying doublet hybridizes strongly with the Pr hexagonal site

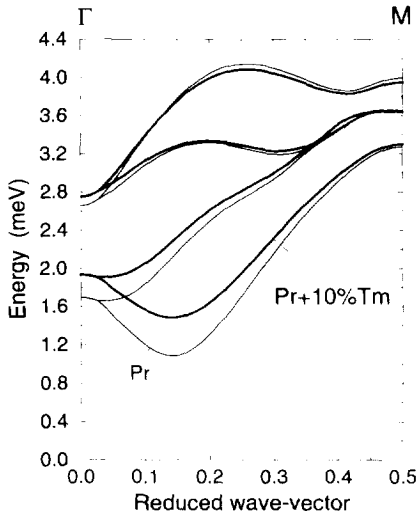

Fig. 2. The dispersion of the hexagonal site excitations in $Pr_{0.9}Tm_{0.1}$ (bold lines) calculated by the VCA method. The dispersion in pure Pr is also shown.

excitations, but the lowest Ce mode (unresolved doublet) is still at higher energies than that of the pure Pr-mode.

The experimental results seem to indicate the opposite behaviour, with the excitations much more affected by the Tm than by the Ce-ions. In $Pr_{0.9}Tm_{0.1}$, we find a strong modification of the cubic site excitations around 8 meV. This is unlikely to be caused by the Tm ions on the hexagonal sites. The effect is so big that it is probably associated with a Tm level on the cubic, rather than the hexagonal, sites at round 6 meV (most clearly seen in the c-axis scan). In $Pr_{0.9}Ce_{0.1}$ it is possible that a Ce-mode around 2–3 meV is present but has not been resolved in this experiment. Measurements with higher resolution are necessary to elucidate this question. The PRISMA-II spectrometer, currently under construction, will operate with significantly lower analyser energies and will allow survey measurements with a correspondingly improved resolution to be made [7].

References

[1] J. Jensen and A.R. Mackintosh, Rare Earth Magnetism: Structures and Excitations (Oxford Univ. Press, Oxford, 1991).

[2] J.G. Houmann, B.D. Rainford, J. Jensen and A.R. Mackintosh, Phys. Rev. B 20 (1979) 1105.

[3] K.A. McEwen, U. Steigenberger and J. Jensen, Phys. Rev. B 43/4 (1991) 3298.

[4] U. Steigenberger, M. Hagen, R. Caciuffo, C. Petrillo, F. Cilloco and F. Sacchetti, Nucl. Inst. Methods B 53 (1991) 87.

[5] J. Jensen, J. Magn. Magn. Mater. 14 (1979) 224.

[6] B.D. Rainford, L. Cussen, J. Jensen and D. Fort, J. Magn. Magn. Mater 76&77 (1988) 399.

[7] C. Petrillo, F. Sacchetti, M. Hagen and U. Steigenberger, RAL Report RAL-93-091.