Journal of Magnetism and Magnetic Materials 14 (1979) 224–226

CPA – CALCULATION OF THE PARAMAGNETIC EXCITATIONS IN Pr + 5%Nd

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The paramagnetic excitation spectrum of a substitutional disordered alloy consisting of two different types of magnetic ions is derived in the coherent potential approximation (CPA). The results are applied to the case of dhcp Pr + 5%Nd, which has been studied experimentally by Houmann et al. using inelastic neutron scattering.

1. Theory

We consider a binary system of magnetic ions, 1 and 2, which are characterized by their non-interacting susceptibilities, $\chi_l(\omega) = (g_l\mu_B)^2 u_l(\omega)$; $l = 1$ or 2. We assume the susceptibilities and the two-ion couplings to be diagonal in the Cartesian components, so that each component, α, may be considered independently. The ions are arranged randomly in a fixed lattice, and the two-ion interaction is

$$-\tfrac{1}{2}\sum_{\alpha}\sum_{i\neq j}\mathcal{J}^{(\alpha)}(ij)[(1-p_i)J_1^{\alpha,i}+\lambda p_i J_2^{\alpha,i}] \times [(1-p_j)J_1^{\alpha,j}+\lambda p_j J_2^{\alpha,j}]\,. \quad (1)$$

$p_i = 1$ if the ith ion is of type 2, otherwise $p_i = 0$. $J_1^{\alpha,i}$ denotes the αth component of the total angular momentum of a type 1 ion at site i. The couplings $J_1^\alpha - J_2^\alpha$ and $J_2^\alpha - J_2^\alpha$ are considered to be λ and λ^2 times the corresponding $J_1^\alpha - J_1^\alpha$ coupling, respectively. In the case of a binary rare-earth system $\lambda = (g_2-1)/(g_1-1)$. A 2 × 2 matrix of Green's functions may then be defined, for example:

$$G_{12}(ij,t) = -\mathrm{i}\theta(t)\langle[(1-p_i)J_1^{\alpha,i}(t), p_j J_2^{\alpha,j}(0)]\rangle\,. \quad (2)$$

The neutron scattering cross-section is proportional to $\mathrm{Im}\,\chi(\boldsymbol{q},\omega)/[1-\exp(-\hbar\omega/k_BT)]$, where $\chi(\boldsymbol{q},\omega)$ is the Fourier transform of the configurational average of $-2\pi\mu_B^2\sum_{l,l'} g_l g_{l'} G_{ll'}(ij,t)$. In the paramagnetic phase $\chi(\boldsymbol{q},\omega)$ may be expressed in terms of the two Green's functions: $G_l(ij,\omega) = G_{1l}(ij,\omega) + \lambda G_{2l}(ij,\omega)$, using an RPA-decoupling of the equations of motion (utilizing the standard basis excitation operators) where

$$G_l(ij,\omega) - [u_1(\omega) - p_i\{u_1(\omega)-\lambda^2u_2(\omega)\}] \times \sum_{i'}\mathcal{J}(ii')G_l(i'j,\omega) = -\frac{1}{2\pi}\delta_{i,j}[\delta_{l,1}(1-p_i)u_1(\omega)+\delta_{l,2}\lambda p_i u_2(\omega)]\,. \quad (3)$$

This equation is equivalent to that considered for a dilute Van Vleck paramagnet ($u_2(\omega)\equiv 0$) by Schmidt [1] and in more general cases by Lage and Stinchcombe [2]. Using the CPA-result obtained by these authors, we get (p is the average value of p_i):

$$\chi(\boldsymbol{q},\omega) = \mu_B^2\Gamma(\boldsymbol{q},\omega)\Big[(1-f_\omega)g_1^2u_1(\omega)+f_\omega g_2^2u_2(\omega) + \{p-f_\omega-\mathcal{J}(\boldsymbol{q})[p(1-f_\omega)u_1(\omega) - f_\omega(1-p)\lambda^2u_2(\omega)]\} \times \frac{(\lambda g_1-g_2)^2u_1(\omega)u_2(\omega)}{\{u_1(\omega)-\lambda^2u_2(\omega)\}}\Big]\,, \quad (4)$$

where

$$\Gamma(\boldsymbol{q},\omega) = [1-(1-f_\omega)u_1(\omega)\mathcal{J}(\boldsymbol{q}) - f_\omega u_2(\omega)\lambda^2\mathcal{J}(\boldsymbol{q})]^{-1} \quad (5)$$

and

$$f_\omega = p\left[\frac{1}{N}\sum_q \{1 - u_2(\omega)\lambda^2 \mathcal{J}(q)\}\Gamma(q,\omega)\right]^{-1} . \quad (6)$$

The virtual-crystal result (VCA) is given by eqs. (4) and (5) if f_ω is replaced by p.

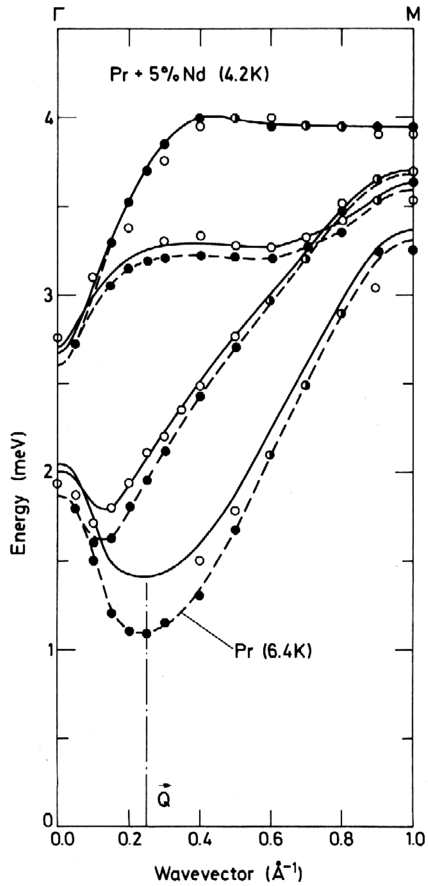

Fig. 1. The energies of magnetic excitations propagating on the hexagonal sites in the longitudinally polarized antiferromagnetic phase of Pr + 5%Nd in a [100]-direction. The experimental results (○) were obtained by Houmann et al. (private communication). The solid lines are the VCA-results deduced from the Pr-results at 6.4 K (–•–•–) after ref. [3]. At 4.2 K the amplitude of the modulated structure is ~0.8 μ_B/ion [4], corresponding to an effective ferromagnetic moment of a factor $\sqrt{2}$ smaller (or an applied field of about 10 kOe [3]). In the calculation we neglected $\chi_{Nd}(\omega)$, which should be justified except near the Nd level around 1.5 meV. Further, we neglected the off-diagonal coupling between excitons at $\boldsymbol{q}$ and $\boldsymbol{q} \pm 2\boldsymbol{Q}$, which gives rise to energy gaps of ~0.1 meV or less. No well-defined low-energy excitations were observed close to $\boldsymbol{Q}$.

2. Excitations in Pr + 5%Nd

The magnetic excitation spectrum of dhcp Pr + 5%Nd has been studied experimentally by Houmann and coworkers (private communication). The crystal was found to order at $T_N \cong 7.5$ K. The modulation vector, $\boldsymbol{Q}$, of the antiferromagnetic structure is the wavevector at which the dispersion relation in Pr has a minimum eneegy in the ΓM direction (see fig. 1).

The experimental results in the paramagnetic phase of Pr + 5%Nd (at 8.5 and 11.5 K) for the excitons propagating on the hexagonal sites show a strong hybridization of the Pr-modes with an excited state of the Nd-ions around 1.5 meV. A representative example is given in fig. 2.

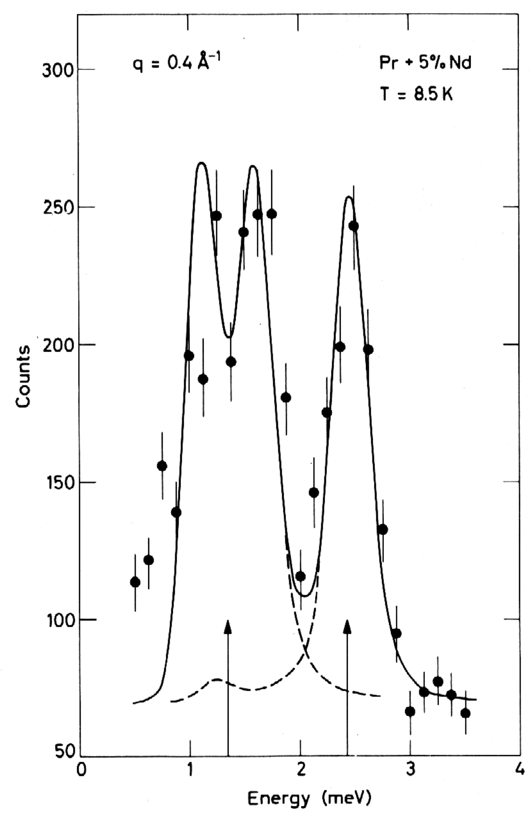

Fig. 2. Neutron group observed at $q = 0.4$ Å^{-1} along ΓM by Houmann et al. (private communication) in the paramagnetic phase of Pr + 5%Nd. The arrows indicate the mode-positions in Pr. The solid lines give the calculated result using an experimental resolution function with a full width at half maximum of 0.30 meV.

In the calculations we incorporated the intrinsic linewidth of the pure Pr-modes due to single-site fluctuations as derived by Bak [5]. We avoided the unphysical feature of the CPA-result (at low frequencies), which is discussed by Lage and Stinchcombe [2], by replacing f_ω by p in this limit (for energies somewhat below 0.5 meV). The summation over q-space, eqs. (5) and (6), was transformed into a self-consistent integral-equation in ω-space, involving the density of states of the exciton modes in Pr calculated by Bak [5]. When the crystal-field levels of the Nd-ions are appropriately adjusted, the theory gives a reasonable account of the neutron scattering results at 8.5 and 11.5 K, except for $\boldsymbol{q} \approx \boldsymbol{Q}$, where the observed low-energy neutron group is much broader than that calculated. The discrepancy may be ascribed to critical fluctuations. The crystal-field parameters used for the Nd-ions are $B_{20} = 0.06$ meV, $B_{40} = (-1.5 \times 10^{-4})$ meV, and $B_{60} = -\frac{8}{77} B_{66} = (-2.5 \times 10^{-5})$ meV (defined in [3]), producing dipolar-excited levels at 0.27 and 1.45 meV. We used $\lambda = 1.135$ (instead of 15/11) in order to obtain $T_N = 7.5$ K (within VCA). The observation of the Nd-level at 0.27 meV would require a high experimental resolution if it should be distinguished from the elastic peak due to the degenerate states of the Nd-ions. In the model the cubic ions are only included through the use of an effective coupling within the hcp-sublattice (see [3]), and corrections are expected. However, the hybridization of the optical modes is certainly due to the Nd-ions on the hexagonal sites, as it is present also near Γ.

In order to obtain better resolved spectra close to $\boldsymbol{Q}$ and T_N, experiments with a Pr + 2.5%Nd crystal are planned for the future.

References

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