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Line Shape of Magnetic Excitations in
Singlet-Ground-State Systems*

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The excitation spectrum in a paramagnetic singlet doublet system is calculated using a diagrammatic expansion technique, and the theoretical predictions are compared with experiments on praseodymium. The theory gives an accurate description of the dramatic temperature dependence of the energies and lineshapes for the exciton modes.

For the last several years there has been a great interest in the properties of localized magnetic systems which possess a nonmagnetic singlet ground state. The ordering in such systems occurs as an exchange polarization of the ground state, provided the exchange interaction between the magnetic ions exceeds a certain threshold value. Here we shall present a calculation of the paramagnetic excitation spectrum in a singlet doublet system using a diagrammatic Green's function expansion method. A similar technique has been applied to a ferromagnet by Vaks, Larkin and Pikin [1], and to the Ising model in a transverse field by

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Stinchcombe [2]. In contrast to other techniques, based on various decouplings of equations of motion for spin operators, this kind of theory gives a well defined high-density expansion parameter $(1/Z)$ where Z is an effective number of interacting neighbors. The approximations are based on physical, not technical reasons. Praseodymium is an example of a singlet ground state magnet in which the exchange is only slightly undercritical with respect to magnetic ordering. The ground state on the hexagonal sites is the pure $|J^Z=0\rangle$ singlet, and the first excited state is the doublet $|x\rangle, |y\rangle$, where $|x\rangle = 1/\sqrt{2} (|1\rangle + |-1\rangle)$ and $|y\rangle = -i/\sqrt{2} (|1\rangle - |-1\rangle)$. The paramagnetic excitation spectrum has recently been measured by Houmann *et al.* [3] using inelastic neutron scattering technique. The lowest lying mode shows a clear tendency towards softening as the temperature is lowered towards 0K, and a dramatic increase of the intrinsic linewidth takes place as the temperature increases from 6K to 30K, where well-defined modes cease to exist. In Praseodymium seems to be the simplest real singlet-ground-state magnet, and significantly more information is now available on the excitations in this material than in any other paramagnetic system. This makes the element Pr almost ideal for a confrontation between experiment and theoretical model calculations.

The effective Hamiltonian describing the magnetic ions on the hexagonal sites in Pr may be written

$$\mathcal{H} = \sum_i \Delta (S_i^z)^2 - \sum_{ij, \alpha\beta} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta, \quad \alpha, \beta = x, y$$

Δ is the crystal field splitting (≈ 3.2 MeV). We introduce the Green's functions

$$G^{\alpha\beta}(r_1, \tau_1; r_2, \tau_2) = \langle T_\tau S^\alpha(r_1, \tau_1) S^\beta(r_2, \tau_2) \rangle ,$$

where $\langle T_\tau \dots \rangle$ denotes the thermal average of the τ ordered product of operators in the interaction representation. The Green's function can be represented by a sum of connected diagrams consisting of single-cell blocks joined by interaction lines, $J(\vec{q})$. For a detailed description of the technique, see Ref. [4]. The zeroth order diagrams (RPA) simply consist of noninteracting Green's function connected with interaction lines. The poles of the RPA-propagators, which give the energy spectrum are

$$(\omega_{\vec{q}}^N)^2 = \Delta^2 - 4\Delta R \int^N(\vec{q})$$

where $R = \{1 - \exp(-\beta\Delta)\} \{1 + 2\exp(-\beta\Delta)\}^{-1}$ and $\int^N(\vec{q})$ are eigenvalues of a 4×4 matrix describing the exchange interactions between the ions. The measured dispersion relations were fitted to this expression and inter-atomic exchange parameters were derived. Figure 1 shows the polarization of two of the four modes as a function of q in the Γ -K-M direction. The polarizations change rapidly as a function of q : at the zone center the polarization is linear in the x or y direction, but in general the modes are elliptically polarized in the xy plane. The eigenvectors of the modes can be measured by neutron scattering technique.

The first order diagrams give rise to small shifts of the excitation energies and, more interesting, by extending the theory one order beyond the RPA we obtain the leading contributions to the lifetimes of the excitations. It is interesting that the damping occurs to first order in the expansion. For boson or fermion systems, the lowest order imaginary part of the self energy occurs in second order in the high

density expansion due to interaction between excitations. The spectral functions, which are proportional to the neutron scattering cross-section, can easily be expressed in terms of the Green's functions using the fluctuation-dissipation theorem. The theoretical lineshapes convoluted with the experimental resolution function are compared with experiment in Fig. 2. Almost complete agreement between positions of peaks, intensities and lineshapes is observed at any temperature. The agreement is least perfect at the highest temperatures, where the linewidth, as calculated to first order in the $(\frac{1}{2})$ expansion, is comparable to the energy as calculated to zeroth order. In addition to the diagrams considered here, there are also diagrams which, to first order in $\frac{1}{2}$, describes quasielastic contributions to $S(\vec{q}, \omega)$. Recently, a weak central peak has been observed in Pr at low temperatures [6]. It would be interesting to calculate this effect within the present formalism and compare with experiment.

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FIGURE CAPTIONS

- Fig. 1 Polarization of exciton modes in praseodymium as a function of wavevector, q , in the ΓM direction.
- Fig. 2 Spectral functions for magnetic excitations in Pr. Points: Neutron measurements (Ref. [5]); lines: theoretical curves folded with experimental resolution.

FIG. 1

FIG. 2