TWO-MAGNON RAMAN SCATTERING IN QUADRATIC DOUBLE-LAYER ANTIFERROMAGNETS

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Two-magnon Raman spectra of ordered $K_3Mn_2F_7$ and $K_3Ni_2F_7$ are interpreted in terms of a two-dimensional four-sublattice spin-wave theory. In solving the equation of motion of the Green functions it is essential to include terms of the spin-wave Hamiltonian coupling the two magnon branches.

The magnetic double-layer structure $K_3Mn_2F_7$ ($T_N = 58.3$ K) may be considered as intermediate between the two-dimensional (2D) antiferromagnet K_2MnF_4 and the 3D $KMnF_3$. Its magnetic thermodynamic properties have only recently been considered in some detail [1, 2]. Here, we explore the spin-wave dispersion by observing the Raman spectra and comparing them with the results computed by Green-function methods.

The spin Hamiltonian is given by

$$\mathcal{H} = |J| \sum_{j, d} S_{j, a} \cdot S_{j+d, b}$$
$$- g\mu_B H_A \left(\sum_j S^z_{j, a} - \sum_j S^z_{j, b} \right), \qquad (1)$$

where J is the exchange constant, S_a and S_b are up and down spins, respectively, and H_A represents the temperature-dependent staggered anisotropy field parallel to the tetragonal axis. The summation over j runs over both layers; d is associated with summation over the five nearest neighbors of site j, one of which is on the adjoining layer. A 2D four-sublattice (two on each layer) spin-wave treatment based on eq. (1) leads to a magnon dispersion consisting of two-fold degenerate magnon branches [1]. The treatment has been particularly successful in describing the thermodynamic properties well below ordering, and we have therefore used it here to calculate the Raman-scattering cross section. The calculational technique is the equation of motion of the retarded Green functions $\langle\langle \mathcal{H}_R; \mathcal{H}_R \rangle\rangle$ [3], where the Raman scattering tensor $\mathcal{H}_R$ complies with the symmetry of the five nearest-neighbor pairs of site j and the symmetry of the lattice. A point of special note is that there is an essential contribution to the scattering from four-magnon terms in the Hamiltonian of the form $\alpha^\dagger_k \beta^\dagger_k \delta_k \epsilon_k$, i.e. involving the creation of two magnons of the lower branch (in-phase precession of the paired layers)

and the destruction of two magnons of the upper branch (out-of-phase precession). These terms have no effect in calculating thermodynamic quantities since in the random-phase approximation they do not renormalize the magnon energies.

In fig. 1 the experimental Raman spectrum at 2 K is given together with the theoretical zero-temperature spectrum calculated with decoupling of the equation of motion to first-order, i.e. without magnon damping. Adopting $\Delta = g\mu_B H_A/5|J|S = 0.002$, we have adjusted the exchange parameter J
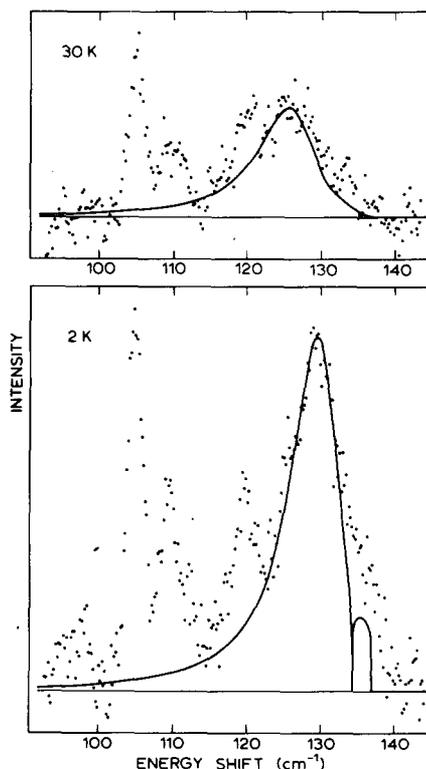

Fig. 1. Two-magnon Raman spectra in $K_3Mn_2F_7$ in the $z(xx)y$ polarization configuration. Solid curves are theoretical spectra.

so as to make the theoretical peak position coincident with experiment. The resulting $J = (-7.68 \pm 0.07)$ K is in excellent agreement with $J = (-7.59 \pm 0.03)$ K from analysis of the sublattice magnetization [1], as well as $J = (-7.62 \pm 0.15)$ K and $J = (-7.7 \pm 0.2)$ K from single-crystal data of the perpendicular and parallel susceptibility, respectively [4]. Furthermore, when comparing the shape of the calculated spectrum with experiment we see good accord, taking into account the finite resolution of the spectrometer (~ 3 cm^{-1}).

In second order, the equation of motion technique provides a damping of the magnons. The damping of magnons primarily contributing to the Raman intensity, i.e. with k near the zone boundary and, for the upper branch, near maximum dispersion, has been expressed in an approximate analytical form. In fig. 1 the resulting theoretical spectrum at 30 K, as an example, is compared with experiment. Again there is good agreement. Spectra at other temperatures are not shown here, but we note that the computed second-order spectra account for the observed temperature dependence of both the peak position and the width of the scattering up to 1.1 T_N.

We have finally compared the experimental low-temperature spectrum in the isomorphic $K_3Ni_2F_7$, reported earlier by Ferguson et al. [5], with our calculations. In this compound the anisotropy Δ is also small, but not yet known precisely. The comparison therefore does not yield J itself, but the combination $J(1 + 1.04\ \Delta) = (-95.4 \pm 0.5)$ K. As in the case of $K_3Mn_2F_7$ the theoretical result nicely fits the experimental spectrum.

References

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