MAGNON-PAIR MODES IN TWO DIMENSIONS

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Magnon-pair modes have been studied in the quadratic layer antiferromagnet K_2NiF_4 at temperatures between 5 and 150°K ($T_N = 97.1$°K) using the technique of second-order Raman scattering. At low temperatures the effect of magnon-magnon interactions is observed to be in excellent agreement with theoretical predictions. The temperature evolution of the pair-mode spectrum demonstrates striking differences between two and three dimensions in the frequency renormalization and damping of short-wavelength magnons.

We report here the first observation of coupled two-magnon excitations (magnon-pair modes) in a two-dimensional magnetic system (the quadratic layer antiferromagnet, K_2NiF_4). The magnon-pair modes are zero-wave-vector excitations of zero spin consisting of one magnon from each antiferromagnetic sublattice.[1] The spectrum of light inelastically scattered by these magnon pairs is determined by the magnon dispersion relation, the joint magnon density of states (weighted according to the requirements of crystal symmetry), and the effects of magnon-magnon interactions. At zero temperature the pair-mode spectrum has been calculated[2] incorporating these factors. The predicted frequency and line shape are in excellent agreement with our experimental observations. We have also studied the temperature dependence of the magnon-pair-mode spectrum over a range between (0.1 and 1.5)T_N. For K_2NiF_4, $T_N = 97.1$°K.[3] Both the frequency and linewidth of the pair-mode spectrum exhibit much less drastic changes as T is increased above T_N than has been found for three-dimensional systems.[4-6] Comparison of our two-dimensional K_2NiF_4 results with those previously obtained in closely related three-dimensional systems (NiF_2[4] and $KNiF_3$[6]) demonstrates significant differences in the wave-vector dependence of the magnon frequency renormalization with temperature, and suggests quite different temperature behavior of the magnetic "coherence length."[7] These observations in two-dimensional K_2NiF_4 may be especially helpful in guiding a theory for magnon pairs at finite temperatures. Such a theory might be more readily developed in two dimensions because spin-wave theory remains valid to higher temperatures (relative to T_N) than in three dimensions. For example, the condition for applicability of spin-wave theory, $(T/JZS) \ll 1$, limits one to $T \ll 2T_N$ in the three-dimensional $KNiF_3$, but only to $T \ll 4T_N$ in two-dimensional K_2NiF_4.

The apparatus and techniques employed in these Raman experiments have been fully described elsewhere,[1] with the exception of the magnetic field apparatus. These magnetic field experiments were performed in a split-coil superconducting solenoid, having four directions of optical access, a variable temperature capability, and a maximum field strength of 100 kOe. The K_2NiF_4 crystals were grown by the horizontal zone-melting method, described previously.[3] The sample of K_2NiF_4 used here was ~2 mm on a side, of rather poor optical quality, and was not entirely a single crystal. These factors presumably prevented our observing the one-magnon scattering at the antiferromagnetic resonance (AFMR) frequency[8] and introduced some ambiguity into experimental determination of the magnon-pair polarization selection rules (see below).

The K_2NiF_4 crystal structure[9] consists of successive simple square NiF_2 planes separated by two KF planes, all stacked along the c direction. Both theory[10] and experiment[3,11] indicate that there is essentially no magnetic coupling between planes. Thus K_2NiF_4 may be considered a two-dimensional Heisenberg antiferromagnet, whose magnetic properties are adequately described by a single isotropic exchange interaction J and a small c-directed anisotropy field, H_A, both acting on atoms within the plane. At zero temperature the magnon energies E_k are[12]

$$E_{\vec{k}}^2 = (JSZ + g\mu H_A)^2 - (JSZ\gamma_{\vec{k}})^2, \tag{1}$$

where $\gamma_{\vec{k}} = \frac{1}{2}(\cos k_x a + \cos k_y a)$, and $Z = 4$ is the number of nearest neighbors. The scattering of an incident optical field, $\vec{E}_1$, into the scattered field, $\vec{E}_2$, by a two-magnon excitation is described by the interaction Hamiltonian[1,2]

$$H^{II} = A \sum_{\langle ij \rangle} (\vec{E}_1 \cdot \vec{\sigma}_{ij})(\vec{E}_2 \cdot \vec{\sigma}_{ij})(\vec{S}_i \cdot \vec{S}_j), \tag{2}$$

where $\vec{\sigma}_{ij}$ is a unit vector connecting the nearest-neighbor sites i and j. The magnon-pair spec-

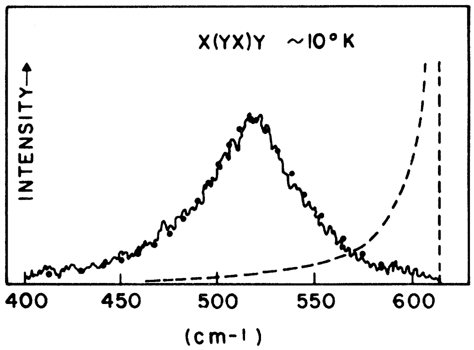

FIG. 1. Low-temperature Raman spectrum from magon-pair modes in K_2NiF_4. Dashed curve is theory neglecting magnon interaction effects. Dotted curve is theory including magnon interaction effects. Solid curve is experimental observation.

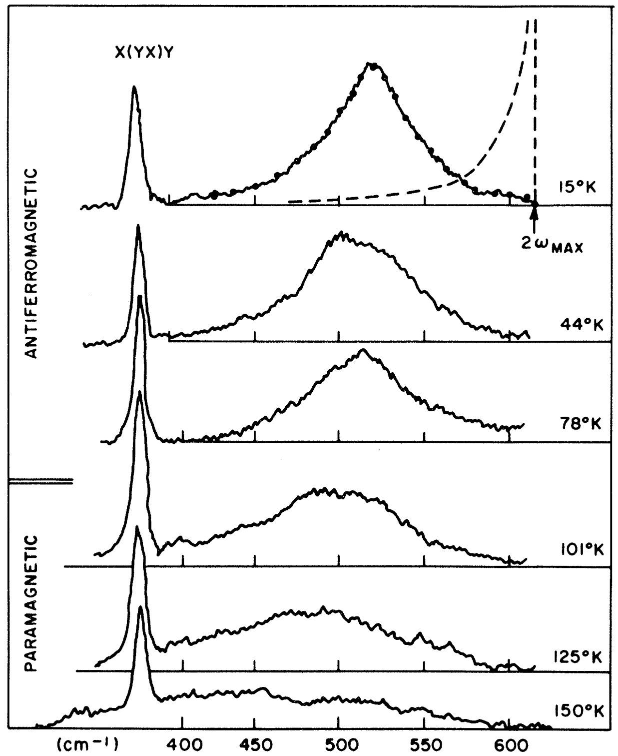

FIG. 2. Temperature evolution of magnon-pair-mode spectrum in K_2NiF_4. The sharp feature at ~375 cm^{-1} is an optic phonon. The maximum frequency at zero temperature is designated $\omega_{max} = 308$ cm^{-1}.

trum is calculated from Eq. (2) using Eq. (1) to provide the density of states. The predicted line shape[2] in the absence of magnon-magnon interactions is given by the dashed curve in Fig. 1. The predicted line shape taking magnon-magnon interactions into account (necessary even at zero temperature because of the local nature of the interactions in H^{II}) is given by the solid dots.[2] This is in excellent agreement with the observed spectrum (solid curve in Fig. 1). The values $J = 76.8$ cm^{-1} and $g\beta H_A = 0.59$ cm^{-1} determined from neutron-scattering experiments[11] have been used in Parkinson's line shape expression.[2] We emphasize that there are no adjustable parameters (aside from an intensity normalization) in the theory, so that the pair-mode Raman spectrum can be considered a means of measuring the dominant exchange, J, to an accuracy of a few percent or better. The value of H_A can in principle be inferred from the one-magnon light scattering at the AFMR frequency, not observed here.

We have also examined the magnetic field behavior and the polarization selection rules of the low-temperature pair-mode spectrum. At $T \approx 10$°K with the magnetic field parallel to c direction, no splitting or change in intensity, shape, or width could be observed for field strengths of up to 100 kOe. This result confirms the "spin zero" assignment[1] (one magnon from each sublattice) of the magnon-pair mode. Since $\vec{\sigma}_{ij}$ lies in the x-y plane, Eq. (2) requires that both the incident and scattered fields be polarized perpendicular to the c axis. While the Raman tensor elements α_{xy} and α_{yx} observed are at least a factor of 5 larger than α_{yz} or α_{xz}, some scattering was observed in these latter geometries. We tentatively ascribe this, however, to the less than ideal condition of the sample described above.

As the temperature is increased, the magnon-pair spectrum of K_2NiF_4 shown in Fig. 2 broadens and shifts to lower frequencies. Comparison of the 78 and 101°K traces of Fig. 2 illustrates the insensitivity of the pair-mode spectrum to the phase transition at 97.1°K. The persistence of a well-defined pair-mode spectrum up to ~1.5 $\times T_N$ is also evident in Fig. 2. In these respects the K_2NiF_4 spectra are qualitatively similar to those previously reported for three-dimensional antiferromagnets.[4-6] However, a quantitative comparison between two- and three-dimensional behavior reveals significant differences.

In Fig. 3 we compare the temperature dependence of the normalized pair-mode frequency, $\omega(T)/\omega(0)$, as well as the full width at half-maximum $\Gamma(T)$ for the two-dimensional K_2NiF_4 and its closest three-dimensional counterparts, $KNiF_3$[6] and NiF_2.[4] We take $\omega(T)$ to be the average frequency (zeroth moment) of the pair-mode

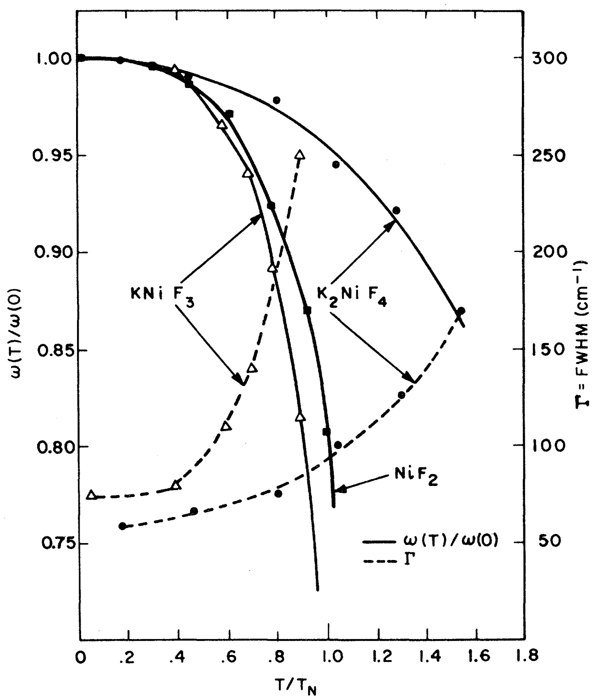

FIG. 3. Comparison of temperature dependences for normalized magnon-pair-mode frequencies $[\omega(T)/\omega(0)]$ and full widths at half-maximum Γ for two-dimensional K_2NiF_4 and three-dimensional $KNiF_3$ and NiF_2.

spectrum.

As with all other three-dimensional cases studied to date,[4-6] the pair-mode frequencies in $KNiF_3$ and NiF_2 are renormalized by 25% or more at T_N. This contrasts sharply with the less than 5% renormalization in K_2NiF_4. Similar differences exist for the temperature behavior of the pair-mode linewidth. For $KNiF_3$[6] and NiF_2[4] the ratios $\Gamma(T_N)/\Gamma(0)$ are ~4 and 5.2, respectively, much greater than the value 1.5 found in K_2NiF_4.

Since no finite-temperature theory exists for magnon pairs in either two or three dimensions, a quantitative discussion of our results is not possible at present. Instead, we will correlate our observations with other experiments and will indicate qualitatively their likely theoretical significance.

The magnon-pair-mode spectrum is dominated by short-wavelength (Brillouin-zone boundary) magnons, so that its temperature evolution is determined by the temperature dependence of (1) short-wavelength magnon energies and (2) lifetimes, and (3) those magnon-interactions effects described at zero temperature in Fig. 1. If we ignore (3) (because it probably varies weakly with temperature), we may discuss the results of Fig. 2 in terms of the temperature behavior of short-wavelength magnon energies and lifetimes. It is enlightening to compare this behavior with (a) the behavior of long-wavelength magnons in two dimensions (K_2NiF_4) and (b) the behavior of short-wavelength magnons in three dimensions.

Comparison (a) is available from the antiferromagnetic-resonance experiments of Birgeneau, DeRosa, and Guggenheim.[8] They find that the frequency of the long-wavelength magnon in K_2NiF_4 approaches zero at T_N and scales with the sublattice magnetization. Thus the Brillouin-zone–center magnons are 100% renormalized at T_N, whereas the zone-boundary magnons we observe are renormalized by less than 5% at T_N. It is evident therefore that the magnon energy renormalization with temperature is strongly dependent upon wavelength.

While no neutron-scattering studies of short-wavelength magnons in two dimensions at elevated temperatures have been reported, such experiments have been performed for fairly long wavelength magnons.[12] Magnons of wavelength 110 Å were followed up to $1.1T_N$ and exhibited no appreciable thermal effects.[12] Thus only very long wavelength ($\lambda > 100$ Å) magnons are strongly temperature dependent in two dimensions.

Wavelength dependence of the magnon renormalization has also been distinguished experimentally for three-dimensional systems.[4,13,14] However, the difference between zone-center and zone-boundary renormalizations is less striking than for two dimensions. The curves in Fig. 3 thus illustrate that appreciable energy renormalization as well as appreciable damping of zone-boundary magnons sets in at much higher temperatures, relative to T_N, in two dimensions than in three dimensions.

The above experimental results and discussion clearly indicate that there should be some characteristic (temperature dependent) length L_c for which magnons with $\lambda < L_c$ experience much less damping and energy renormalization than do magnons with $\lambda > L_c$. Lines has recently identified a length with similar properties within an approximate theory.[7] This length, which he calls the "coherence length," is distinct from, but related to, the usually defined correlation length. Our results appear to be in qualitative agreement with the behavior expected from Lines' theory. They are also consistent with the direct measurements of the correlation length[11] in K_2NiF_4 which show that its value for a given $(T-T_N)$ is much greater than for three-dimensional systems.

In summary, we have observed magnon-pair excitations in two-dimensional K_2NiF_4 at a variety of temperatures and magnetic fields. The results imply very little energy renormalization at T_N and persistence well into the paramagnetic phase of zone-boundary magnons. The excellent quantitative success of Parkinson's calculation for the low-temperature magnon-pair line shape together with the qualitative association between the pair-mode spectrum and the wavelength dependence for magnon renormalization at finite temperatures give rise to the hope that a quantitative finite-temperature theory of magnon pairs may soon be developed. Such a theory is especially desirable because the magnon-pair Raman spectrum directly measures aspects of a four-spin correlation function,[4] from which several quantities of interest, such as the magnetic contributions to the specific heat[15] and to sound-wave attenuation,[16] may be extracted.

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EXTENSION OF THE VARIABLE MOMENT OF INERTIA MODEL TOWARD MAGIC NUCLEI

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Ground-state bands of the quadrupole type are found in even-even nuclei ranging from the most deformed to doubly magic. Their analysis indicates how an "internal stress" which tends to keep the nucleus spherical is overcome as the angular momentum increases.

An analysis[1] of ground-state bands in even-even nuclei led to a remarkable result: While the unified model contains the assumption that the moment of inertia of a deformed nucleus is, in first approximation, independent of the angular momentum, and that a nucleus which has a spherical ground state remains spherical[2] even if its surface undergoes 1-, 2-,···, n-phonon oscillations of the quadrupole type, the new analysis yielded the variable moment of inertia (VMI) "model." This model consists of two simple empirical laws:

(I) For rotational as well as "transition" and "vibrational" nuclei, the energy is given by

$$E(J)=\tfrac{1}{2}C(\mathscr{I}-\mathscr{I}_0)^2+J(J+1)/2\mathscr{I}\quad(\hbar^2=1),\qquad(1)$$

where $\mathscr{I}_J$ is chosen to minimize $E(J)$,

$$\partial E(J)/\partial\mathscr{I}=0;\qquad(2)$$

and

$$C>0\qquad(3)$$

for each state.[3] The limit of validity for this law was taken to be $\mathscr{I}_0>0$, equivalent to the condition