and 162.5/165 cm^{-1} (Ni^{2+}:MnF_2) lines. Measurement of the possible antisymmetry of the scattering tensor for this line has been precluded so far by the small size of the available samples. This measurement should be done since a one-magnon line would have an antisymmetric scattering tensor ($\alpha_{xz} + \alpha_{zx} = 0$).

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OBSERVATION OF LOCALIZED MAGNONS BY RAMAN SCATTERING IN Ni-DOPED MnF_2

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Weak Raman scattering from nickel-doped MnF_2 observed below T_N has been identified with the simultaneous excitation of pairs of single-spin-deviation impurity magnon modes. The polarization and scattering frequency are accurately predicted by a simple model allowing for an interaction between the modes.

In this Letter we describe the first experimental observation of localized magnons using Raman spectroscopy.[1] Localized magnon modes have already been identified by other experimental techniques such as fluorescence studies,[2] ESR,[3] and neutron scattering.[4] The localized magnons associated with nickel impurities in MnF_2 comprise a particularly favorable system for investigation since the spin-wave spectrum of MnF_2 is well known and a magnon impurity mode has been observed in fluorescence at 120.4 cm^{-1}. Futhermore, both the Ni^{2+} and Mn^{2+} low-lying states have essentially no orbital momentum, which greatly simplifies their theoretical description.

Raman scattering was studied in a single crystal of MnF_2 containing 1-2% Ni using a double-grating CODERG monochromator and a linearly polarized argon ion laser providing 30-100 mW in the 4880-A line. The Raman scattered light was detected by photon counting using a cooled photomultiplier with "S" sensitivity. The spectral slit width was energy limited to about 3 cm^{-1}. For low-temperature studies the sample

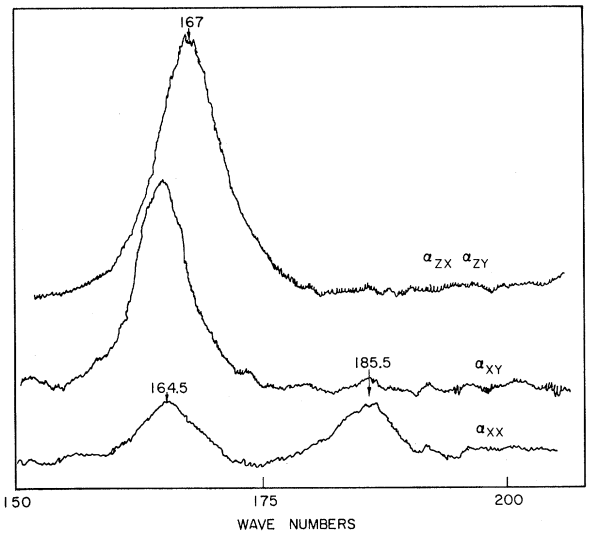

FIG. 1. Experimental Raman spectra in 1-2% Ni-doped MnF_2 at 8°K between 150 and 200 cm^{-1} for different polarizations. The relative intensity among the different spectra is only approximate. The lines near 165 cm^{-1} are observed only in crystals containing nickel impurities, at temperatures below 67.3°K (T_N). The weak scattering near 185 cm^{-1} is not understood.

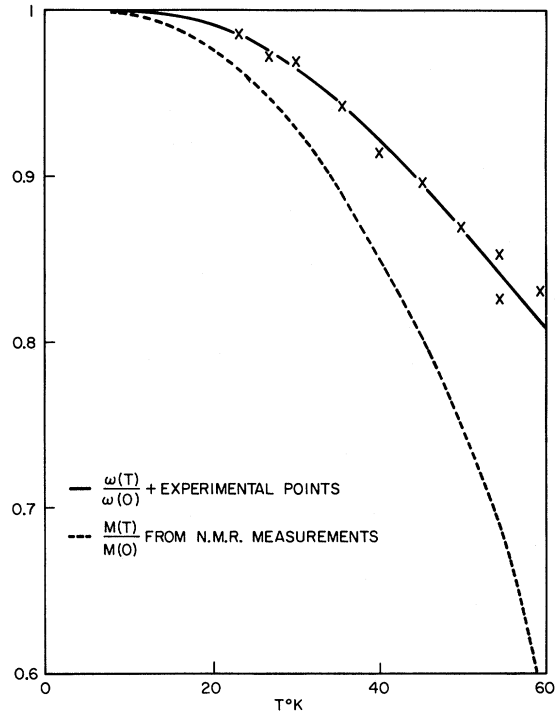

FIG. 2. Temperature dependence of the peak frequency observed for α_{zx} relative to the position at 2°K. The dashed curve gives the temperature dependence of the magnetization as measured by NMR [see V. Jaccarino, in Magnetism, edited by G. T. Rado and M. Suhl (Academic Press, Inc., New York, 1965), Vol. II(A), p. 331].

was immersed in superfluid helium ($T \sim 2$°K), while higher temperatures could be maintained using a gas-exchange cryostat. The geometrical form of our x-ray oriented crystal permitted all polarizations[5] to be studied except α_{zz}. However, to improve signal-to-noise ratio, some spectra as shown in Fig. 1 were taken without polarizing the scattered light.

At 2°K, in addition to the phonon and two-magnon scattering observed in the pure MnF_2, scattering was observed between 160 and 170 cm^{-1}, with integrated intensities for α_{xy} and α_{zx} some five or six times smaller than for the corresponding intrinsic two-magnon scattering. A line centered at 164.5 cm^{-1}, with an experimental half-width of 6.5 cm^{-1}, is polarized principally for α_{xy}. An α_{zx} line is centered at 167 cm^{-1}, with a half-width of 7 cm^{-1}. The α_{xx} spectrum is similar to α_{xy}, but significantly less intense. Finally a very weak line is found at 185.5 cm^{-1}, appearing only in α_{xx}. Figure 1 shows (α_{zx}, α_{zy}), α_{xx}, and α_{xy} at 8°K.

As shown in Fig. 2, the relative frequency $\omega(T)/\omega(0)$ of the α_{zx} peak is considerably less sensitive to temperature up to 60°K than the relative sublattice magnetization $M(T)/M(0)$ of pure MnF_2. Between 2 and 60°K, the half-width increases from 7 to about 40 cm^{-1}, and the integrated intensity diminishes by a factor of 10.

This behavior is similar to that observed for the intrinsic two-magnon scattering,[5] which corresponds to the excitation of a magnon on each sublattice with equal and oppositely signed wave vectors. As with the nickel-doped crystals, two scattering peaks are observed at energies slightly smaller (because of a magnon-magnon interaction[6]) than the sum of the energies of the two magnons, with the frequency of the α_{xz} peak slightly larger than that for α_{xy}. The similarity of the intrinsic to the nickel impurity spectrum suggests that the scattering at 164 (α_{xy}) and 167 cm^{-1} (α_{zx}) corresponds to the simultaneous excitation of two impurity magnons, one localized on the impurity ion and one on the other sublattice, and with the strength of the scattering derived from exchange interactions involving second neighbors, as suggested for the intrinsic case.[7]

In the paramagnetic phase, the site group of the nickel ion is D_{2h}, but in the ordered, antiferromagnetic phase this is lowered to the unitary magnetic site group C_{2h} by the restrictions

placed on the antiunitary time-reversal operator. Since Raman scattering proceeds without a change of parity and since we may take the ground state to be A_g, then the excited states reached by Raman scattering must also be of even parity, and therefore must belong to either A_g or B_g. Polarizations α_{xx} or α_{xy} will involve two magnons having A_g as their direct product, while for α_{zx} or α_{zy} the two magnons will have B_g. Assuming that one of the scattered modes is the s_0 (120.4-cm^{-1}) mode observed in fluorescence[8] (this is the only impurity mode residing mainly on the impurity sublattice), which has the representation A_g, then the representations given for the product states are also the representations for the impurity mode on the sublattice opposite to that of the impurity. The s_0 mode has been shown to be highly localized on the Ni ion.[8]

Although we may expect the s_0 mode to couple to an infinite number of even-parity impurity modes on the other sublattice which may be constructed from the complete set of MnF_2 wave-vector states, only those low-order modes which have their spin deviations concentrated on the second neighbors to the nickel ion will have appreciable Raman intensity. Theoretical calculations recently reported for MnF_2[9,10] show that one s (called s_1), three p, three d, and one f mode can be constructed from the eight second neighbors (these considerations neglect the first-neighbor exchange, J_1, and the possible inequivalence of exchange between the impurity ion and the two classes of second neighbors). Of these, only s_1 and the three d modes, d_{xy}, d_{xz}, and d_{yz}, have even parity. Since s_1 may also comprise spin deviations on the nickel ion, its energy is expected to be smaller than that of the d modes. The ratio of the energy of the d to that of the s_0 mode of Ni in MnF_2 has been estimated[10] by a Green's function calculation (neglecting J_1) to be 0.426. From the experimental value for s_0, we then estimate a value of 51.3 cm^{-1} for the d mode. Thus, the 185-cm^{-1} mode, which like s_1 is A_g, cannot be assigned to s_1, and it seems likely that s_1 is not observed. The 164-cm^{-1} mode (observed in α_{xx}, α_{xy}) transforms as A_g, and we assign it to d_{xy} while the 167-cm^{-1} (α_{zx}) mode transforms as B_g, or d_{xz}, d_{yz}. The splitting between these modes arises from the first-neighbor exchange, which affects, again, d_{xy} differently from d_{xz}, d_{yz}. Since the d_{xy} mode does not depend on J_1, its energy should agree well with the Green's function estimate. The α_{xy} peak frequency should equal the sum of the s_0 and d modes.

However, this sum, 171.7 cm^{-1}, is 7.2 cm^{-1} larger than the observed α_{xy} scattering peak. This is to be expected, since we have neglected the interaction between the s_0 and d modes which, like that observed in the intrinsic two-magnon scattering,[11] is significant. Since a Green's function calculation of the interaction between impurity magnons is not yet available, we present a simpler, Ising model estimate of is s_0-d_{xy} scattering frequency.

If i and j represent second neighbors (on opposite sublattices), the interaction between two Mn ions is $-2J_{\mathrm{MnMn}}\vec{S}_{i\mathrm{Mn}}\cdot\vec{S}_{j\mathrm{Mn}}$ while the interaction between a Ni and a Mn ion is $-2J_{\mathrm{MnNi}}\vec{S}_{i\mathrm{Mn}}\cdot\vec{S}_{j\mathrm{Ni}}$. For 1-2% Ni in MnF_2, the most probable situation for a Ni-Mn pair corresponds to the Ni ion surrounded by eight Mn second neighbors with the Mn ions each surrounded by an additional seven Mn second neighbors. In the scattering process, $M_{s_{\mathrm{Ni}}}$ changes from -1 to 0, while $M_{s_{\mathrm{Mn}}}$ changes from $+\frac{5}{2}$ to $+\frac{3}{2}$.

For large molecular fields, the s_0 mode energy can be represented[8] by an exchange term and a crystal field term:

$$E_{s_0} = -40J_{\mathrm{NiMn}} - D = 120.4\ \mathrm{cm}^{-1},$$

where $J_{\mathrm{NiMn}} = -3.11\ \mathrm{cm}^{-1}$ and $D = 4.05\ \mathrm{cm}^{-1}$. The d_{xy} mode created separately from the s_0 mode is

$$E_d = -35J_{\mathrm{MnMn}} - 2J_{\mathrm{NiMn}} = 48.9\ \mathrm{cm}^{-1}.$$

The energy for both modes simultaneously at the same impurity is

$$E_{s_0-d} = -40J_{\mathrm{NiMn}} - D - 35J_{\mathrm{MnMn}} = 163.1\ \mathrm{cm}^{-1}.$$

Thus the interaction energy ΔE is

$$\Delta E = E_{s_0} + E_d - E_{s_0-d} = +2J_{\mathrm{NiMn}} = -6.22\ \mathrm{cm}^{-1}.$$

Note that E_d compares favorably with the Green's function result (which includes anisotropy), and E_{s_0-d} is in excellent agreement with the α_{xy} peak at 164.5 cm^{-1}. In fact, if we add to the d-mode energy 0.8 cm^{-1}, which corrects[12] for the anisotropy energy of the Mn ions, we obtain 163.9 cm^{-1}, even closer to the experimental value. This agreement may be considered evidence that the d modes are mainly localized on the second neighbors to the nickel impurity as assumed in the above calculation. However, this conclusion applies strictly to the d mode while interacting with the s_0 mode since the interaction

may significantly contribute to its localization.

Further analysis of the shape and intensity of the scattering lines and of their temperature dependence must await a more sophisticated model. At the present time the 185-cm^{-1} scattering is not understood.

We have also observed an absorption band in the infrared for $E \| c$ at 167 cm^{-1}, similar to the α_{zx} Raman scattering peak. Such absorption must arise from odd-parity modes such as the p and f modes mentioned above. In fact, the Green's function calculations[10] predict that these modes lie very close to the d modes. Details of this work will be published in a future communication.

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[12]This number corrects for crystal-field and dipolar anisotropy appropriate to the perfect crystal.

REORIENTATION MEASUREMENT IN Mg^{24}

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The reorientation effect in Coulomb excitation with S^{32} ions was used to deduce the static quadrupole moment of the first excited $J^\pi = 2^+$ state in Mg^{24}. The value obtained in this experiment is $Q_{2^+} = -0.26 \pm 0.08$ b.

In the last few years the reorientation effect in Coulomb excitation was successfully used[1] to determine the quadrupole moments of excited states in the mass region $A > 100$. This Letter describes a measurement of the static quadrupole moment of the first excited $J^\pi = 2^+$ state in Mg^{24}. In this case, the influence of the static quadrupole moment of the 2^+ state on the multiple Coulomb excitation is expected to be large. Assuming an intrinsic quadrupole moment $Q_0 \sim 0.8$ b for Mg^{24} and using S^{32} ions, the reorientation effect amounts to a 25% change in the excitation cross section for backward angles. The change due to the population of the 2^+ state through other low-lying excited states in Mg^{24} is smaller than 1%. The estimated correction due to transitions via the giant dipole resonance states[1] is also less than 1%. Therefore, reorientation measurements in light nuclei should provide reliable values of the static quadrupole moment of the first excited state.

In the present experiment, sulfur beams of 42, 48, and 55.6 MeV produced by the High Voltage Engineering Corporation Model MP tandem Van de Graaff accelerator of the Max-Planck-Institut was used to bombard Mg^{24} self-supporting targets of 76, 150, 310, and 550 $\mu g/cm^2$ thickness. At the bombarding energies chosen, the elastic scattering cross section was measured in this laboratory and does not show any deviation from the Rutherford cross section. The gamma rays following the Coulomb excitation have been de-