obtained above with the results from other compounds. For Fe^{57} in MgO we adopt an average value $\Delta E(4.2°\mathrm{K}) = 0.315\pm0.015$ mm/sec.[7] For this case Ham[8] estimates the combined effects of random strains and Jahn-Teller coupling to yield $F\simeq 0.09$ ($\pm 5\%$). Thus for MgO, $\Delta E_0 = 3.5\pm0.3$ mm/sec. Presumably the 14% reduction in $\langle 1/r^3\rangle_{\mathrm{eff}}$ from that in $\mathrm{FeSiF_6\cdot 6H_2O}$ is due to greater covalency for MgO. The same qualitative conclusion may be drawn from the isomer shifts (IS), which are 1.06 ± 0.01 and 1.41 ± 0.01 mm/sec for divalent Fe^{57} in MgO and $\mathrm{FeSiF_6\cdot 6H_2O}$, respectively (measured at room temperature relative to iron).

If it is assumed that covalency reduces the spin-orbit parameter in proportion to $\langle 1/r^3\rangle_{\mathrm{eff}}$, one arrives at $-\lambda\simeq 80$, 90, and 100 cm^{-1} for Fe^{2+} in MgO, $\mathrm{FeSiF_6\cdot 6H_2O}$, and the free ion, respectively. Corresponding values for $\langle 1/r^3\rangle_{\mathrm{eff}}$ would be approximately 3.0, 3.5, and $4.0a_0^{-3}$.

As a final remark, we note that a recent analysis[9] of the problem of Fe^{2+} in FeF$_2$ yields $\Delta E_0\simeq 3.5\pm0.1$ mm/sec, IS $=1.36$ mm/sec, and $-\lambda\simeq 85$ cm^{-1}, which suggest covalency reduction effects between those found in MgO and $\mathrm{FeSiF_6\cdot 6H_2O}$. In contrast, the quadrupole splitting and IS of divalent Fe^{57} in highly doped CaF$_2$ [10] appear to be greater than that in $\mathrm{FeSiF_6\cdot 6H_2O}$, suggesting even less covalency.

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Raman Scattering from Localized Magnons*

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Additional observations on Raman scattering from localized spin waves are reported. Spectra showing the one-magnon S_0-like mode in MnF$_2$:Fe and one d-like mode in RbMnF$_3$:Ni are presented.

In the past two years there have been several observations of Raman scattering from pairs of localized magnons in antiferromagnetic MnF$_2$.[1–3] These experiments have been interpreted in terms of molecular-field models and simple combinations of Green's-function and molecular-field calculations. Recently, Thorpe[4] has completed a calculation which predicts the line shape for the two-magnon impurity mode in RbMnF$_3$:Ni and fitted it to some unpublished data taken subsequent to our original measurements.[1] We present here both the new two-magnon data and also observations of what we believe to be two different types of one-magnon modes localized at an impurity site.

RbMnF$_3$ is in many respects a better host for our purposes than MnF$_2$. It has cubic, rather than tetragonal, symmetry; with a single exchange constant and negligible anisotropy. When doped with Ni^{2+}, the energy of the impurity-centered S_0 mode is 238 cm^{-1},[5] which is far higher than the maximum pure-crystal spin-wave energy of 70.5 cm^{-1}. In addition, in contrast to the case of MnF$_2$:Ni and MnF$_2$:Fe, the energy of the localized d-like modes of the neighboring host spins is several cm^{-1} above the top of the spin-wave band of the pure host.[6] As a result, it becomes possible to observe the d-like localized modes.

The high S_0 energy fixes this mode almost entirely on the impurity, and as in the MnF$_2$ systems, it has the effect both of making the molecular-field model an excellent approximation and of allowing one to decouple

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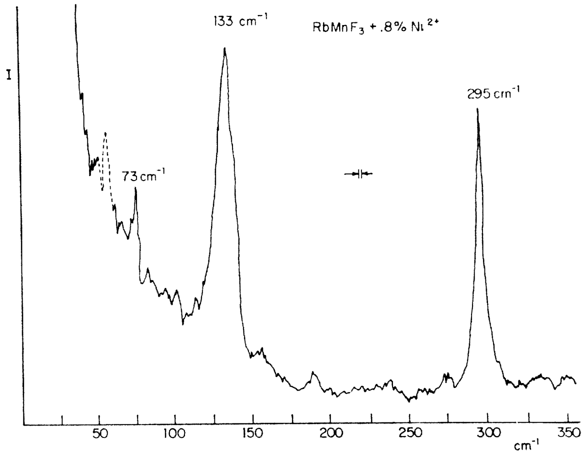

FIG. 1. Raman spectrum of 0.8%-Ni^{+2}-doped RbMnF$_3$. The dashed line is an emission line. The 73- and 295-cm^{-1} lines are associated with the Ni^{2+} impurities. The spectrum is taken near 8°K and contains both α_{zz} and α_{xy} polarizations.

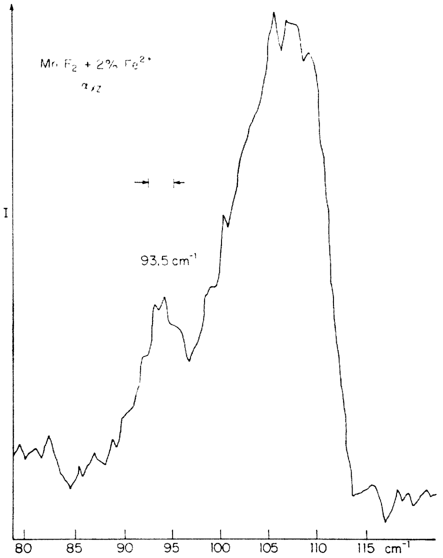

FIG. 2. Raman spectra of 2%-Fe^{2+}-doped MnF$_2$ at 8°K. The 93.5-cm^{-1} line is the one-magnon impurity mode of S_0 symmetry. The line near 106 cm^{-1} is due to intrinsic two-magnon scattering.

the three-body Green's functions into more readily calculated two-body equations. The molecular-field model predicts the energies of the d modes and the $S_{0\text{-}d}$ modes to be

$$E_d = 74.6 \text{ cm}^{-1},$$

$$E_{S_{0\text{-}d}} = 238 + 74.6 - 15.8 = 295.2 \text{ cm}^{-1},$$

where the negative term is the magnon-magnon binding energy $2J'$. Green's-function calculations give[6]

$$E_d = 73.5 \text{ cm}^{-1},$$

$$E_{S_{0\text{-}d}} = 291 \text{ cm}^{-1},$$

and both computations agree well with the experimental values, which are accurate to ±1 cm^{-1}:

$$E_d = 73 \text{ cm}^{-1},$$

$$E_{S_{0\text{-}d}} = 295 \text{ cm}^{-1}.$$

A typical experimental spectrum is shown in Fig. 1.

We do not find the S_0 mode itself, nor has this excitation been observable in MnF$_2$:Ni either with Raman scattering or with infrared absorbtion.[7] However, for the first time in a light-scattering experiment, we do find the S_0 mode in MnF$_2$:Fe at 93.5 cm^{-1}, in good agreement with infrared measurements[8] (Fig. 2).

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