tion direction of the incident light and magnetization direction, respectively, with the local trigonal axis of the site j, and assuming that the anisotropy energy for an Fe^{2+} ion can be expressed as

$$\epsilon_j = W\cos^2\beta_j,$$

then the torque (that is, the derivative of the anisotropy energy) is given by

$$L = A_1 \sin 2\varphi + B_1 \cos 2\varphi + \text{higher terms},$$

where φ is the angle between the magnetization and the [100] direction during the torque measurement, and

$$A_1 = \frac{2WN}{9C}(b\sin 2\theta + c\sin 2\Phi),$$

$$B_1 = \frac{WN}{18C}(b + c - b\cos 2\theta - c\cos 2\Phi),$$

where θ, Φ are angles between the polarization direction and the applied field during irradiation. Thus, if we keep the polarization direction θ fixed, then A_1 will be proportional to $\sin 2\Phi$, while B_1 is proportional to $\cos 2\Phi$. This is confirmed by the results in Fig. 2.

Further experimental results not indicated in the graph showed a slight dependence of the photomagnetic effect on the direction of the applied field during cooling to 4.2°K. This would indicate that some thermally induced anisotropy remains in the crystal even after prolonged irradiation.

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SPIN-OPTICAL-PHONON INTERACTION IN ANTIFERROMAGNETIC CoF_2

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(Received 23 October 1968)

Spin–optical-phonon interaction is observed in CoF_2 by the transfer of magnetic dipole intensity to the otherwise optically inactive E_g phonons. Theory predicts that the temperature dependence of the intensity is caused by the change in longitudinal spin correlation, and the change in energy and thermal population of the magnetic excitations. Experimentally, only the latter are important in CoF_2.

Unquenched orbital motion in the single-ion ground state can significantly alter the magnetic properties of the concentrated salt. Off-diagonal exchange coupled with the spin-orbit interaction produces multipole spin-spin interactions.[1-3] Purely electrostatic interactions between charge distributions can also lead to complex effective spin interactions.[4,5] Equally important is the presence of spin-lattice coupling which produces a large single-ion anisotropy field and is expected to couple magnetic excitations to phonons. The virtual emission and reabsorption of phonons will also generate multipole spin-spin interactions in suitable systems.[6-9] Since the above spin-spin interactions often have the same phenomenological form, it is difficult to extract unambiguously from measured spin-wave energies or ion-pair energy levels the various contributions to the ion-pair interactions and it is desirable to perform other measurements which are more sensitive to a particular interaction.

In the present Letter we discuss an experiment which directly measures the interaction between magnetic excitations and the phonons in the antiferromagnet, CoF_2, by observing the transfer of magnetic-dipole intensity to the otherwise optically inactive, $q = 0$, E_g phonon.[10] Since the experiment measures changes in the phonon state, it is sensitive to spin-phonon coupling but relatively insensitive to the complicated exchange interactions between Co^{2+} ions. The observed spin-phonon interaction is sensitive, however, to the gross changes in magnetic state produced by ordering and thermal population of the magnetic excitations, and it is the manifestation of these effects in the spin-phonon coupling that is discussed in the following.

Extrapolation of the E_g phonon frequency in

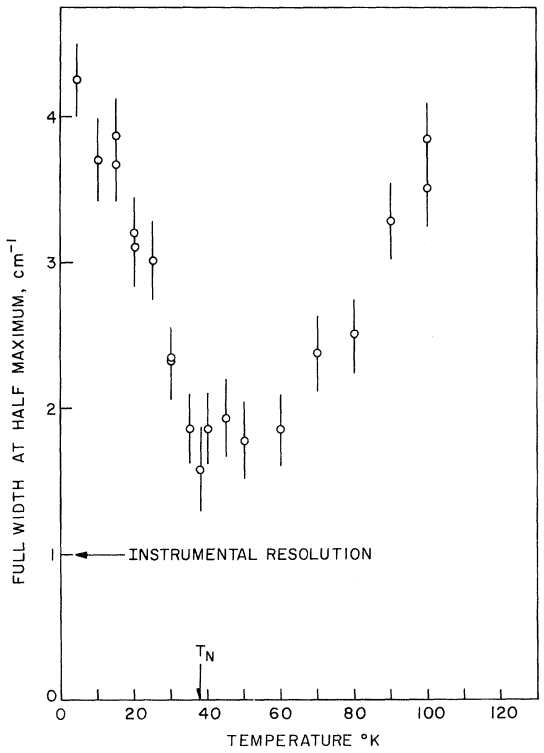

FIG. 1. Full width at half-maximum, uncorrected for instrumental resolution, as a function of temperature.

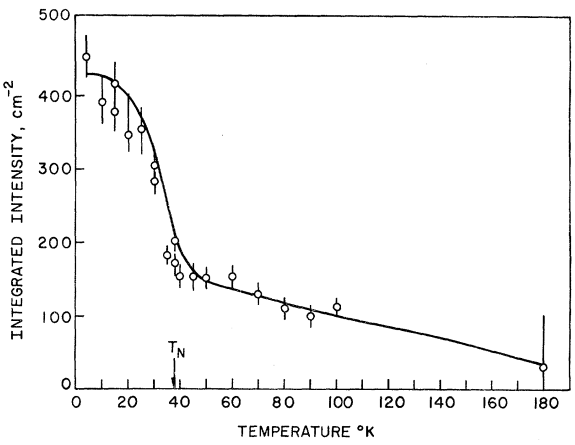

FIG. 2. Transverse magnetic dipole intensity as a function of temperature. Solid line is the theory which ignores intensity that is sensitive to the long-range longitudinal correlations.

MnF_2 and FeF_2[11] to CoF_2 predicts the optically inactive E_g mode to lie near 267 cm^{-1}. Far-infrared-absorption experiments in nominally pure CoF_2 reveal a temperature-sensitive magnetic-dipole transition in the vicinity of 256 cm^{-1}, polarized perpendicular to the c axis. A magnetic-impurity effect is precluded by the strength of the absorption (~426 cm^{-2} at 4.2°K), the lack of sample dependence, and weak magnetic-field dependence. Since the frequency is only weakly temperature dependent, multipole spin-wave excitations[12] are not considered. The line is interpreted as the E_g phonon allowed as a magnetic dipole transition by virtue of the strong spin-lattice coupling.

At 4.2°K the resonance occurs at 256 ± 0.5 cm^{-1}. As the temperature is raised, the frequency drops until it reaches ~250 cm^{-1} at ~140 °K. A weak discontinuity is noted at the Néel point. More striking behavior, obtained by measuring the linewidth as a function of temperature, is shown in Fig. 1. The linewidth narrows by more than a factor of 2 when approaching the critical point from the low-temperature side. In Fig. 2 the measured intensity is shown as a function of temperature. As the temperature is raised the intensity drops to ~$\frac{1}{3}$ the value at 4.2 °K. A much slower dependence is observed from T_N (38°K) to $4.5T_N$ (180°K). Above 180°K the line is too broad to accurately measure the intensity.

A theoretical discussion of the transfer of magnetic dipole intensity to the phonon best proceeds by starting in the disordered state. In the mean-field approximation, the local field acting on the $S = \frac{3}{2}$ state contains only the single-ion anisotropy

$$-\delta S_z^{\,2} \pm \gamma(S_x^{\,2} - S_y^{\,2}), \tag{1}$$

where the ± sign refers to the two different sites in the rutile unit cell which are designated 1 and 2. The anisotropy separates the excited doublet from the ground doublet by ~160 cm^{-1}.[12] At temperatures above the Néel point but not high enough to populate the excited doublet, the state of the ion on the α site in the jth unit cell, $|j, \alpha\rangle$, will be a random linear combination of the two possible states $|g_{\pm}^{\alpha}\rangle$ in the ground doublet. Despite the random character of the ground state we may still define local magnetic-exciton creation and destruction operators $a_{j,r}^{\dagger}, a_{j,r}$, where r runs over the different excitations which connect the ground state to one of the four possible singly excited states in the jth unit cell.

Interaction with the phonons is given by the fol-

lowing Hamiltonian:

$$N^{-1/2}\sum_{j}\sum_{\vec{q},p} i(b_{\vec{q},p}-b_{-\vec{q},p}^{\dagger})(\hbar/2m\omega_{\vec{q},p})^{1/2}\exp(-i\vec{q}\cdot\vec{r}_j)$$
$$\times\{\beta_1 f_1(\vec{q},p)(S_zS_x+S_xS_z)_j^1-\beta_2 f_2(\vec{q},p)(S_zS_x+S_xS_z)_j^2$$
$$-\beta_2 f_2(\vec{q},p)(S_zS_y+S_yS_z)_j^1+\beta_1 f_1(\vec{q},p)(S_zS_y+S_yS_z)_j^2\}, \tag{2}$$

where $f_1(\vec{q},p)$ and $f_2(\vec{q},p)$ are the structure factors for the interaction of the $\vec{q},p$ phonon with the Q_{zx} quadrupole moments on sites 1 and 2, respectively. β_1 and β_2 measure the strength of the coupling. Expressing the quadrupole operators as exciton creation and destruction operators generates the following total Hamiltonian:

$$H=\sum_{\vec{q},p}\hbar\omega_{\vec{q},p}b_{\vec{q},p}^{\dagger}b_{\vec{q},p}+\sum_{j,r}\hbar\omega_0 a_{j,r}^{\dagger}a_{j,r}$$
$$+\sum_{\vec{q},p}\sum_{j,r}\{R_{j,r;\vec{q},p}^{*}a_{j,r}b_{\vec{q},p}+R_{j,r;\vec{q},p}a_{j,r}^{\dagger}b_{\vec{q},p}^{\dagger}$$
$$+S_{j,r;\vec{q},p}b_{\vec{q},p}^{\dagger}a_{j,r}+S_{j,r;\vec{q},p}^{*}b_{\vec{q},p}a_{j,r}^{\dagger}\}, \tag{3}$$

where R and S are functions of the random ground state in the jth cell. Since exciton dispersion is of minor consequence, it is ignored throughout and we write $\hbar\omega_{j,r}=\hbar\omega_0$ as shown in (3).

Although the Hamiltonian is bilinear, interesting behavior, as a function of temperature, is obtained for the following reasons: (1) The random character of the coupling is partially removed for $T<T_N$, (2) the exciton energy $\hbar\omega_0$ is a strong function of temperature below T_N, and (3) the excitons cease to behave as bosons for $kT\sim\hbar\omega_0$. The effect on the intensity transferred to the phonon can be calculated from the frequency-dependent magnetic susceptibility obtained from a suitable set of retarded Green's functions. The system is decoupled by the following approximation:

$$\langle\langle[a_{j,r}a_{j'r'}^{\dagger}]b_{\vec{q},p}|a_{i''r''}\rangle\rangle=\langle[a_{j,r}a_{j'r'}^{\dagger}]\rangle\langle\langle b_{\vec{q},p}|a_{i''r''}\rangle\rangle,$$

where $\langle[a_{j,r}a_{j'r'}^{\dagger}]\rangle=\delta_{j,j'}\delta_{r,r'}\tanh(\hbar\omega_0/2kT)$, and solved by perturbation theory.[13]

We obtain for the real part of the transverse susceptibility χ_{xx}, correct to second order in the coupling constants,

$$\chi_{xx}(\omega)=\frac{(g\mu_B)^2}{\Omega_0}(s_x^2+s_y^2)\frac{2\hbar\omega_0}{(\hbar\omega_0)^2-(\hbar\omega)^2}I(T)$$
$$+3I(T)^2\frac{(g\mu_B)^2}{\Omega_0}\sum_{\vec{q},p}\left(\frac{\hbar}{2m\omega_{\vec{q},p}}\right)\frac{2\hbar\omega_{\vec{q},p}}{(\hbar\omega_{\vec{q},p})^2-(\hbar\omega)^2}\frac{4(\hbar\omega_0)^2}{[(\hbar\omega)^2-(\hbar\omega_0)^2]^2}$$
$$\times\{(1/N)|\sum_j[\beta_1 f_1(\vec{q},p)s_xS_z^1(j)-\beta_2 f_2(\vec{q},p)s_yS_z^2(j)]\exp(-i\vec{q}\cdot r_j)|^2$$
$$+|\beta_1 f_1(\vec{q},p)s^y-\beta_2 f_2(\vec{q},p)s^x|^2\delta_{\vec{q},0}\delta_{p,Eq}\}, \tag{4}$$

where $s_x=\langle e_-^1|S_x|g_+^1\rangle$, $s_y=\langle e_-^1|S_y|g_+^1\rangle$, $S_z^{\alpha}(j)=\langle j,\alpha|S_z|j\alpha\rangle/\langle g_+^1|S_z|g_+^1\rangle$, and $I(T)=\tanh(\hbar\omega_0/2kT)$.

The first term simply describes resonance at the exciton frequency $\hbar\omega_0$, while the second term gives the transfer of magnetic dipole intensity to the phonons. Since $S_z^{\alpha}(j)$ is a random variable in the disordered state, the first phonon term couples to all phonons. We obtain a singular term at $\vec{q}=0$ only if there is a long-range correlation in S_z. Consequently, sharp-line phonon absorption from this term monitors a combination of the the infinite-range correlation functions $\langle S_z^1(0) \times S_z^1(\infty)\rangle$ and $\langle S_z^1(0) S_z^2(\infty)\rangle$ and will only be seen below T_N. The second part of the phonon coupling is independent of the random character of the ground state and will persist in both the ordered and disordered states. Nevertheless, it will exhibit a temperature dependence through $I(T)^2$ and the temperature variation in $\hbar\omega_0(T)$.

Symmetry requires that $\chi_{yy}=\chi_{xx}$. A similar expression for χ_{zz} can also be derived. Only infinite-range correlations in the transverse-spin components will give sharp-line phonon absorption in this polarization. Since there are no such correlations at any temperature in CoF_2, no sharp-line E_g phonon absorption should be seen in σ polarization as is the case.

Above T_N only the second phonon term in (3) can give absorption. Since we know $\hbar\omega_0(T)$, we can then fit the high-temperature part by adjusting the interaction parameter

$$|\beta_1 f_1(0,E_g)s_y - \beta_2 f_2(0,E_g)s_x|\,(\hbar/2m\omega_{0,E_g})^{1/2} = 18\ \mathrm{cm}^{-1}.$$

Since $\hbar\omega_0(T)$ [12] is independent of T above T_N, the slow variation in intensity is produced by $I^2(T)$, i.e., it is caused by the thermal population of the exciton states. The behavior of this term below T_N can be had by including the temperature variation of $\hbar\omega_0$. The results are shown by the solid line in Fig. 2. No room is left for a contribution sensitive to the infinite-range correlation implying that

$$|\beta_1 f_1(0,E_g)s_y - \beta_2 f_2(0,E_g)s_x|^2 \geq 10|\beta_1 f_1(0,E_g)s_x + \beta_2 f_2(0,E_g)s_y|^2. \quad (5)$$

A priori there does not appear to be any reason why the correlation-sensitive term should be small, and it must be regarded as peculiar to the interaction constants in the CoF_2 system.

Similar effects should be seen in other systems that give evidence for a strong spin-lattice coupling. In particular the phonon absorption in UO_2,[9] seen only in the ordered state, may also be amenable to the same analysis.

The effect of the spin–optical-phonon interaction on the magnetic excitations in CoF_2 will be discussed in a future publication.

The authors would like to thank P. Martel of the Chalk River Nuclear Laboratories for providing a sample of CoF_2 and M. E. Lines for several interesting discussions. The technical assistance of F. DeRosa is gratefully acknowledged.

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