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Origin of circular dichroism in simple antiferromagnetic fluorides

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A very small circular dichroism (CD) was observed nearly 30 years ago in the magnon sidebands of the simple rutile antiferromagnetic fluorides, MnF_2 and CoF_2, at low temperatures. The source of this CD was not understood. Recent Raman circular intensity differential studies of the one-magnon Raman scattering in FeF_2 has revealed that the degeneracy of the antiferromagnetic magnon branches is lifted by a weak magnetic dipole-dipole interaction, as predicted by Pincus and Loudon and by White four decades ago. The origin of the observed CD in the magnon sidebands can now be traced to this same magnetic-dipole induced splitting.

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I. INTRODUCTION

The optical properties of antiferromagnetic insulators were actively investigated in the late 1960s and 1970s. This activity was spurred by the discovery of magnon sidebands in MnF_2 (Ref. 1) and the observation of two-magnon absorption in the far-infrared (FIR) spectrum of FeF_2 (Ref. 2). These two processes were shown to have a common origin by Tanabe and by Loudon and their respective co-workers.[3,4] In these materials, coupling between the intrinsic excitations of the lattice, i.e., excitons, magnons, and phonons, leads to the formation of cooperative transitions that account for much of the optical spectral structure. A fairly comprehensive review of this field appeared in 1986.[5]

The magnetic-field dependence of the magnon sidebands was investigated by Sell *et al.*[6] and no visible splittings of the sidebands were observed even at relatively high applied magnetic fields. The reason for this is that the magnon sideband is a compound excitation consisting of an exciton on one sublattice coupling with a magnon on the opposite sublattice. Since these excitations are on opposite sublattices, the application of a unidirectional magnetic field produces shifts in the exciton and magnon energies that are in opposite directions in energy, leading to a nearly complete cancellation of the splitting depending on the difference between the g values of the exciton and magnon. The net g factors of the sidebands were determined later to be of the order of $g^{\mathrm{eff}} = 0.005$.

To further explore the magnetic-field dependences, magnetic circular dichroism (MCD) was studied in a number of these rutile structure antiferromagnets in the mid 1970s, and it was demonstrated in a series of papers[7–9] that such techniques provided a very sensitive tool with which to probe the various excitations and provide complementary data to standard (or laser based) optical absorption, FIR, and Raman spectra of these materials. Dichroism in a material is defined as the difference between the absorption coefficients at a given wavelength of a medium for two conjugated polarizations of the incident light; thus circular dichroism (CD) arises from the difference in the optical absorption for right and left circularly polarized radiation. When these differences are induced by the application of an external magnetic field, the effect is termed MCD. In both CD and MCD, a unique direction is necessary in order to define the sense of circulation of the circularly polarized light; this is provided either by some unique crystalline axis or by the direction of the applied magnetic field. Materials showing CD are normally called ‘‘optically active’’ and such crystals are relatively rare, whereas MCD is quite common in paramagnetic systems where the applied magnetic field provides the unique axis. MCD is conjugated to Faraday rotation by the Kramers-Kronig relations and constitutes the resonant rather than the dispersive phenomena in the transformation.[10] The origins of CD and MCD are different and thus the effects are independent of each other and are simply additive.

Using these techniques, we were able to measure the magnetically induced splittings of sidebands; the effective g factor cited above resulted from such measurements, for example. We also reported at that time the observation of CD in certain magnon sidebands of MnF_2 (Ref. 7) and CoF_2 (Ref. 8), but not in FeF_2.[9] Based on the static rutile structure we would not expect such a CD response. We were able to eliminate experimental artifacts as the source of the observed CD, but were not able at that time to identify the physical basis for this unexpected observation. In this paper, we revisit this problem, which has lain fallow for nearly three decades. Although it is small, the existence of CD indicates that some aspect of the physics of these systems is not being incorporated.

The clue to understanding the CD results has come from a recent Raman-scattering experiment on one of these compounds.[11] One of us (W.M.Y.) had earlier proposed the combination of Raman-scattering and modulated dichroism techniques to explore the spin signature of Raman spectra of lattice excitations especially those intrinsic to magnetic materials.[12,13] These techniques, known generically as Raman optical activity (ROA), have been employed by atomic and molecular spectroscopists to study vibrational and magnetic properties of molecules in solution.[14] Analogous to the case of CD, ROA is observed through the difference in the intensity of the Raman line when excited with right and left circularly polarized incident light. For simple antiferromagnetic materials, the use of circularly polarized light leads to the excitation of magnons on one or the other magnetic sublattice. Hoffman *et al.* have successfully observed ROA in magnons, as well as in phonons, in transition-metal fluorides.[15,16] The results obtained from studies of $\mathrm{FeF_2}$ are particularly relevant to our deliberations here; Lockwood *et al.* were able to measure, using a 90° scattering geometry, a splitting of $0.09\pm0.02\ \mathrm{cm}^{-1}$ in the one-magnon Raman spectrum of $\mathrm{FeF_2}$ at low temperature in zero applied magnetic field.[11] Thus the sensitive ROA technique employed allowed a splitting of the two branches to be resolved and this indicated that the two antiferromagnetic branches of $\mathrm{FeF_2}$ are intrinsically nondegenerate. These results served to confirm our earlier CD results and gave us an indication as to the reason and origin of our observations. We conclude that the source of the CD is to be found in the magnetic dipolar interactions that exist between the two antiferromagnetic sublattices and which render them inequivalent when magnons are excited.[17]

II. OPTICAL DICHROISM IN RUTILE ANTIFERROMAGNETS

The compounds that we discuss here are the common transparent transition-metal difluorides $\mathrm{MnF_2}$, $\mathrm{CoF_2}$, and $\mathrm{FeF_2}$, all of which possess the tetragonal rutile structure. At room temperature, in the absence of magnetic ordering, the crystal symmetry is such as to preclude optical activity and no dichroism is observed in the optical absorption bands, which are dominated by phonon or vibronic assisted transitions. Once cooled below the antiferromagnetic ordering temperature (the Néel temperature T_{N}), these materials are describable in terms of a simple two-sublattice antiferromagnet and in principle they are ideal systems for studying magnetically dichroic phenomena. This is because for temperatures well below their Néel temperatures, the effective exchange fields are such that the two sublattices are totally spin-polarized parallel and antiparallel, respectively, to the unique c axis of the antiferromagnetic crystal. The ground states of ions in each sublattice as well as the collective magnetic excitations (spin waves or magnons) are then identical to each other in every respect except their polarization. The sole remaining energy degeneracy, which will respond to an applied magnetic field, is that of the two sublattices; hence, the origin of magnetic dichroism in these systems is totally related to the lifting of this energy equivalence for the electronic (exciton), spin (magnon), and the combined (magnon sideband) excitations.[6]

Since a unique axis of magnetization (the c axis in rutile structures) is involved, dichroic phenomena are only observed in transitions that are α polarization active. In this polarization, the **E** and **H** vectors of the transmitted light are perpendicular to the c axis thus defining the sense of circulation of the circularly polarized light. The α-polarized spectra studied in our earlier work contain pure electronic as well as compounded transitions such as magnon and phonon sidebands. The observed dichroic properties were reported as a function of applied magnetic field, as a function of applied uniaxial stress, and as a function of temperature.[18] At low temperatures, a very small zero-field CD signal was observed in two of the three compounds investigated.[7–9]

As noted above, for simple antiferromagnets in their ordered state, the ground state of one sublattice is nondegenerate and is identical to that of the other sublattice except for the sense of their magnetizations with respect to the unique crystalline tetragonal axis. If the eigenstate for the "up" or A sublattice is given by $|L,S\rangle$, then the corresponding "down" or B sublattice is represented by $|L,-S\rangle$ where L and S are the orbital and spin quantum numbers. For pure electronic transitions, the appropriate dipolar transition operators for circular polarized radiation transform as $L_{\pm}+2S_{\pm}$, where the $+$ and $-$ subscripts refer to right and left circular polarizations, respectively. If magnetic ions in one of the sublattices have a nonvanishing transition probability between two states for one of these operators, then the ions in the opposite sublattice will have an identical element for the corresponding conjugated operator. In the absence of an applied magnetic field, the sublattices are degenerate in energy and no CD should be observed, as the crystal symmetry precludes it. With an applied field, this energy degeneracy is lifted and the transition will manifest MCD. Since the two components of the transmitted light are 180° out of phase, phase sensitive detection techniques can provide the sense of the splitting, i.e., the ordering direction of the sublattice with respect to the applied field.

A similar argument applies for compounded excitations such as the electric dipole allowed magnon sidebands. The active operators for the latter transform as $L_{\pm}$ and selection rules govern the intermediate odd states, which must have well-defined L values differing from the ground state by one unit. Again, because of the equivalence of the two sublattices, the assisted transitions are not expected to show any dichroic properties at zero magnetic field. For the magnon sideband, as noted earlier, the exciton and the magnon are created on opposite sublattices and the opposing shifts produced by an applied field on the composite of the sideband result in a near cancellation, because of their quite similar g values. The field induced splitting, as registered by the MCD signal, is thus small but demonstrably observable.[6,7] Generally, the splitting of the sideband may be written as

$$\Delta=E_{+}-E_{-}=\tfrac{1}{2}[(H_{AA}-H_{BB}+2g\mu_{\mathrm{B}}H)^{2}+4H_{AB}^{2}]^{1/2}. \tag{1}$$

The terms H_{AA} and H_{BB} are the diagonal terms of the Hamiltonian that represent the energies of the ions on the individual sublattices A and B, respectively. H_{AB} represents any nondiagonal coupling existing between the sublattices, μ_{B} is the Bohr magneton, and H is the applied field. The splitting or g factor for the compound excitation is given by the difference

$$g=(g^{e}-g^{m}), \tag{2}$$

where g^e and g^m are the effective g factors for the exciton and magnon, respectively.

The term, $H_{AA}-H_{BB}$, in Eq. (1) is a measure of the inequivalence between sublattice energies and for $H_{AA}\neq H_{BB}$ it is the source of CD. The existence of nondiagonal terms may be detected by using phase-sensitive techniques. For equivalent sublattices and because of the quadratic dependences in Eq. (1), when H_{AB} is present the phase of the signal will not reverse upon reversal of the field direction. This type of field dependence is observed in CoF_2, where nondiagonal Davydov splitting exists between the ions of the rutile unit cell.[8,19]

III. EXPERIMENTAL RESULTS

The general experimental details have been described previously in a number of places;[8,20] the apparatus is essentially the same as would be required for absorption measurements in the Zeeman configuration. The sample was placed in the center of a superconducting solenoid so that the magnetic field **H** can be applied parallel or antiparallel to the c axis of the crystals. Light was passed through a monochromator and a dichroic modulator, and entered the sample with wave vector $\mathbf{k}\|c$ and parallel or antiparallel to the applied field. The dichroic modulator consisted of a fused quartz block driven by a transducer, as developed by Jasperson and Schnatterly.[21] The modulator provided alternately left and right circularly polarized light at a frequency of 50 kHz. The ac component of the transmitted radiation was detected using a phase-sensitive lock-in detector; the in-phase signal gave the difference spectrum between the two senses of polarization, that is,

$$S(\omega)=A\frac{[\alpha^{+}(\omega)-\alpha^{-}(\omega)]}{[\alpha^{+}(\omega)+\alpha^{-}(\omega)]}, \tag{3}$$

which is the CD. In Eq. (3), A is an empirically determined proportionality constant and $\alpha^{\pm}$ are the right $(+)$ and left $(-)$ circularly polarized absorption coefficients related to the up and down sublattices, respectively. Since the coefficients α are positive quantities, the polarity of the signal yields information on the ordering of the sublattice splitting relative to the direction of propagation **k**.

For simple absorption line shapes such as those encountered in pure electronic or exciton transitions, a splitting of the sublattices results in the typical dispersive S-shape signal $S(\omega)$ from which the value of Δ may be derived.[22] For broad and diffuse spectra, and when the splittings to be detected are small compared to the width of the absorption line shape, $\Gamma(\omega)$, then

$$S(\omega)=\frac{\Delta}{2}\frac{d}{d\omega}\alpha(\omega), \tag{4}$$

where

$$\alpha(\omega)=\alpha^{+}(\omega)+\alpha^{-}(\omega) \tag{5}$$

and

$$\alpha^{\pm}(\omega)=\left(\frac{2\pi}{c}\right)|\langle e|H_{\mathrm{int}}|g\rangle|^2_{\pm}\Gamma(\omega). \tag{6}$$

where c is the speed of light and H_{int} is the appropriate multipolar operator connecting the ground and excited states g and e, respectively. Substitution of Eq. (6) into Eq. (5) yields

$$S(\omega)=B\left(\frac{\Delta}{2}\right)\Bigg(|\langle e|H_{\mathrm{int}}|g\rangle|^2\frac{d\Gamma(\omega)}{d\omega}+\Gamma(\omega)[|\langle e|H_{\mathrm{int}}|g\rangle|^2_{-} - |\langle e|H_{\mathrm{int}}|g\rangle|^2_{+}]\Bigg). \tag{7}$$

In the above equation, we have assumed that the α^{+} and α^{-} for right and left circularly polarized light are approximately equal to each other. B is again an experimentally determined constant proportional to the modulated signal intensity. The signal is thus composed of a component proportional to the derivative of the line-shape function $\Gamma(\omega)$ and a residual term, which reflects changes in the transition probability induced by the applied field or other perturbations. This result agrees with the line-shape function obtained by Buckingham and Stephens[10] except for a thermally dependent population term, which has been ignored here.

A finite CD was observed at low temperatures in two of the three difluorides studied and only in compounded magnetic transitions, i.e., in sideband transitions. Signals were observed in the magnon sidebands of the ${}^6A_{1g}({}^6S)$ ground state to ${}^4T_{1g}({}^4G)$ and ${}^4A_{1g}$, ${}^4E_g({}^4G)$ absorptions of MnF_2, and of the ${}^4T_{1g}({}^4F)$ to ${}^2A_{1g}$, ${}^2T_{1g}({}^2P)$ of CoF_2; no CD signals were observable for FeF_2.

The magnitude of the CD signal $S(\omega)$ depends critically on the alignment of the crystalline c axis to the light propagation vector; the signal decreases as the degree of misalignment increases. The CD signal reverses polarity when the sample is rotated by 180° about an axis perpendicular to the c axis; this implies that the c axis is unidirectional in these materials. The application of external perturbations such as external magnetic fields and uniaxial stresses can nullify all or various portions of the CD signal.[18] The magnitude of the fields and/or stresses required to negate a signal depend on the portion of the CD spectra being considered and their direction relative to the c axis. Finally, the CD signal weakens as the temperature of the sample is increased and the component proportional to the derivative of $\Gamma(\omega)$ in Eq. (7) vanishes at higher temperatures. For example, CD becomes unobservable in MnF_2 at about 25 K for the ${}^4T_{1g}$ transition and at a somewhat higher temperature for the other transitions studied (${}^4A_{1g}$, ${}^4E_{1g}$).

Figure 1 shows the absorption spectrum of a magnon sideband of the (${}^2A_{1g}$, ${}^2T_{1g}$) transition of CoF_2 along with the observed CD in zero magnetic field. The CD is purely differential in shape (see Fig. 1) and is estimated to denote an intrinsic splitting of $\Delta_0=0.015\ \mathrm{cm}^{-1}$. The CD in CoF_2 becomes unobservable in the vicinity of 10 K. Recent results by Eremenko and co-workers generally confirm our results on the MCD in this compound, although they make no mention of the presence of CD.[23]

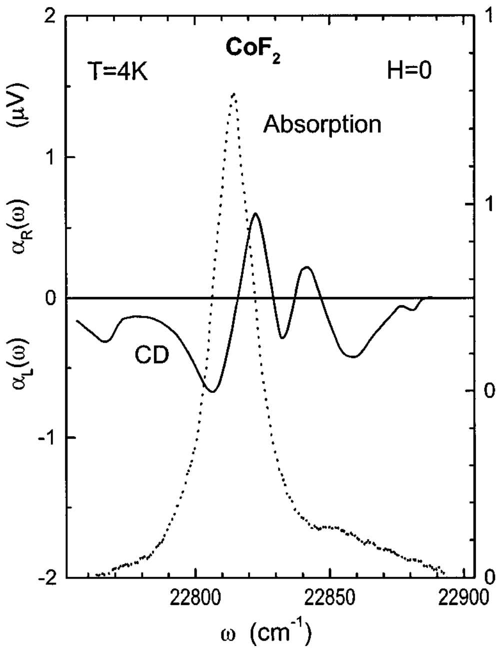

FIG. 1. The CD spectrum of the one-magnon sideband near 22 800 cm^{-1} of CoF$_2$ at 4 K and the corresponding absorption spectrum of the sideband.

Figure 2 shows the absorption spectrum of the so-called σ_1 and σ_2 sidebands accompanying the ground state to $^4T_{1g}$ transition in MnF$_2$.[1,24] These transitions are α-polarization active. A small CD signal is observed that is clearly related to these absorptions. The actual line shapes for these magnon sidebands and their frequency derivative are depicted in Fig. 3; these results are used in Fig. 4 to obtain a fit for the CD observed in this spectral region. In addition to the differential component, the results of the fitting indicate the presence of a residual contribution that could arise from the second term of Eq. (8); this term, which appears to scale as the absorption, denotes oscillator strength changes in the sideband transitions. The fit to the CD shown in Fig. 4 yields the zero-field splittings for the sidebands to be $\Delta(\sigma_1)=0.07$ cm^{-1} and

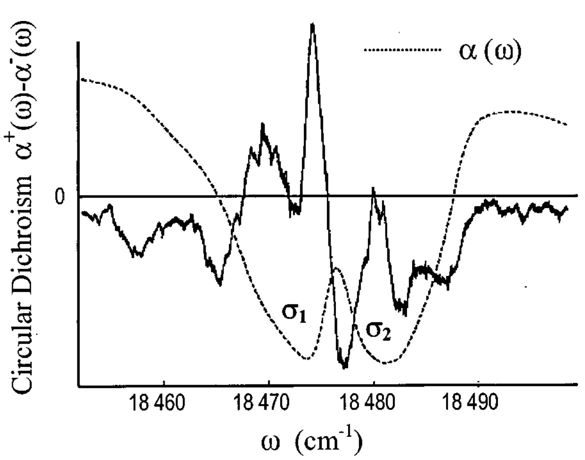

FIG. 2. The absorption and CD spectra of the so-called σ_1 and σ_2 sidebands of MnF$_2$ at 2 K; these traces connect the observed CD to the magnon sidebands (Ref. 20).

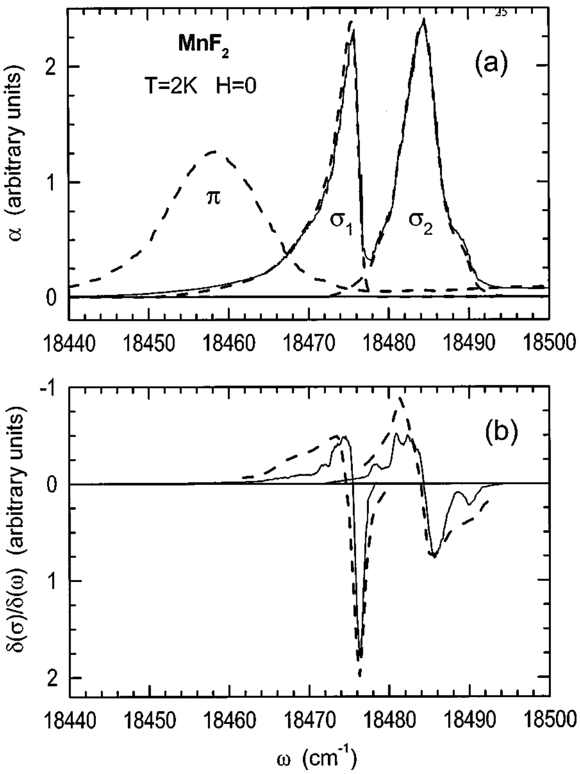

FIG. 3. (a) The experimental magnon sideband absorption spectra observed in the $^4T_{1g}$ region of MnF$_2$ at low temperature [Wilson (Ref. 20)]. Sidebands corresponding to coupling to different points of the Brillouin zone are shown in π and σ polarizations; these transitions are electric dipole in nature, hence, the σ polarized transitions also appear in α polarizations and it is these transitions that show CD and MCD. (b) Derivatives of the experimental line shapes of $\sigma_1(\omega)$ and $\sigma_2(\omega)$ are represented by solid lines; the differentials are compared to the line shapes used by Scarpace (Ref. 20) and by Wong (Ref. 20) to fit their CD and MCD results (dashed lines) calculated from the sidebands predicted theoretically in Ref. 32. Clearly, the experimentally derived traces contain greater detail.

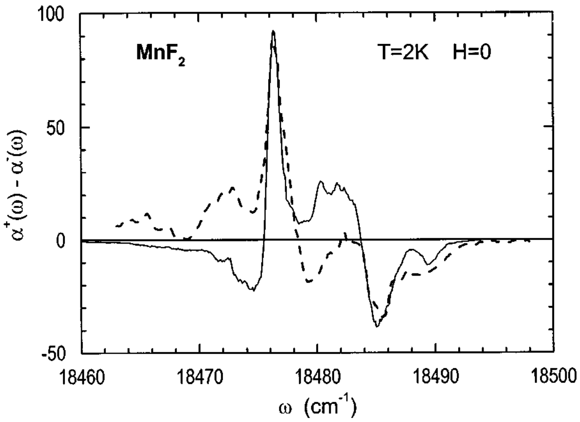

FIG. 4. Fit (solid line) to the observed (dashed line) CD in the sideband of MnF$_2$ at low temperature. Though the fit exhibits a shift in the baseline, the main features of the CD signal are well replicated.

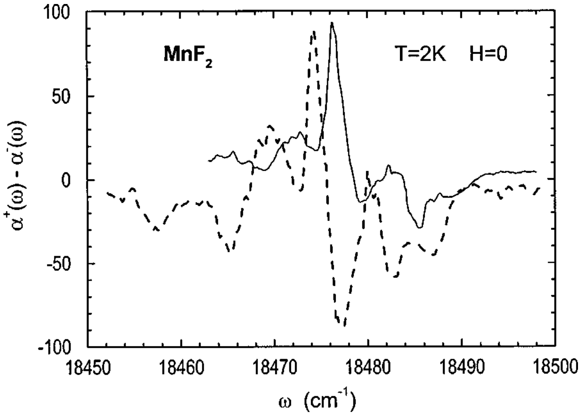

FIG. 5. The CD signal in two MnF_2 samples of different thicknesses (solid and dashed lines refer to thinner and thicker samples, respectively) [Wong (Ref. 20)]. (One spectrum has been shifted sideways for clarity.) This figure illustrates that the total absorption, $\alpha^+ + \alpha^-$, can influence the nature of the observed signal and that if the absorption is too strong, as was the case in FeF_2 sidebands where the absorption was saturated, the signal becomes too small to be observed. We no longer have all the relevant sample information concerning these traces, but in principle corrections can be made in Eq. (7) to account for the changing total absorptions.

$\Delta(\sigma_2) = -0.05\ \mathrm{cm}^{-1}$. The negative sign in $\Delta(\sigma_2)$ signifies that the sense of the splitting in this sideband is out of phase with that of σ_1. Such a change in the sense of the splitting is also observed when uniaxial stresses are applied to MnF_2 along the [110] direction in the basal plane.[25] Because of this similarity, the behavior of the CD was studied as a function of basal plane stress. The portion of the CD signal that is proportional to the splitting was found to increase linearly with stress applied along the [110] direction. A stress applied in the $[1\bar{1}0]$ direction, on the other hand, produces a "negative" or reduced splitting, which at a certain point reaches the "zero" stress CD signal. The applied stress ranged up to about 30 kg/cm^2; in this limited range, the line shape of the CD signal did not change noticeably apart from its strength. Thus, oscillator strength changes as measured by the residual signal (Fig. 4) also increase or decrease linearly with increasing stress and can actually be nulled out.[18] With these stress results and using elastic constants for MnF_2 measured by Gibbons,[26] we estimate that an intrinsic strain $\Sigma = \Delta d/d = 1.2\times 10^{-5}$ in the [110] direction exists in the basal plane when the material is in the ordered state.

FeF_2 was studied in the 21 500–28 500 cm^{-1} spectral region.[9] No CD signals were encountered in any of the α-active magnon sideband transitions investigated. With the benefit of hindsight, we now believe that we failed to observe CD in this material simply because the magnon sideband transitions studied were very strong, and necessitated the use of very thin samples. From Eq. (3), it is clear that a large denominator term comprising the total strong absorption in both polarizations would tend to conceal small splittings, as shown by the sample thickness dependence of the MnF_2 CD illustrated in Fig. 5. We estimated at the time of the experiments that our apparatus was capable of detecting splittings down to of the order of $\Delta \approx 10^{-3}$ cm^{-1} per cm of sample.

Several additional statements can be made regarding the CD signal.[18,20]

(a) The nature of the CD signal was found to be sample independent. Samples used in these measurements were obtained from diverse sources (e.g., H.J. Guggenheim at Bell Laboratories, R. Feigelson at Stanford University, and a number of commercial concerns) and the properties of the CD signal did not change from sample to sample.

(b) The CD signal strength was found to be proportional to the sample thickness whenever experimental circumstances allowed such a determination.

(c) All samples were annealed by recycling them from room temperature to several hundred degrees Celsius. No changes were observed in either the size or the nature of the signal after the temperature recycling.

(d) CD is observed in magnon-assisted transitions exclusively.

We believe that the last observation gives us a key to the source of the unexpected CD in these samples.

IV. THEORY

In the rutile structure the $a(x)$ and $b(y)$ direction lattice spacings are equal. To observe a CD signal in the absence of any lattice strain, it is required that this degeneracy be lifted. One way this can occur is through demagnetization effects arising from the dipole-dipole interactions. Ordinarily, one does not associate demagnetization effects with antiferromagnets. However, when the spin wave is excited in an antiferromagnet (either at $\mathbf{k}=0$ or at $\mathbf{k}\neq 0$) the two sublattices are canted slightly with respect to one another. This means that the sublattices exert torques on one another leading to a dipolar splitting of the spin-wave frequencies. Loudon and Pincus[17] carried out one of the first theoretical attempts to investigate how dipolar effects modify the antiferromagnetic resonance (AFMR) frequencies. Their calculation, however, was based on a molecular-field approach that neglected the affect of the Lorentz field of one sublattice upon the other sublattice. When this effect is included, the resonant frequencies are given for $\mathbf{k}=0$ by[27]

$$\omega(k=0)_{1,2} = \pm\gamma\left[H_A\left(H_A - \frac{8}{3}\pi M_s\right) + 2H_A(H_E + 4\pi M_s N_x)\right]^{1/2}, \tag{8a}$$

$$\omega(k=0)_{3,4} = \pm\gamma\left[H_A\left(H_A - \frac{8}{3}\pi M_s\right) + 2H_A(H_E + 4\pi M_s N_y)\right]^{1/2}. \tag{8b}$$

and for $\mathbf{k}\neq 0$ by

TABLE I. Magnetic parameters at low temperature for various transition-metal fluorides possessing the rutile structure. The calculated dipole-dipole induced splittings $\Delta\omega_{\mathrm{k}=0}$ and $\Delta\omega_{\mathrm{k}=\pi/2}$ are also given.

Parameter	MnF_2[a]	FeF_2[b]	CoF_2[c]
Effective spin S	5/2	2	1/2
H_{E} (T)	51.5	53.313	37.6
H_{A} (T)	0.84	19.745	9.54
g	2.00	2.25	2.80
γ (cm^{-1}/T)	0.934	1.05	1.31
M_{S} (T)	0.060	0.056	~0.06
$\Delta\omega_{\mathrm{k}=0}$ (cm^{-1})	0.016	0.065	0.083
$\Delta\omega_{\mathrm{k}=\pi/2}$ (cm^{-1})	0.02	0.08	0.12

[a] References 24, 28, and 29.

[b] References 28 and 30.

[c] References 28 and 31.

$$\omega(k)_{1,2}=\pm\gamma\left[H_A^2+2H_AH_E+H_E^2(1-\gamma_k^2)+\frac{8}{3}\pi M_s[H_A+H_E(1-\gamma_k)]\right]^{1/2}, \tag{9a}$$

$$\omega(k)_{3,4}=\pm\gamma\left[H_A^2+2H_AH_E+H_E^2(1-\gamma_k^2)+8\pi M_s\times\left(\frac{1}{3}-\sin^2\theta_k\right)[H_A+H_E(1-\gamma_k)]\right]^{1/2}, \tag{9b}$$

where $\gamma=g\mu_{\mathrm{B}}$ is the gyromagnetic ratio with g the Landé g factor and μ_{B} the Bohr magneton, H_A is the uniaxial anisotropy field, H_E is the effective exchange field, M_s is the sublattice magnetization, **N** is the demagnetization dyadic,[27] and θ_k is the angle between the easy axis z of magnetization and the direction of propagation of the spin wave **k**. γ_k is defined as $(1/n)\Sigma_{\delta}\exp(i\mathbf{k}\cdot\boldsymbol{\delta})$, where the $\boldsymbol{\delta}$'s are the vectors to the n nearest neighbors.

Notice that for the uniform spin precession ($\mathbf{k}=0$) in a sphere ($N_x=N_y=N_z=1/3$), the Lorentz field just cancels the surface demagnetizing field. Therefore, there is no shift in the resonant frequency as compared to nondipolar cases. The symmetric occurrence of N_x and N_y in Eq. (8) is due to the fact that the normal modes labeled 1, 2 and 3, 4 are characterized by net transverse magnetizations that are linearly polarized in the x and y directions, respectively. Notice also that although the 1, 2 spin-wave modes are independent of θ they are shifted by the Lorentz field.

Since the samples used in the optical studies had rectangular cross sections in the basal plane, there will be dipolar splittings of the AFMR frequencies as well as for the spin waves. Thus, from Eq. (8),

$$\Delta\omega_{k=0}=\omega_1-\omega_2=\frac{4\pi M_sH_A(N_x-N_y)}{\sqrt{2H_AH_E+H_A^2}}. \tag{10}$$

The demagnetizing factors are given by integrals over the surface.[27] In order to obtain estimates of these splittings we approximate N_x and N_y by 1/2 and 1/4, respectively, based on the sample dimensions. The parameter values used in the calculations and the resulting splittings are listed for MnF_2, CoF_2, and FeF_2 in Table I.

V. COMPARISON OF THEORY WITH EXPERIMENT

The calculated splittings given in Table I are quite small—at most a fraction of a wave number—compared with the spin-wave energies in these compounds. Given the nature of the natural linewidths, splittings of this magnitude are difficult to measure and only recently have they been resolved for the first time using Raman spectroscopy.[11,15] As noted in the Introduction, Lockwood *et al.*[11] were able to measure using a 90° scattering geometry a $\Delta\omega$ of $0.09\pm0.02\ \mathrm{cm}^{-1}$ in FeF_2 at low temperatures.

The calculated values for $\Delta\omega$ in MnF_2 and CoF_2 (see Table I) are of similar magnitude to those found experimentally for the magnon sidebands in the visible region: 0.02 cm^{-1} for CoF_2 and 0.07 and 0.05 cm^{-1} for MnF_2. Thus the origin of the observed splittings reported in the CD observations is totally encompassed by the effects of the magnetic dipole-dipole interaction in these simple rutile antiferromagnets. The size of the splittings detected by CD cannot be compared directly with those calculated in Table I, as the processes involved in the formation of magnon sidebands are not as simple and direct as those encountered in Raman scattering. For MnF_2, for example, the sideband involves a combination of an exciton on one sublattice with essentially zero dispersion[25] with a magnon on the other with nearly 50 cm^{-1} dispersion; **k** conservation requires the creation of exciton-magnon pairs with equal and opposite wave vectors from throughout the Brillouin zone. Although the splitting is mostly uniform across the Brillouin zone, the CD is, in effect, sampling the derivative of the joint density of states. It follows that an accurate calculation of the CD requires the use of Eq. (9), but the theory needs to be extended to include specific symmetry points in the Brillouin zone and an appropriate summation over the entire zone.

Conceivably, another possible cause of CD in these compounds could be a slight tilting of the spins away from their ordered positions along the c axis. However, at low temperatures and in zero applied magnetic field, all experimental results to date point to collinear spin alignment in all of these compounds.[28]

VI. CONCLUSIONS

The magnon sideband CD observed in rutile structure antiferromagnets can be quite readily explained as being the result of magnetic dipole-dipole interactions breaking the degeneracy of the two spin-wave branches. As such, these optical measurements performed three decades ago are seen now to represent the first observation of such splittings predicted by one of us (R.M.W.) in 1965. It would be informative to perform further experiments to confirm the shape dependence of the splitting by varying the dimensions of the samples.

The splitting assignable to the dipolar interaction can also be changed by the application of stress. It would be interesting to correlate the magnetic interactions with the elastic properties of the ordered lattices; undoubtedly this would lead to a better understanding of magnetorestrictive properties.

In this study we have employed the data contained in a number of University of Wisconsin theses dating back several decades; the passage of time and physical moves have reduced our capabilities to conduct additional experiments on this subject. It is our sincere wish that these results may perhaps rekindle interest in this area of magnetism.

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