Measurement of the effective-g-factor renormalization of zone-edge antiferromagnetic magnons in MnF_2†

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The temperature-dependent renormalization of the effective g factor of zone-edge magnons has been obtained through the use of magnetic-circular-dichroism-measurement techniques. The magnetic field splitting of a two-magnon optical sideband in MnF_2 has been studied between 4 and 25°K with this method. Combined with the g-factor renormalization of the associated electronic transition, these results yield the temperature dependence of the magnon g factor. Our findings are in agreement with the theoretical prediction of a $g_0\chi_\parallel/2\chi_\perp$ depression of g for antiferromagnetic magnons, independent of the magnon wave number.

I. INTRODUCTION

The technique of magnetic-circular-dichroism (MCD) measurement has proven to be a valuable tool for the study of weak-optical-absorption features in magnetic insulators.[1] We have used this method to measure the temperature-dependent renormalization of the effective g factor for zone-edge magnons in the antiferromagnet MnF_2. Antiferromagnetic resonance has also been used to observe magnon g-factor renormalization[2,3]; however, this technique only permits the study of $k=0$ magnons. Exciton-magnon optical sidebands, on the other hand, are dominated by zone-edge magnons, for which the magnon energy is comparable to the exchange energy. The study of these sidebands has already yielded the magnon energy renormalization at finite temperatures.[4] A detailed study of the splitting of such an absorption line in an applied magnetic field can also furnish the effective-g-factor renormalization for $k\neq 0$ magnons. To our knowledge, this experiment represents the first such measurement.

We have studied the purely excitonic transitions at 18 418 and 18 435 cm^{-1} and their associated magnon sidebands within the ${}^6A_{1g}\to{}^4T_{1g}$ absorption manifold in MnF_2.[5] The magnetic-field splitting of the one-magnon sidebands does not yield the magnon g-factor directly, but rather, the difference between the magnon and the exciton g factors.[6] If g^{mag} and g^{exc} have nearly the same value, the observed splitting will be difficult to detect. In previous work, the magnetic field dependence of the one-magnon sidebands was studied in pulsed fields to 150 kOe with no visible splitting.[7]

This situation will be altered for a two-magnon sideband, such as the one identified at 18 533 cm^{-1} in MnF_2.[8] From its energy, effective g factor, and electric dipole nature, it has been assigned as a two-magnon sideband of the purely excitonic transition $E2$ at 18 435 cm^{-1}, with the two magnons created on one sublattice and the exciton on the other. In this case, the g factor that we measure will be $g^{\text{obs}}=2g^{\text{mag}}-g^{\text{exc}}$; so even if the magnon and exciton g factors have nearly the same value the observed splitting can be quite large, and its change as a function of temperature can be accurately determined. Since $\Delta g^{\text{obs}}(T)=2\Delta g^{\text{mag}}(T)-\Delta g^{\text{exc}}(T)$, we need to measure the renormalization of the exciton g factor as well in order to obtain

$$\Delta g^{\text{mag}}=\tfrac{1}{2}(\Delta g^{\text{exc}}+\Delta g^{\text{obs}}). \qquad (1)$$

II. THEORETICAL REVIEW

The predicted behavior of g^{mag} as a function of temperature is discussed by Saslow.[9] He starts with an expression for the energy of a sublattice magnon at finite temperature in an applied field,

$$\omega_{\vec{k}}^{\alpha,\beta}(H,T)=\omega_{\vec{k}}(0,0)\left(1-\frac{1}{NS}\sum_{\vec{q}}\frac{\omega_{\vec{q}}}{2JzS}(n_{\vec{q}}^{\alpha}+n_{\vec{q}}^{\beta})\right)\pm\left(g_0\mu H+\frac{2Jz}{N}\sum_{\vec{q}}(n_{\vec{q}}^{\alpha}-n_{\vec{q}}^{\beta})\right), \qquad (2)$$

where J is the intersublattice exchange integral, z is the number of nearest neighbors on the opposite sublattice, $n_{\vec{q}}$ is the magnon-occupation number, α and β refer to the different sublattices, N is the number of spins in the crystal, $\omega_{\vec{k}}(0,0)=2JzS(1-\gamma_{\vec{q}}^2)^{1/2}$, and $\gamma_{\vec{q}}=(1/z)\sum_{\vec{\delta}}e^{i\vec{q}\cdot\vec{\delta}}$. The first bracket in Eq. (2) is the energy renormalization in zero-external field $\omega_{\vec{k}}(0,T)$, and the second bracket includes applied-field effects.

To obtain the g-factor renormalization, we insert the expression for occupation number

$$n_{\vec{q}}=(e^{\beta\omega_{\vec{q}}(H,T)}-1)^{-1}, \qquad (3)$$

keeping terms to first order in H, and expand Eq. (2) in a Taylor series about $H=0$ to obtain

$$\omega_{\vec{k}}(H,T)=\omega_{\vec{k}}(0,T)+(g_0^{\text{mag}}+\Delta g^{\text{mag}})\mu H+O(H^2). \qquad (4)$$

This leads to the result

$$\frac{\Delta g^{\mathrm{mag}}}{g_0^{\mathrm{mag}}} = \frac{-4Jz}{Nk_BT} \sum_{\vec{q}} n_{\vec{q}}(1+n_{\vec{q}}) , \qquad (5)$$

where $n_{\vec{q}}$ is now the occupation number in zero-external field (which is no longer sublattice dependent).

From the elementary theory of antiferromagnets, we may write an expression for the net magnetization parallel to the applied field (along the c axis),

$$M_{\parallel} = g_0 \mu \sum_{\vec{q}} (n_{\vec{q}}^{\alpha} - n_{\vec{q}}^{\beta}) . \qquad (6)$$

The low-field parallel susceptibility, defined as $(\partial M_{\parallel}/\partial H)_{H=0}$, then becomes

$$\chi_{\parallel} = \frac{2g_0^2\mu^2}{kT} \sum_{\vec{q}} n_{\vec{q}}(n_{\vec{q}}+1). \qquad (7)$$

Using also the expression for perpendicular susceptibility,

$$\chi_{\perp} = Ng_0^2\mu^2/4Jz , \qquad (8)$$

we then find

$$\Delta g^{\mathrm{mag}}/g_0^{\mathrm{mag}} = -\tfrac{1}{2}\chi_{\parallel}/\chi_{\perp} . \qquad (9)$$

This expression is independent of $\vec{k}$; that is, the g-factor renormalization is predicted to remain constant throughout the Brillouin zone.

One point needs to be noted here. Equation (2) is only an approximation, as it does not include the effects of anisotropy and intrasublattice exchange, which result in the observed $\vec{k}$ dependence of the *energy* renormalization in zero field.[10] These effects contribute additional terms to $\omega_k(0, T)$, but influence $\Delta g^{\mathrm{mag}}/g^{\mathrm{mag}}$ only through the temperature dependence of $\chi_{\parallel}/\chi_{\perp}$. Thus they are included through the use of empirical values for $\chi_{\parallel}$ and $\chi_{\perp}$.

Subsequently, Passow *et al.*[11] discussed the g-factor renormalization for a purely electronic transition, in their case, the $E1$ exciton line in MnF_2. They arrive at the expression

$$\Delta(T) = (4zH/Ng_0\mu)[J_iS - J_f(S-1)]\,\chi_{\parallel}(T), \qquad (10)$$

where $\Delta(T)$ is the change in energy separation of the peaks in the applied field H as a function of temperature, J_i and J_f are the interlattice exchange integrals for the ground and excited state, respectively, and S is the ground-state spin. We note, however, that using Eq. (8) and the definition of g, this expression can be reduced to the form

$$\frac{\Delta g^{\mathrm{exc}}}{g_0^{\mathrm{exc}}} = -\frac{1}{2}\,\frac{\chi_{\parallel}}{\chi_{\perp}}\left(S - \frac{J_f}{J_i}(S-1)\right) , \qquad (11)$$

differing from Eq. (9) only because the excited atom experiences an exchange field different from that experienced by the ground-state atoms. In Ref. 11, the change in splitting at 25 kOe was plotted against $\chi_{\parallel}$, obtained from the susceptibility data of Trapp and Stout.[12] A good fit was obtained with $J_f/J_i \approx 1.3$.

In the same paper, the differential broadening of the split $E1$ lines was also measured, with the higher-energy peak exhibiting a slightly greater temperature broadening. This effect was attributed to Raman scattering of magnons by nearest neighbors on the opposite sublattice with slightly different sublattice magnon populations induced by the applied field.

III. EXPERIMENTAL DETAILS

The MCD apparatus that we have used has been described in detail elsewhere[8,13] and will only be summarized. The sample sits within the evacuated bore of a superconducting solenoid with the magnetic field applied parallel or antiparallel to the c axis of the crystal. Light from a tungsten source is passed through a 1-m Czerny-Turner scanning monochromator and then through a photoelastic modulator which alternately produces right- and left-circularly-polarized light at 50 kHz.[14] After the modulated beam passes through the sample, it is detected by a photomultiplier tube (EMI 9558), the output of which is monitored by a lock-in detector (PAR HR-8). The signal in phase with the 50 kHz reference frequency yields the difference spectrum between the two senses of polarization, which is proportional to the MCD. The output of the lock-in amplifier is plotted on a chart recorder as a function of wavelength for different temperatures.

The sample temperature is measured and regulated using a capacitance thermometer and an ac capacitance bridge patterned after Griffin.[15] At the start of each run, the capacitance thermometer, which is unaffected by high magnetic fields but recycles poorly, is calibrated in zero field against a germanium thermometer, also in contact with the sample.

Our MCD spectra consist of the differential absorption between left- and right-circularly polarized light of two closely spaced Zeeman lines. The resulting traces appear as the difference between two overlapping absorption curves. For a splitting significantly smaller than the linewidths, one obtains the typical S-shaped signal. The true separation of the peaks may be obtained only through a knowledge of both the apparent (measured) splitting and the line-shape function. For weak and noisy signals, considerable error is frequently introduced in the process of data reduction.

If the applied magnetic field is strong enough that the separation is larger than the linewidths, the two curves will overlap only slightly, and thus one can directly measure the actual splitting. These two situations are depicted in Fig. 1. For small separation, the signal amplitude is propor-

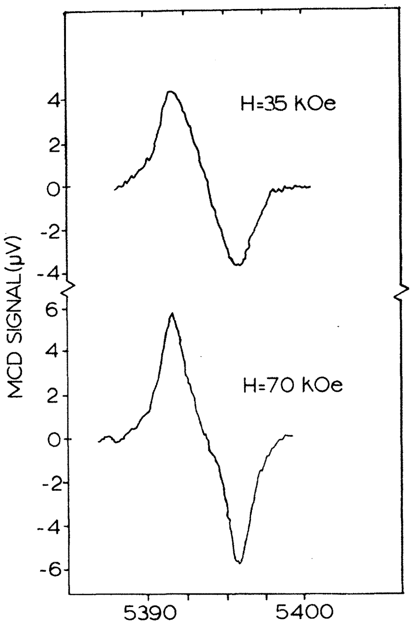

FIG. 1. MCD spectra of the two-magnon sideband for different field strengths. Upper curve, 35 kOe, small-peak separation. Lower curve, 70 kOe, peaks resolved.

tional to H,[16] approaching its intrinsic value as H increases and the lines separate. We obtained our MCD spectra in an applied field of 70 kOe. In this high field, the two peaks corresponding to the two-magnon sidebands on opposite sublattices were completely resolved, and therefore the g factor was directly proportional to the measured peak separation; no fit to a normalized line shape was necessary, and consequently our experimental uncertainty was significantly smaller than the predicted effect.

The exciton absorption lines $E1$ and $E2$, magnetic

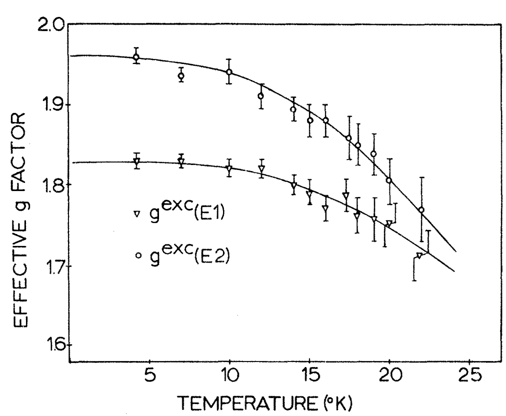

FIG. 2. Temperature dependence of the effective g factors for the $E1$ and $E2$ exciton lines in MnF_2 at 18 418 and 18 435 cm^{-1}, respectively. Solid lines represent the best fit with Eq. (11) and susceptibility data from Ref. 12 (see text).

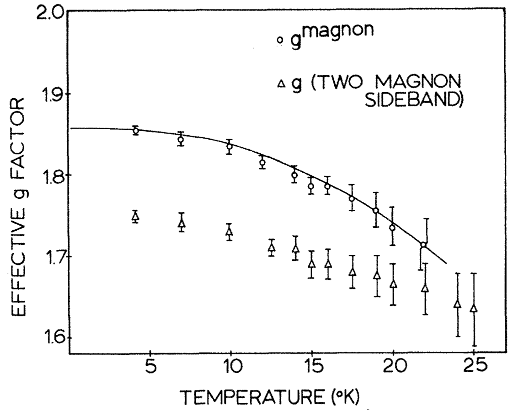

FIG. 3. Temperature dependence of the effective g factor for the two-magnon sideband in MnF_2 at 18 533 cm^{-1} and the calculated magnon g factor. Solid line is $g_0^{mag}(1-\chi_{\parallel}/2\chi_{\perp})$, using susceptibility data from Ref. 12.

dipole transitions,[6] show up only under σ polarization, whereas with MCD we measure the axial spectrum. However, with the use of two small right-angle prisms affixed to the sample, we were able to measure the σ absorption spectrum within the bore of the superconducting magnet and thereby obtain directly the temperature-dependent renormalization of the $E2$ effective g factor which appears in Eq. (1).

IV. RESULTS AND DISCUSSION

We present the results of our measurements in Figs. 2 and 3. In Fig. 2, we have plotted $g(T)$ at 66 kOe for both $E1$ and $E2$. (Data taken at 35 kOe were found to be in agreement with the above values.) Again, by comparison with the Trapp and Stout data, we find that for $E1$ $J_f/J_i = 1.2 \pm 0.05$, and for $E2$, $J_f'/J_i = 0.88 \pm 0.04$. The different values of J_f are attributed to differences in the wave functions of the $E1$ and $E2$ excitons. We also observed a differential broadening of the two $E2$ lines, with the higher-energy peak exhibiting the greater broadening, thereby agreeing qualitatively with the model developed in Ref. 11. However, since $E2$ is a weaker line which washes out at 22 °K and overlaps the tail of a one-magnon sideband, a quantitative measurement of this effect would have been subject to large uncertainties and therefore was not undertaken.

The lower points in Fig. 3 show the measured values of g^{obs} for the two-magnon sideband, and the upper points show the values of g^{mag} calculated from Eq. (1). We see that our results are in very good agreement with the predicted value $g_0^{mag} \times (1-\chi_{\parallel}/2\chi_{\perp})$, the solid line of Fig. 3. Also, comparison with the data of Refs. 2 and 3 indicates that the g factors of the zone-boundary and zone-center magnons are renormalized by the same factor, in agreement with Ref. 9.

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