Origin of Circular Dichroism in Simple Antiferromagnets

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In the 1970s, we studied the magnetic circular dichroic (MCD) properties of a number of rutile antiferromagnets (AFMs) and demonstrated that such techniques provided a very sensitive tool with which to probe the spectra and to provide complementary data to absorption spectra or to far-infrared and Raman spectra of these materials [1–3]. We also reported at that time the observation of circular dichroism (CD) in certain magnon sidebands of MnF_2 [1] and CoF_2 [2]. CD occurs in the absence of an applied magnetic field and corresponds to zero-field MCD though the origins of the dichroism may arise from different sources. 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 the unexpected observation.

For simple AFMs in their ordered state, the ground state of each sublattice is non-degenerate and is identical to the other except for their sense of magnetization with respect to the unique crystalline axis. If the state for the "up" sublattice is given by |L,M> then the corresponding "down" sublattice is represented by |L,–M>. 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 polarizations, respectively. If ions in one of the sublattices have a non-vanishing 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 a magnetic field, the sublattices are degenerate in energy and no circular dichroism will be observed. With an applied field, this energy degeneracy is lifted and the transition will manifest MCD. Since the two components of the light are 180° out of phase, phase sensitive detection techniques can provide the sense of the splitting, i.e., the ordering 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 experimental 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 **H** field could 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 $\mathbf{k} \parallel c$ and parallel or antiparallel to the applied field. The dichroic modulator consisted of a fused quartz block driven by a transducer. The modulator provided alternatively 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

$$S(\omega) = A\frac{[\alpha^{+}(\omega) - \alpha^{-}(\omega)]}{[\alpha^{+}(\omega) + \alpha^{-}(\omega)]} \qquad (1)$$

which is the CD or MCD signal. In Eq. (1), A is an empirically determined proportionality constant and $\alpha^{\pm}$ are the right (+) and left (-) circular polarized absorption coefficients related to the "up" and "down" sublattices, respectively. Since the coefficients, α's, are positive quantities, the polarity of the signal yields information on the ordering of the sublattice splitting. Figure 1 shows the absorption spectrum of a magnon sideband of the (^4A, $^4T_1$) transition of CoF_2 along with the observed CD and MCD in a 20 KOe applied H field. Though the CD may not be purely differential in shape, the observed weak CD corresponds to an additive intrinsic splitting of the order of $\Delta_0 = 0.02$ cm^{-1}. The absorption spectrum of the so-called σ_1 and σ_2 magnon sidebands accompanying the ground state to $^4T_{1g}$ transition in MnF_2 are α-polarization active. A small CD signal is observed that is clearly related to this transition. A fit for the CD observed in this spectral region yields the zero field splittings for the sidebands to be $\Delta(\sigma_1) = 0.07$ cm^{-1} and $\Delta(\sigma_2) = -0.05$ 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.

These unexpected CD results in MnF_2 and CoF_2 can now be explained from the fact that a recent CD study of the one-magnon Raman scattering in FeF_2 [4] has revealed directly for the first time the presence of a zero field splitting of the AFM sublattices of 0.09 cm^{-1}. Such a splitting was predicted previously through the presence of magnetic dipole-dipole interactions [5] in rutile structure AFMs. Although the strength of these interactions is poorly known, an approximate calculation based on the theory of Ref. 5 gives a splitting of about 0.03 cm^{-1} for MnF_2, in good agreement with the observed result. Thus the CD observed for magnon sidebands of these AFMs is mainly due to dipolar interactions.

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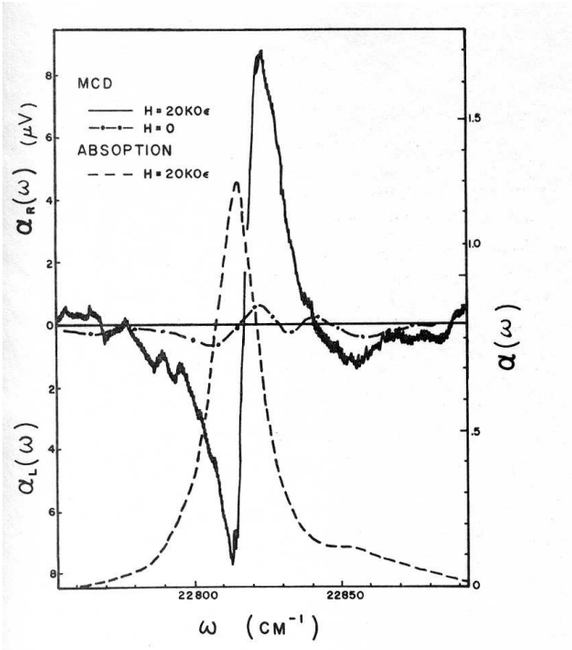

Fig. 1. Optical absorption, CD and MCD of CoF_2 at 4 K.