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OPTICAL PROPERTIES AND SPIN DYNAMICS OF FeCl_2

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Résumé. — Le spectre d'absorption optique de FeCl_2 a été mesuré à des températures comprises entre 1,2 et 300 °K et dans le domaine d'énergie 0,6-3 eV. Les bandes d'absorption observées sont faibles et ont un comportement anormal en température en dessous de la température de Néel ($T_N = 23,5$ °K). Pour expliquer ces variations anormales, le modèle faisant intervenir le couplage spin-exciton est proposé. D'après ce modèle, des informations sur la fonction de corrélation de spin à différentes températures entre plus proches voisins peuvent s'obtenir très simplement à partir des résultats d'absorption optique à ces mêmes températures. Les résultats présentés sont comparés aux différents modèles dans les divers domaines de température.

Abstract. — The optical absorption spectrum of FeCl_2 is reported in the energy range 0.6-3 eV and for temperatures between 1.2 and 300 °K. The observed absorption bands are weak and some have an anomalous temperature dependence below T_N , Néel temperature (23.5 °K). A model is proposed to explain the anomalous temperature dependence involving spin-exciton coupling. According to the model information about the nearest-neighbor spin correlation function can be obtained simply from optical absorption data at all temperatures. Results are presented and compared with various models in the different temperature regions.

FeCl_2 has a layer structure similar to MoS_2 in which a hexagonal layer of Fe^{++} ions is sandwiched between two hexagonal layers of Cl ions, the three being bound together by ionic and covalent forces. The binding between one such triplet and the next is much weaker, probably being mostly Van der Waals. The result is a soft hygroscopic crystal which cleaves (or scrapes) easily, looks muddy brown in color, and is antiferromagnetic with a Neel temperature of about 23.5 °K. The magnetic ordering is such that all the Fe^{++} spins in one layer are parallel, and alternate layers are antiparallel. Thus although FeCl_2 is antiferromagnetic, the predominant interaction within one plane is ferromagnetic. We shall see that this has some striking consequences for the optical behavior of the crystal at low temperature.

The Fe^{++} ions are located at centers of inversion and are octahedrally coordinated with the nearest neighbor Cl^- ions. The crystal field acting on the Fe^{++} ions is nearly cubic with approximately a one percent trigonal distortion [1].

The optical spectrum of Fe^{++} [2] as well as various ground state properties [3] have been used as an example of the dynamic Jahn-Teller effect. The lowest observed optical transition is from the ${}^5T_{2g}$ ground state to the 5E_g state split by the cubic crystal field parameter $10 Dq \approx 8\,600 \text{ cm}^{-1}$. The absorption

band has a characteristic double peaked shape expected for a transition to a two-fold degenerate state coupled to lattice vibrations [4].

In this paper we present measurements of the optical spectrum of FeCl_2 in the energy range 0.5 eV-3 eV, and for temperatures varying between 1.2 °K and room temperature. Section I presents the results and describes the general features of the absorption bands which were observed. Section II presents a model which describes the anomalous low temperature variation of the strength and shape of some of the bands. Section III presents results of circular dichroism measurements made on the absorption bands at low temperature.

The optical absorption measurements were made using a Cary model 14 spectrophotometer. The sample was surrounded by gas or liquid coolant during the measurements. The temperature was varied by partially transferring He to the dewar and allowing the temperature to slowly rise during the experiment. The temperature sensor used was a Au : Fe-chromel thermocouple.

The magnetic circular dichroism measurements were made using a small superconducting solenoid to produce fields up to 30 kG. A circular polarizer was placed in both the sample and reference channel of the Cary 14 and the absorption measured for field parallel and anti-parallel to the optical beam.

The samples used were grown by M^{me} R. Saint-James at the Physique des Solides department of CEA Saclay. Although the impurity content is not known,

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it is estimated to be approximately 0.1 at %. The optical data reported here were independent of sample with the exception of the shape of the IR peak which showed some variable structure.

I. — Figure 1 shows the observed absorption spectrum for four temperatures. The band at 0.9 eV has

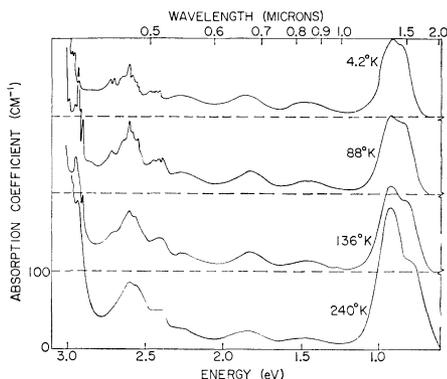


FIG. 1. — Absorption coefficient versus energy for FeCl_2 measured at four different temperatures.

structure similar to that reported previously [2] with the separation between the two main peaks apparently increasing with temperature — a characteristic of the dynamic Jahn-Teller effect. In addition, however, the strengths of the two peaks vary differently with temperature. This transition, being spin allowed but parity forbidden, is weakly allowed by virtue of the coupling of the T_2 and E_g states to odd-parity electronic states caused by odd-parity phonons. The different variation in strength of the two peaks with temperature is quite puzzling since it implies different phonons are coupled to the states giving rise to the two peaks, and yet according to the Jahn-Teller interpretation these states have the same symmetry and hence must couple with equal strength to other electronic states. Further work must be done to unravel this mystery. One possible explanation is that impurities are in some way responsible.

At higher energy several absorption bands appear, all of which are temperature-independent in strength for temperatures above T_N . This indicates no phonons are needed for the transitions to take place. Their weakness however, indicates that these transitions are not completely allowed. Undoubtedly the transitions are spin-forbidden but parity allowed. Table I shows the peak positions and oscillator strengths of the major lines observed.

The strength of spin-forbidden electric dipole transitions made allowed by spin-orbit mixing can be roughly estimated by

$$f_{\text{sf}} = f_{\text{sa}} \cdot \left(\frac{\lambda}{\Delta E} \right)^2$$

where f_{sf} is the oscillator strength of the spin-forbidden

TABLE I

Oscillation Strengths of Absorption Bands in FeCl_2

The oscillator strengths were determined using

$f = 6.57 \times 10^{-11} \int \alpha(\nu) d\nu$. This assumes the index of refraction is 1.5 and no local field correction.

$E(\text{eV})$	$10^6 f$
0.9	10.3
1.48	2.6
1.85	3.4
2.27	1.6
2.45	1.0
2.6	2.3
2.90	1.09
2.93	3

transition f_{sa} is the oscillator strength for transitions to a different state of the same spin, λ is the spin-orbit mixing parameter which couples the two states and ΔE is their energy separation. Reasonable guesses for these numbers for Fe^{++} are ; $f_{\text{sa}} \sim 0.1$, $\lambda = .01 \text{ eV}$, $\Delta E = 3 \text{ eV}$. This combination of numbers produces $f_{\text{sf}} \simeq 10^{-6}$ which is the observed order of magnitude.

The observed energy spectrum of states should be compared with the crystal field calculations of Sugano and Tanabe [5]. Their calculated spectrum, when adjusted (by inserting the Racah parameters appropriate for Fe^{++}) shows that there are several odd parity states having different spin than the ground state in the energy range of the observed absorption bands, but the positions of these states do not agree well at all with the observed spectrum. Further work on this problem using adjusted Racah parameters and perhaps higher term states as well will be necessary to achieve an identification of the observed absorption bands. We will not pursue that problem here but will describe the observed properties of the absorption bands which will aid in an identification when such a calculation is carried out.

There are two systematic variations of the observed bands with energy. First the phonon coupling strength of the excited states giving rise to the bands decreases with increasing energy. The three bands between 1.5 and 2.3 eV are broad and structureless, while all the higher energy absorption consists of narrow line groups. In addition, the three lower energy bands show no temperature dependence at all, while all the higher bands decrease in strength strongly below T_N . The group around 2.6 eV decreases to around 70 % of its high temperature value, and the narrow line at 2.9 eV disappears completely at low temperatures. These temperature dependences will be discussed in detail in the next section.

The 2.9 eV line broadens and disappears as the temperature is raised towards room temperature. A

similar behavior is found for a variety of zero phonon lines and associated vibrational structure in the spectra of electronic states weakly coupled to lattice vibrations [6].

II. — A spin-forbidden transition can be made stronger either by the spin-orbit mixing mechanism described in the last section or by virtue of exciton-magnon coupling as first described by Sell, Greene and White [7]. In this mechanism the spin change on the ion where the transition takes place is compensated by an opposite spin flip on a neighboring ion so the total spin in the excited state is the same as in the ground state. The strength of the absorption depends on the strength of the exciton-spin fluctuation coupling for neighboring ions.

In the case of FeCl₂ all the nearest-neighbor Fe⁺⁺ ions have parallel spins at zero temperature with maximal values of m_j , so opposite spin flips on nearest neighbor sites are impossible. Only when two neighboring ions do not have parallel spins with extremal m_j value can two simultaneous spin flips in opposite directions occur, enhancing the optical absorption. Thus for FeCl₂ the oscillator strength of the exciton-spin fluctuation optical absorption increases with the disorder in the spin system rather than the order, as for MnF₂ where nearest neighbor spins are antiparallel at low temperatures.

To be more precise, let the spin of an ion be J , the spin projection quantum number along the C axis be m_j , and the probability of an ion occupying state m_j be $P(m_j)$. Then the observed strength of an absorption line due to this mechanism is proportional to

$$A(T) \propto 1 - K \sum_{ij} (P_i(-J) P_j(-J) + P_i(J) P_j(J)) \quad (1)$$

where j and i label the positions of two ions, assumed henceforth to be nearest neighbors. The area of the absorption line is thus a measure of the nearest neighbor spin correlation function.

Eq. (1) can be readily evaluated in the high and low temperature limits as well as for all temperatures in the molecular field model. At zero temperature $P(-J)$ for all ions approaches unity and the strength is zero. At small but finite temperatures,

$$P(-J) \simeq 1 - P(-J + 1)$$

$$P(J) \simeq 0$$

Using the magnon model, the area of the line is approximately given by

$$A(T) = |c|^2 \sum_k \frac{k}{\partial E(k)} |S_k|^2 \exp\left(-\frac{E(k)}{kT}\right) \quad (2)$$

where $E(k)$ is the magnon energy as a function of wave vector, $S_k = \sum_j \sin \mathbf{k} \cdot \mathbf{a}_j$ where \mathbf{a}_j are the vectors

connecting nearest neighbor ions and the density of states factor

$$\frac{k}{\partial E(k)} \frac{\partial E(k)}{\partial k}$$

is appropriate for two dimensions. Using

$$E(k) = A(B - \gamma_k^2)^{1/2} \quad \text{where} \quad \gamma_k = \frac{1}{n} \sum_a e^{i\mathbf{k} \cdot \mathbf{a}}$$

(n = number of nearest neighbors) and assuming small values of $k \cdot a$,

$$A(T) = CT^2 \exp\left(-\frac{E_0}{kT}\right) \quad (3)$$

where E_0 is the lowest excitation frequency for magnons, known from antiferromagnetic resonance measurements to be 16.4 cm^{-1} [8].

Figure 2 shows a series of measurements of the

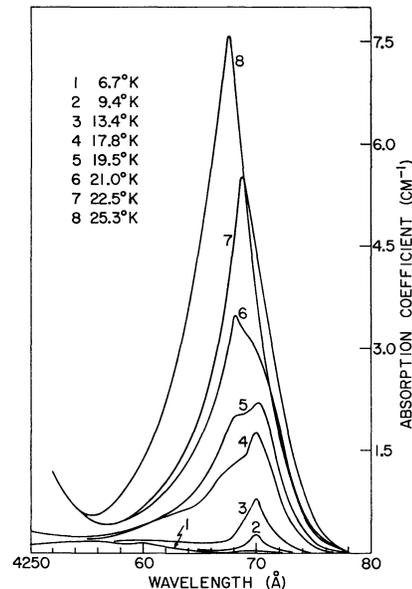


FIG. 2. — Optical density of the 4268 Å band at various temperatures.

narrow line at 4268 Å which disappears completely at low temperature ; thus essentially all of its strength is proportional to the spin fluctuation coupling. At the lowest temperatures a single narrow line grows rapidly with temperature. Figure 3 shows a test of eq (3). The straight line has a slope giving $E_0 = 16 \pm 2 \text{ cm}^{-1}$ in good agreement with the result of Jacobs *et al.*

At high temperature $A(T)$ should become constant since $P(m_j) = 1/(2J + 1)$ for all m_j , independent of temperature. Figure 4 shows a plot of $A(T)$ (in arbitrary units) as a function of temperature showing $A(T)$ does indeed become constant for temperatures above about $1.6 T_N$.

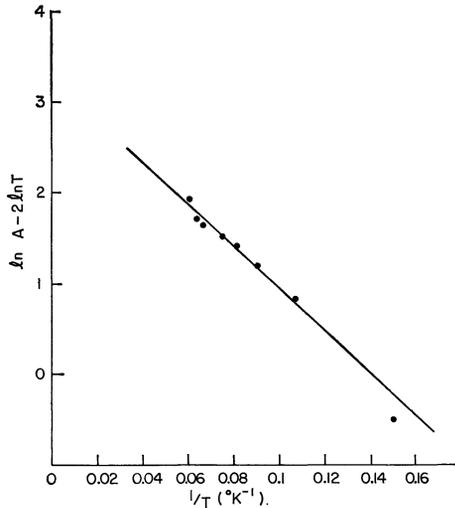


FIG. 3. — Graph of $(\ln A - 2 \ln T)$ vs $1/T$. The straight line corresponds to a magnon gap of 16 cm^{-1} .

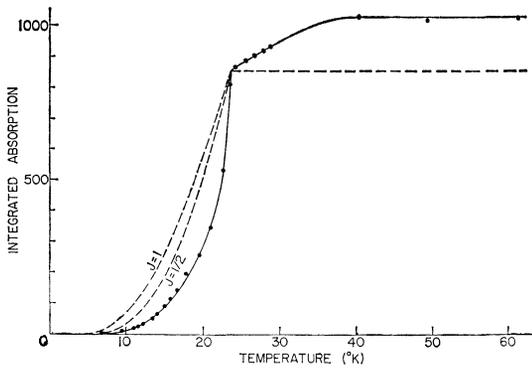


FIG. 4. — Solid line drawn through circles : area of the 4268 \AA band vs. temperature. Dashed lines : molecular field calculation of the same quantity for $J = 1$ and $J = \frac{1}{2}$.

The classical molecular field theory can be used to evaluate (1) and we present the results here for purposes of illustration. We assume a molecular field H_m acts on each spin in one sub-lattice and in the absence of external fields $-H_m$ acts on the spins in the other sub-lattice.

Then

$$P(m_j) = \frac{\exp\left[-\frac{g\mu_B m_j H_m}{kT}\right]}{\sum_{-J}^J \exp\left[-\frac{g\mu_B m_j H_m}{kT}\right]} \equiv \frac{e^{-m_j x}}{\sum_{-J}^J e^{-m_j x}} \quad (4)$$

where

$$x = \frac{g\mu_B H_m}{kT}.$$

The function $x(T)$ can be obtained by solving simultaneously the two eq [9]

$$\frac{M(T)}{M(0)} = B_J(Jx) \quad (5)$$

$$\frac{M(T)}{M(0)} = J \frac{J+1}{3J} \left(\frac{T}{T_N}\right) x \quad (6)$$

where $B_J(x)$ is the Brillouin function.

The Fe^{++} ground state in FeCl_2 is fairly complicated due to the fact that the trigonal crystal field interaction is of the same order of magnitude as the spin-orbit interaction within the ground state manifold [1]. For zero trigonal field the ground state has $J = 1$, and for small trigonal field and even smaller spin-orbit parameter the ground state is a doublet which can be characterized by $J = \frac{1}{2}$.

According to Carrara, the spin orbit and trigonal interactions are about equal and the splitting between the ground state doublet and the low lying singlet, which together make up the $J = 1$ ground state in the absence of a trigonal interaction, is about 8 cm^{-1} . Thus we expect the effective spin to be $\frac{1}{2}$ for lowest temperature and 1 near T_N due to the thermal population of the low lying singlet. Both $J = \frac{1}{2}$ and $J = 1$ will be used for comparison purposes.

For $J = 1$ straight forward evaluation of (4) yields

$$\frac{A(T)}{A(T_N)} = \frac{9}{7} \frac{(2 \cosh x + 1)^2 - 2 \cosh 2x}{(2 \cosh x + 1)^2} \quad (7)$$

and for $J = \frac{1}{2}$

$$\frac{A(T)}{A(T_N)} = \frac{2}{1 + \cosh x} \quad (8)$$

By combining these results with equations (5) and (6) for the appropriate J values the curves shown in figure 4 were obtained. The deviation between the molecular field calculation and the measured result is greater for this experiment than for a measurement of most macroscopic quantities such as magnetization, or neutron scattering intensities, since short range correlations are involved. The molecular field theory misses completely the residual correlations above T_N and does not describe the shape below T_N very well.

The critical behavior of the nearest neighbor spin correlation function near T_c can be directly measured both above and below T_c providing among other things a check on the symmetry implied by scaling [10].

Assuming that the spin correlation function is independent of spin projection number, we have

$$1 - \frac{A(T)}{A(T_\infty)} = C \langle S_0 S_R \rangle$$

where A = the area of the absorption band. Thus optical absorption measurements can give direct information about the temperature dependence of $\langle S_0 S_R \rangle$ both above and below T_N . The temperature resolution employed in this experiment was not fine enough to provide a determination of this behavior for sufficiently small values of $(T_c - T)/T_c$. Further work in this direction is under way.

It is clear from figure 2 that the shape of the 4 268 Å line is complex and changes with temperature. Again the low and high temperature results can be readily obtained but nothing whatever can be predicted about the intermediate range.

At low temperature the one magnon model described above predicts for the optical absorption coefficient

$$\begin{aligned} \alpha(w) &= (w - w_0) e^{-\hbar w/kT} \quad w > w_0 \\ \alpha(w) &= 0 \quad w < w_0. \end{aligned} \quad (9)$$

Where w is the frequency of the light measured downwards from the magnetic dipole transition energy. This equation is not obeyed well at all. The observed low temperature peak is quite symmetric while the shape implied by eq (9) is not. This probably indicates that the exciton-magnon interaction alters the magnon energies appreciably, an effect ignored in eq (9). Other effects which will change the line shape at intermediate temperatures are magnon-dispersion, which may differ from the above approximation, and frequency renormalization, which is ignored in deriving eq (9).

At high temperatures when the spins have randomly chosen values of m_j , the shape can again be evaluated. Let the exchange energy of ion 1 due to nearest neighbors labeled by i be

$$E_{ex} = \sum C_i = C m_{J1} \sum_i m_{Ji}. \quad (10)$$

Then the transition energy for a purely magnetic dipole transition is

$$E = E_0 + C \Delta m_{J1} \sum_i m_{Ji}. \quad (11)$$

The energy needed to change the nearest neighbor spin an amount Δm_{J2} is

$$\Delta E_{ex2} = \Delta m_{J2} C \sum_j m_{Jj} \quad (12)$$

where j labels the nearest neighbors of ion 2 which itself is a nearest neighbor to ion 1. The total energy for the transition then is, using $\Delta m_{J1} = -\Delta m_{J2}$;

$$E = E_0 + C \left(\sum_i m_{Ji} - \sum_j m_{Jj} \right). \quad (13)$$

Each ion has six nearest neighbors, two of which are in common and so cancel. The four remaining can each have $(2J + 1)$ values of m_j so there are $(2J + 1)^4$ possible environments for each ion. Since the distribution of the m_j 's is independent of the sign of m_j this is the same problem as the calculation of the distribution for a single ion. It is well known from the F -center spin resonance line shape problem that the shape of the resulting line approaches a Gaussian in the limit of large numbers of possible environments [11]. Figure 5 shows a test of this result. As the temperature increases the lineshape approaches a Gaussian, while

at lower temperatures the shape is more peaked near the center, indicating that near neighbor spins tend to be parallel.

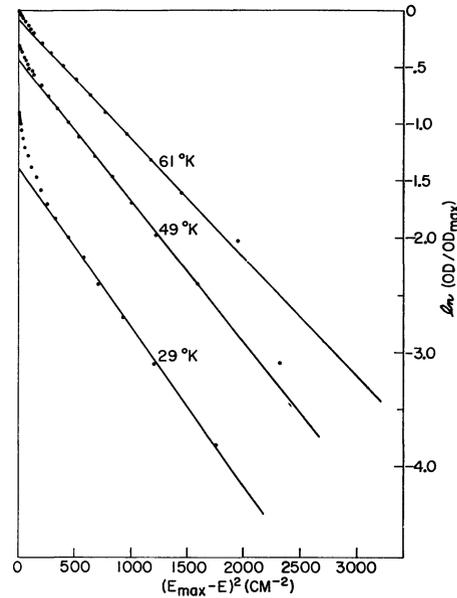


FIG. 5. — Plot of $\ln(OD/OD_{MAX})$ vs. $(E_{MAX} - E)^2$ for the low energy side of the 4 268 Å band. A linear relation indicates a Gaussian shape.

The slope of the straight line in Figure 5 gives a measure of Γ in the relation

$$\alpha(E) = \alpha_0 \exp \left[- \frac{(E_p - E)^2}{\Gamma^2} \right].$$

The slope is nearly the same for all temperatures, but is somewhat smaller at the highest temperature measured. The high temperature value is $\Gamma^2 = 1040 \text{ cm}^{-2}$ ($\Gamma = 32.2 \text{ cm}^{-1}$). This can be used to obtain a value for C in eq. 10-13. A calculation identical to that in reference [1] for the F center spin resonance line shows that the second moment of the absorption line is given by

$$\begin{aligned} \langle (E - E_0)^2 \rangle &= \\ &= C^2 \left\{ \sum_i \langle m_{i1}^2 \rangle + \sum_i \langle m_{j2}^2 \rangle \right\}. \end{aligned} \quad (14)$$

Using

$$m_{ij}^2 = \frac{1}{3} J(J + 1)$$

we have

$$\begin{aligned} \langle (E - E_0)^2 \rangle &\equiv \frac{1}{2} \Gamma^2 = \frac{16}{3} C^2 \quad J = 1 \\ &= 2 C^2 \quad J = \frac{1}{2} \end{aligned} \quad (15)$$

or,

$$\begin{aligned} C &= 9.85 \text{ cm}^{-1} \quad J = 1 \\ &= 16.1 \text{ cm}^{-1} \quad J = \frac{1}{2}. \end{aligned}$$

This can be used to estimate a molecular field at zero temperature :

$$E_{\text{ex}} = g\mu H_m = C \sum_i m_{J_i}. \quad (16)$$

Using $g = 4.1$ for $J = 1$ and 8.2 for $J = \frac{1}{2}$ to agree with the observed saturation magnetization in the saturated paramagnetic state,

$$H_m = \begin{cases} 2.0 \times 10^5 \text{ G} & J = 1 \\ 0.82 \times 10^5 \text{ G} & J = \frac{1}{2}. \end{cases}$$

These numbers bracket the value $1.4 \times 10^5 \text{ G}$ obtained by Carrara for the ferromagnetic molecular field.

Another feature of the data is the shift in peak energy between the low and high temperature absorption bands. This shift is about equal to the splitting of the band at intermediate temperatures. It is as though the high temperature and low temperature spectra simply super-impose at intermediate temperatures. This shift should, according to the magnon model, equal the magnon gap. The observed shift is approximately 2.5 \AA which corresponds to 13 cm^{-1} . Considering that the measurement is not very precise, this should be considered reasonable agreement with Jacobs' value of 16.4 cm^{-1} .

III. — One of the unusual properties of FeCl_2 is its transformation in external fields to a new phase with all spins parallel. This saturated paramagnetic phase is achieved in relatively small external fields (10.6 kG) because of the small size of the antiferromagnetic interaction between layers.

The ability to align the spins combined with the importance of the spin-orbit interaction in the ground state suggests that a large circular dichroism may occur for some of the absorption bands, which could be useful in identifying them.

We measured the circular dichroism at low temperature over the entire wavelength range reported here, and observed a large effect only for the lowest energy

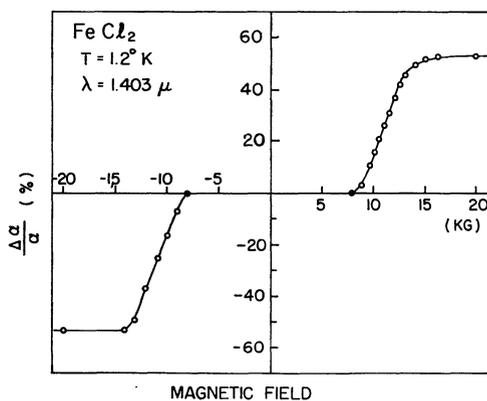


FIG. 6. — Circular dichroism of the IR band as a function of applied field.

band at 0.9 eV . The field dependence and spectrum are shown in figures 6 and 7 respectively. The field dependence shows that the signal is proportional to the magnetization since $M(H)$ behaves in a similar manner [12]. The spectrum shows that there is predominantly a zeroth moment change with a noticeable first moment change as well.

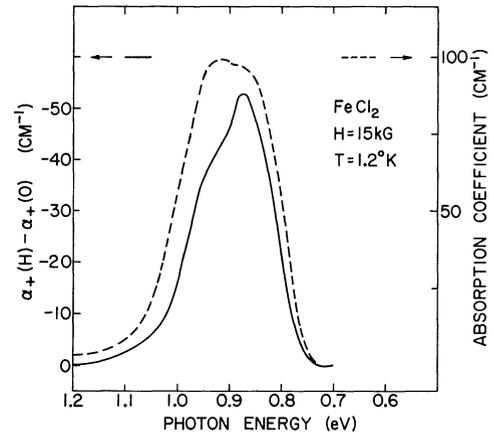


FIG. 7. — Solid line : circular dichroism of the IR band as a function of energy. Dashed line : absorption coefficient as a function of energy.

The magnitude of the circular dichroism in an ideal sample would be larger than that reported here by a constant unknown factor. This correction is due to the depolarization effect of the sample caused by imperfect alignment along the C axis and the fact that it was not a perfect single crystal.

The first moment change with magnetization saturated is measured to be $62 \pm 10 \text{ cm}^{-1}$. This result can be used to estimate an excited state g value, but the uncertainty in signal size due to depolarization make the estimate of little value.

The ground state is characterized by orbital electronic angular momentum projection $m_e = 1$ [1] and the e_g excited state by $m_e = 0$ so we expect a large zeroth moment change using circular polarization. A 50 % change in strength is observed ; this could be as large as 100 % change in an ideal sample. This effect could be used as a simple method for observing the spatial variation of the magnetization in a finite sample as the field is changed.

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