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FAR INFRARED ANTIFERROMAGNETIC RESONANCE IN FeCl_2 , FeBr_2 AND FeI_2

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Résumé. — Nous avons effectué des expériences de résonance antiferromagnétique dans FeCl_2 , FeBr_2 et FeI_2 en utilisant la technique de spectroscopie par transformation de Fourier dans l'infrarouge très lointain. Dans FeCl_2 et FeBr_2 nos résultats s'interprètent bien dans le modèle à deux sous-réseaux avec un paramètre d'anisotropie $D = 9,6$ K. Dans FeI_2 trois pics d'absorption situés à $21,6$; $29,2$ et $32,2$ cm^{-1} ont été attribués à des excitations d'ondes de spin en centre de zone, ce qu'on ne peut comprendre qu'en supposant que FeI_2 possède plus de deux sous-réseaux. Nous avons calculé les fréquences de résonance antiferromagnétique dans un modèle simple à quatre sous-réseaux, et nous avons déduit les paramètres adaptés à FeI_2 . Cependant la séparation de 3 cm^{-1} que l'on observe expérimentalement entre les deux modes de haute fréquence suggère qu'un tel modèle pourrait être trop simple.

Abstract. — A far infrared investigation of antiferromagnetic resonance by means of Fourier-transform spectroscopy has been carried out in FeCl_2 , FeBr_2 and FeI_2 . In FeCl_2 and FeBr_2 our results are found to agree well with the two-sublattice model with an anisotropy parameter $D = 9.6$ K. In FeI_2 three absorption peaks situated at 21.6 cm^{-1} , 29.2 cm^{-1} , and 32.2 cm^{-1} have been attributed to excitation of zone-center spin waves, which can be understood only by assuming that FeI_2 has more than two sublattices. The AFMR frequencies in a simple four sublattice model have been calculated, and the parameters relevant to FeI_2 have been deduced. However the observation of a 3 cm^{-1} splitting between the two higher modes suggests that such a model may be too simple.

1. **Introduction.** — The magnetic properties of FeCl_2 and FeBr_2 , extensively studied in recent years, have been shown [1-4] to be consistent with the model of a two sublattice antiferromagnet having a large anisotropy, strong ferromagnetic interaction between the ions within the c -planes and weak antiferromagnetic interaction between planes. In contrast it has been shown that some magnetic properties [5] and the magnetic structure [6] of FeI_2 are quite different from the other two halides.

Recently the interest in these compounds has focused on their excitations : phonons [7], magnons [8-10] and magnon-phonon interactions [11-13].

In this paper we report the first far-infrared spectra

of FeBr_2 and FeI_2 . The transmission between 5 and 100 cm^{-1} was measured at low temperature using the technique of Fourier-transform spectroscopy.

As will be discussed later, it appears from our results that the spin dynamics of FeI_2 are quite different from those of FeCl_2 and FeBr_2 .

2. **Experimental technique.** — The results described in this paper were obtained using a far infrared Michelson interferometer associated with a cryostat containing both the sample and the bolometer detector. The frequency range of the interferometer is 5 - 500 cm^{-1} and the maximum resolution approaches 0.1 cm^{-1} . The sample temperature can be varied between 1 K and 50 K while the bolometer is kept at a constant temperature of 1.3 K with a proportional-integral-differential temperature controller. Cold fil-

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ters (KBr 1 mm thick) can be inserted before the sample and before the bolometer to remove radiation of higher energy. A detailed description of the experimental setup is given in another paper [14].

The single crystals of FeCl_2 , FeBr_2 and FeI_2 were prepared by S. Legrand at CEN Saclay. The first two were grown using the Bridgman technique. Small slices of approximately $12 \times 12 \times 0.6 \text{ mm}^3$ were cleaved from the boule. FeI_2 was grown by the sublimation method, leaving small circular platelets of about 8 mm diameter and 0.3 mm thickness. All experiments were carried out with the c -axis of the crystal (which is perpendicular to the flat face of the sample) parallel to the propagation direction of the radiation.

3. Experimental results. — **3.1 FeCl_2 .** — A typical result of transmission measurements between 5 and 24 cm^{-1} in FeCl_2 is given in figure 1. The experiments were carried out at 1.2 K with a resolution of 1 cm^{-1} . The frequency of the AFMR, as determined from six experiments made on two different samples, is $16.51 \pm 0.06 \text{ cm}^{-1}$. This is in good agreement with the values found by Jacobs *et al.* [15] ($16.3 \pm 0.3 \text{ cm}^{-1}$) and Johnson and Sievers [16] ($16.7 \pm 0.2 \text{ cm}^{-1}$) using the same technique. This is also in agreement with the results obtained from neutron inelastic scattering experiments by Vettier and Yelon [10] ($16.7 \pm 0.3 \text{ cm}^{-1}$). A previous determination by Birgeneau [8] led to a value of $17.2 \pm 0.4 \text{ cm}^{-1}$ slightly greater than ours.

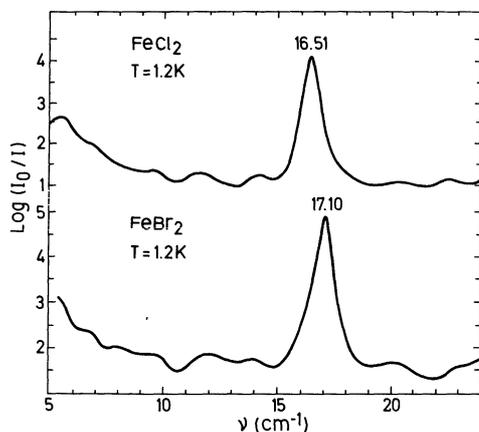


FIG. 1. — Far-infrared absorption spectrum of FeCl_2 and FeBr_2 at $T = 1.2 \text{ K}$.

In addition some transmission measurements were performed between 10 and 100 cm^{-1} which do not reveal any extra absorption in this frequency range. Another experiment has also been made with a resolution of 0.2 cm^{-1} which shows that the width of the resonance is 1.2 cm^{-1} .

3.2 FeBr_2 . — The far-infrared spectrum of FeBr_2 (Fig. 1) is qualitatively similar to that of FeCl_2 with one absorption peak at $17.09 \pm 0.06 \text{ cm}^{-1}$. This can be compared to the value 17.7 cm^{-1} obtained by

Yelon and Vettier [9] from neutron scattering and also to the value of 17.3 cm^{-1} deduced by Fert [17] from magnetic field laser experiments in the paramagnetic phase.

3.3 FeI_2 . — In contrast to the case of FeCl_2 and FeBr_2 , the FeI_2 spectrum exhibits three absorption peaks at $\nu_1 = 21.6 \pm 0.1 \text{ cm}^{-1}$, $\nu_2 = 29.2 \pm 0.1 \text{ cm}^{-1}$ and $\nu_3 = 32.2 \text{ cm}^{-1}$. The relative strengths of these absorption peaks are respectively 1.00, 0.55 and 0.50. A detail of the FeI_2 spectrum between 15 and 37 cm^{-1} , observed at $T = 1.2 \text{ K}$ with an experimental resolution of 0.5 cm^{-1} , is shown in figure 2. An experiment performed at $T = 4.2 \text{ K}$ has not revealed any shift of the peaks, but the three peaks disappeared

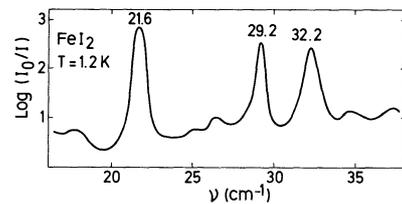


FIG. 2. — Antiferromagnetic resonances in FeI_2 at 1.2 K.

at $T = 12 \text{ K}$ (a temperature higher than $T_N = 9.3 \text{ K}$). Experiments made with a thicker beamsplitter have led us to the conclusion that there is no other resonance line between 6 and 15 cm^{-1} . Finally experiments made with a thinner beamsplitter (Fig. 3) show that, in addition to the three lines considered above, the spectrum exhibits only two small bumps at 70.8 and 83.7 cm^{-1} whose intensity is too small to be attributed to spin waves, and are thought to be due to impurities. The increase of the absorption for frequencies higher than 90 cm^{-1} is thought to arise from the excitons of the $J = 2$ ferrous ion levels.

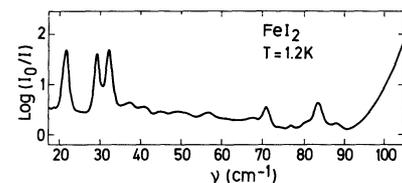


FIG. 3. — Far-infrared spectrum of FeI_2 between 15 and 100 cm^{-1} .

4. Discussion. — **4.1 FeCl_2 - FeBr_2 .** — The far-infrared spectra of FeCl_2 and FeBr_2 , with only one resonance frequency, agree well with the theory of spin waves in a two sublattice model. It has been shown that the doubly degenerate resonance frequency may be written, in the notations of Keffer [18] :

$$E(k=0) = [(g\mu_B H_A - J_0^{ab})^2 - (J_0^{ab})^2]^{1/2} \quad (1)$$

where J_0^{ab} is related to the intersublattice exchange interaction J_1' by

$$J_0^{ab} = 2 z_1' J_1' \quad (2)$$

z'_1 being the number of nearest neighbours which are coupled by J'_1 . H_A is an anisotropy parameter which describes both the single-ion and the exchange anisotropy.

Using the values :

$$\begin{aligned} 2 J'_1 (\text{FeCl}_2) &= - 0.35 \text{ K} \\ 2 J'_1 (\text{FeBr}_2) &= - 2.89 \text{ K} \end{aligned} \quad (3)$$

which are deduced from the metamagnetic fields, and :

$$\begin{aligned} z'_1 (\text{FeCl}_2) &= 6 \\ z'_1 (\text{FeBr}_2) &= 2, \end{aligned} \quad (4)$$

we obtain from our results :

$$\begin{aligned} g\mu_B H_A (\text{FeCl}_2) &= 21.7 \pm 0.1 \text{ K} \\ g\mu_B H_A (\text{FeBr}_2) &= 19.5 \pm 0.1 \text{ K}. \end{aligned} \quad (5)$$

In order to further the analysis we have to consider the detailed spin Hamiltonian appropriate for FeCl₂ and FeBr₂ [8] :

$$\begin{aligned} \mathcal{H} = - \sum_i D(S_i^3)^2 - 2 \sum_{i>j} \frac{(g'_z)^2 - (g'_x)^2}{(g'_x)^2} \\ \times J_{ij} S_i^3 S_j^3 - 2 \sum_{i>j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \end{aligned} \quad (6)$$

where D is the single-ion anisotropy parameter and J_{ij} the exchange constant between effective spins \mathbf{S}_i and \mathbf{S}_j ($S = 1$). The second term is a consequence of the anisotropy of the spectroscopic g' factors relating the true spin Σ ($\Sigma = 2$) to the effective spin \mathbf{S} ($S = 1$).

The first two terms of eq. (6) contribute to H_A :

$$g\mu_B H_A = D + \frac{(g'_z)^2 - (g'_x)^2}{(g'_x)^2} (J_0^{aa} - J_0^{ab}), \quad (7)$$

where J_0^{aa} can be related to the intrasublattice interaction parameters J_1 and J_2 between first and second neighbours by :

$$J_0^{aa} = 2 z_1 J_1 + 2 z_2 J_2, \quad (8)$$

with $z_1 = z_2 = 6$ in both FeCl₂ and FeBr₂.

Using the values deduced from the results of Birgeneau [8] and Yelon [9] :

$$\begin{aligned} J_0^{aa} (\text{FeCl}_2) &= 41.2 \text{ K} \\ J_0^{aa} (\text{FeBr}_2) &= 29 \text{ K} \\ \left(\frac{g'_x}{g'_z} \right)^2 (\text{FeCl}_2, \text{FeBr}_2) &= 0.78, \end{aligned} \quad (9)$$

we deduce from our results and eq. (7) :

$$\begin{aligned} D (\text{FeCl}_2) &= 9.5 \text{ K} \\ D (\text{FeBr}_2) &= 9.7 \text{ K}. \end{aligned} \quad (10)$$

It therefore appears that the two compounds have approximately the same anisotropy parameter. Fert has arrived at the same conclusion (5) from perpendicular susceptibility measurements, although he found a value of D (14 K) greater than ours. However the results of Vettier (10.2 K in FeCl₂ and 10.7 K in FeBr₂) obtained from spin wave analysis are very close to ours.

4.2 FeI₂. — On the contrary the spectrum of FeI₂ is not compatible with the usual theory of two sublattice antiferromagnets since in this model there are only two independent excitations. Our results show that the oscillator strengths of the three main resonances ν_1, ν_2, ν_3 are respectively proportional to 2, 1, 1 which implies that the degeneracy of the first one is twice that of the other two. We therefore infer that the number of independent excitations, and thus the number of sublattices, may be 4, 8, 16... :

i) If the twofold degeneracy associated with the uniaxial AF case is removed (as in the case of MnO) the lines ν_2 and ν_3 are singly degenerate and the number of sublattices is 4.

ii) If this twofold degeneracy applies to ν_2 and ν_3 the number of sublattices is 8 (or a multiple of 8).

This is consistent with the results of neutron diffraction (6) which have been accounted for with a magnetic cell containing 8 or 16 ferrous ions. However this is not consistent with the 12 sublattice model proposed by Graaf and Trooster [19] to explain their Mössbauer experiments. The structure of FeI₂, as determined by Gelard *et al.* [6], may also be regarded as a stack of (101) planes containing parallel spins, the sequence in the spin direction between successive planes being $\uparrow \uparrow \downarrow \downarrow$. Thus it is clear that for such a sequence the minimum number of sublattices is four, independently of the spatial distribution of the spins in the planes. This is why we develop in the following section the calculation of the AFMR frequencies in a four sublattice model.

5. A four sublattice model for FeI₂. — We start with the model shown on figure 4a which consists of four sublattices 1, 2, 3, 4 with spins following the sequence $\uparrow \uparrow \downarrow \downarrow$. We also assume that such a configuration is a simple consequence of the magnetic ordering, so that the symmetry of the interactions is that of the paramagnetic phase. This implies :

$$\begin{aligned} S_1 = S_2 = S_3 = S_4 &= 1 \\ \mathcal{H}_1 = \mathcal{H}_2 = \mathcal{H}_3 = \mathcal{H}_4 & \\ J_{12} = J_{23} = J_{34} = J_{41} & \\ J_{13} = J_{24}, & \end{aligned} \quad (11)$$

where $\mathcal{H}_{1,2,3,4}$ is the single ion Hamiltonian for sublattices 1, 2, 3, 4 and J_{ij} the exchange constants between sublattices i and j .

Such a configuration may be regarded as two imbricated antiferromagnets A and B with sublattices

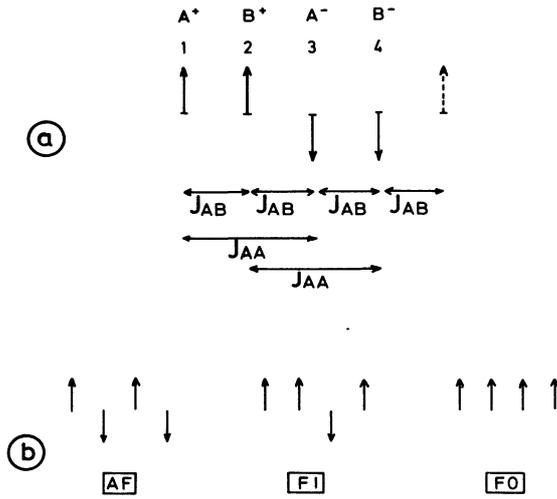


FIG. 4. — a) The four sublattice model and its exchange interactions. b) The three phases with respect to which the four sublattice phase must be stable.

tices $A^+ B^+ A^- B^-$, so that the two independent constants may be renamed J_{AA} and J_{AB} according to figure 4a. The Hamiltonian of the system may be written :

$$\begin{aligned} \mathcal{H} = & -h_A(S_1^z)^2 - h_A(S_2^z)^2 - h_A(S_3^z)^2 - h_A(S_4^z)^2 \\ & - 2K_{AB}(S_1^z S_2^z + S_2^z S_3^z + S_3^z S_4^z + S_4^z S_1^z) \\ & - J_{AB}(S_1^+ S_2^- + S_1^- S_2^+) + \text{c.p.} (1, 2, 3, 4) \\ & - 2K_{AA}(S_1^z S_3^z + S_2^z S_4^z) \\ & - J_{AA}(S_1^+ S_3^- + S_1^- S_3^+ + S_2^+ S_4^- + S_2^- S_4^+), \end{aligned} \quad (12)$$

where h_A is the intrasublattice anisotropy parameter.

According to equation (6) the anisotropic exchange constants $K_{\alpha\beta}$ and $J_{\alpha\beta}$ of (12) are related by :

$$K_{\alpha\beta} = \left(\frac{g'_z}{g'_x} \right)^2 J_{\alpha\beta}. \quad (13)$$

The equation of motion :

$$i\hbar \frac{dS_i^\alpha}{dt} = [S_i^\alpha, \mathcal{H}], \quad (14)$$

when applied to (12) and linearized, gives rise to a set of four coupled equations, the resolution of which is equivalent to the diagonalization of the following matrix :

$$\begin{bmatrix} h_A - 2K_{AA} & -2J_{AB} & -2J_{AA} & -2J_{AB} \\ -2J_{AB} & h_A - 2K_{AA} & -2J_{AB} & -2J_{AA} \\ 2J_{AA} & 2J_{AB} & -h_A + 2K_{AA} & 2J_{AB} \\ 2J_{AB} & 2J_{AA} & 2J_{AB} & -h_A + 2K_{AA} \end{bmatrix} \quad (15)$$

whose eigenvalues are :

$$\begin{aligned} E_{1,2}^2 = & (h_A - 2K_{AA})^2 - 4J_{AA}^2 - \\ & - 4J_{AB}(h_A + 2K_{AA} + 2J_{AA}) \\ E_{3,4}^2 = & (h_A - 2K_{AA})^2 - 4J_{AA}^2 + \\ & + 4J_{AB}(h_A + 2K_{AA} + 2J_{AA}). \end{aligned} \quad (16)$$

In this simple model, the AFMR has two doubly degenerate resonance frequencies and thus cannot explain completely the absorption spectrum of FeI_2 . However if we recall that the energies of the two higher absorption peaks differ by only 3 cm^{-1} , this model may be considered as a first approximation of FeI_2 provided that the energies $E_{1,2}$ and $E_{3,4}$ are respectively assigned to ν_1 and $(\nu_2 + \nu_3)/2$.

However three parameters, h_A , J_{AA} and J_{AB} , are involved in eq. (16) while we are left with only two experimental data, so that our experimental results allowed us only to determine the values of J_{AA} and J_{AB} as a function of h_A (Fig. 5). Another ambiguity with eq. (16) is that $E_{1,2}$ and $E_{3,4}$ are simply interchanged by changing the sign of J_{AB} , so that we can determine only the absolute value of J_{AB} .

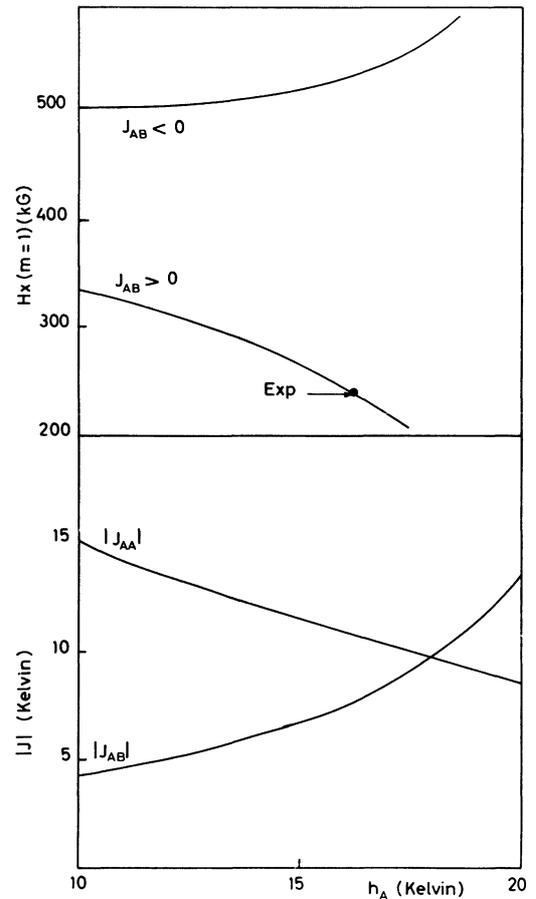


FIG. 5. — Lower curves : the values of J_{AA} and J_{AB} with are compatible with our AFMR results plotted as a function of h_A . Upper curves : the values of $H (m = 1)$ calculated with the same parameters. The point marked « Exp » is deduced from susceptibility measurements of Bertrand [20].

A first restriction in the values of h_A is provided by the stability conditions of the $\uparrow\uparrow\downarrow\downarrow$ phase in zero field. In our model this configuration is stable with respect to the three other phases AF, FI, FO, shown in figure 4 only if :

$$\begin{aligned} J_{AA} &< 0 \\ |J_{AB}| &< |J_{AA}|. \end{aligned} \quad (17)$$

If one of these conditions is not satisfied the stable phase is AF (for $J_{AB} < 0$) or FO (for $J_{AB} > 0$). This restricts the range of h_A to $h_A < 18$ K.

The complete set of values $\{h_A, J_{AA}, J_{AB}\}$ can be unambiguously determined if another property is considered, for example the perpendicular susceptibility. In our model the fractional magnetization per ion m in a perpendicular magnetic field is given by :

$$m = \frac{M}{g_x \mu_B} = \frac{g_x \mu_B H_x}{2(h_A - K_{AA} - J_{AA} - 2J_{AB})}, \quad (18)$$

where g_x is the perpendicular component of the \mathbf{g} tensor ($g_x = 3.14$).

The curve of H_x ($m = 1$) calculated with the values of h_A, J_{AA} and J_{AB} compatible with our results is plotted on figure 5. The upper branch is calculated with $J_{AB} < 0$, the lower one with $J_{AB} > 0$. From the experimental value H_x ($m = 1$) = 240 kG deduced from the perpendicular susceptibility measurements of Bertrand *et al.* [20] we obtain :

$$\begin{aligned} h_A &= 16.2 \text{ K} \\ J_{AB} &= 7.8 \text{ K} \\ J_{AA} &= -10.8 \text{ K}, \end{aligned} \quad (19)$$

and consequently :

$$\begin{aligned} K_{AB} &= 10 \text{ K} \\ K_{AA} &= -13.8 \text{ K}. \end{aligned} \quad (20)$$

In our formalism h_A includes both the single-ion anisotropy and the contribution of the anisotropic intrasublattice interaction J_{11} :

$$h_A = D + 2 \frac{(g'_z)^2 - (g'_x)^2}{(g'_x)^2} J_{11}. \quad (21)$$

The total anisotropy which includes the single-ion and all the anisotropic exchange contributions is :

$$g\mu_B H_A = h_A - 2(K_{AA} - J_{AA}) = 22 \text{ K} \quad (22)$$

Although our experimental results give no information on J_{11} we can estimate that, in the case of FeI₂, $J_{11} \simeq J_{AB}$, because, if we restrict ourselves to the nearest neighbours, the pairs which contribute to both interactions are equivalent. In this approximation we obtain :

$$D = 11.8 \text{ K}, \quad (23)$$

a value roughly similar to that of FeCl₂ and FeBr₂.

From the comparison of (5), (10), (22) and (23) it appears that the dynamics of FeCl₂, FeBr₂ and FeI₂ may be described by similar single-ion parameters; the main difference — namely the existence in FeI₂ of low frequency (21.6 cm⁻¹) and high frequency modes (29.2 cm⁻¹, 32.2 cm⁻¹) — being a natural consequence of the magnetic ordering of FeI₂.

6. Conclusion. — Our experimental results of anti-ferromagnetic resonance in FeCl₂ and FeBr₂ have been found to be consistent with the two sublattice model. On the contrary, in the case of FeI₂, where three absorption peaks have been observed, we have had to consider a more complicated model involving four sublattices. This model has been shown to account for the results of AFMR and perpendicular susceptibility with an anisotropy $h_A = 16.2$ K and two exchange constants $J_{AA} = -10.8$ K and $J_{AB} = 7.8$ K. The splitting between the two higher peaks however could be explained only by a more complex model, involving more than one anisotropy parameter or more than four sublattices. It should be possible to determine whether the number of sublattices is 4 or 8 by means of AFMR experiments using an applied magnetic field : if the lines at ν_2 and ν_3 remain singly degenerate in applied magnetic field there are four sublattices, whereas if they both split the number of sublattices would be eight.

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