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Influence of True Crystallographic Structure on Spin Waves in Ferromagnetic $CrBr_3$†

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Previous spin-wave-renormalization calculations, even those including correlation effects, have been unable to explain correctly both the temperature and field dependence of the magnetization of ferromagnetic $CrBr_3$. However, these calculations were based on a simplified model lattice, which has been shown by recent neutron-diffraction experiments to be only qualitatively correct. In the following, we present a first-order renormalized spin-wave theory which takes into account the true crystallographic structure of $CrBr_3$. This theory explains simultaneously the temperature and field dependence of the magnetization and the magnon dispersion curves. Although the simplified model contains only two exchange parameters, while the exact model contains five, we find that once the true crystallographic structure is introduced, a two-parameter model can explain $M(T, H)$ and the low-energy part of the magnon spectrum. We conclude that the previous discrepancies between theory and experiment were not a result of any failure of spin-wave theory or of the presence of too few exchange parameters, but were due simply to a misrepresentation of the crystal structure.

I. INTRODUCTION

$CrBr_3$ is a rhombohedral insulating ferromagnet with $T_c = 32.5$ °K. Because of its weak layered structure, this system is particularly useful for testing the validity of spin-wave renormalization calculations.[1]

Nuclear-magnetic-resonance (NMR) measurements[2-5] have furnished a very accurate determination of the magnetization $M(T, H)$ versus temperature and field. These results were explained by means of various spin-wave theories. Because of the complicated crystallographic structure of $CrBr_3$, a simplified model was introduced by Gossard, Jaccarino, and Remeika,[2] involving only two exchange parameters: J_L between nearest neighbors in adjacent planes and J_T between nearest neighbors in the same plane. From their measurement of $M(T, 0)$ versus T, Davis and Narath[3] calculated $J_T = 0.497 \pm 0.013$ °K and $J_L = 8.25 \mp 0.10$ °K using a self-consistent first-order renormalized spin-wave theory. However, as has been recently shown,[5] while this set of values of the two exchange parameters gives a very good fit to the magnetization $M(T, 0)$ versus T in zero field, it does not explain the magnetization $M(T_0, H)$ versus H at $T_0 = 18$ °K. In fact, it is shown in Ref. 5 that even a more sophisticated spin-wave theory correct to all orders in the magnon-magnon interaction is unable to explain simultaneously the $M(T, 0)$-vs-T and $M(T_0, H)$-vs-H data. The authors attributed this inability to the inadequacy of the two-parameter model, citing as evidence recent inelastic neutron scattering measurements[6] of the magnon spectrum. Indeed, a five-parameter model (which takes into account the true crystallographic structure) was introduced in Ref. 6 to describe the observed spin-wave spectrum.

It is the object of this paper to derive the first-order renormalized spin-wave theory for the five-parameter model to see whether or not it is possible to obtain a unique set of parameters which gives a good fit to both the $M(T, 0)$ and $M(T_0, H)$ data and, if it is, to compare it to the set determined in Ref. 6. In fact, we may expect some differences between the two sets to appear, since we shall neglect correlations between spin waves. In Sec. II, we develop a formal Hartree-Fock theory of spin waves in $CrBr_3$, closely following Davis and Narath. We then compare the predictions of this theory to the neutron-diffraction experiments. In Sec. IV, we calculate both the temperature and field dependence of the magnetization $M(T, H)$.

II. GENERAL THEORY

$CrBr_3$ has a BiI_3-type crystal structure, which is invariant under the operations of the space group $R\overline{3}$. The unit cell is rhombohedral and contains two Cr^{3+} ions. We consider a system of $2N$ spins with periodic boundary conditions; the spin operators corresponding to the spins of the two sublattices are denoted $\vec{R}_i$ and $\vec{S}_j$ with $|\vec{R}_i| = |\vec{S}_j| = S$. $H_A(T)$ is the anisotropy field directed along z, as measured by Dillon.[7] J_1, J_2, J_4 and J_3, J_5 are, respectively, intersublattice and intrasublattice exchange parameters. $\vec{\delta}'$ and $\vec{\delta}$ are vectors from a given spin site to neighbors belonging, respectively, to the same and different sublattices (the vectors $\vec{\delta}'$ and $\vec{\delta}$ relative to a site R and a site S are opposite). The Hamiltonian is

$$\mathcal{H} = -g\mu_B H_A(T)\left(\sum_i R_i^z + \sum_j S_j^z\right) - \sum_{m=1,2,4} J_m\left(\sum_{i,\delta_m} \vec{R}_i \cdot \vec{S}_{i+\delta_m} + \sum_{j,\delta_m} \vec{S}_j \cdot \vec{R}_{j+\delta_m}\right) - \sum_{m=3,5} J_m\left(\sum_{i,\delta'_m} \vec{R}_i \cdot \vec{R}_{i+\delta'_m} + \sum_{j,\delta'_m} \vec{S}_j \cdot \vec{S}_{j+\delta'_m}\right) . \quad (1)$$

We introduce the angular momentum raising and lowering operators $R_i^{\pm} = R_i^x \pm R_i^y$ and $S_j^{\pm} = S_j^x \pm S_j^y$. Performing the Dyson-Maleev transformation,[8,9] we have

$$\begin{aligned} R_i^z &= S - b_i^\dagger b_i , \\ R_i^- &= (2S)^{1/2} b_i^\dagger , \\ R_i^+ &= (2S)^{1/2} (b_i - b_i^\dagger b_i b_i/2S) , \\ S_j^z &= S - a_j^\dagger a_j , \\ S_j^- &= (2S)^{1/2} a_j^\dagger , \\ S_j^+ &= (2S)^{1/2} (a_j - a_j^\dagger a_j a_j/2S) . \end{aligned} \quad (2)$$

Fourier transforming with, e.g., $a_k = N^{-1/2} \times \sum_i e^{ik\cdot x_i} a_i$, and introducing the quantities

$$E_0 = -2g\mu_B NS H_A(T) - 2NS^2[J_{RR}(0) + J_{SR}(0)] , \quad (3a)$$

$$J_{SR}(k) = \sum_{m=1,2,4} J_m \sum_{\delta_m} e^{i\vec{k}\cdot\vec{\delta}_m} \equiv J_1(k) + J_2(k) + J_4(k) , \quad (3b)$$

$$J_{RS}(k) = J_{SR}(k)^* , \quad (3c)$$

$$J_{RR}(k) = \sum_{m=3,5} J_m \sum_{\delta'_m} e^{-i\vec{k}\cdot\vec{\delta}'_m} \equiv J_3(k) + J_5(k) , \quad (3d)$$

we write

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 , \quad (4)$$

where

$$\begin{aligned} \mathcal{H}_0 = E_0 + \textstyle\sum_k [& g\mu_B H_A(T) + 2SJ_{RS}(0) \\ & + 2SJ_{RR}(0) - 2SJ_{RR}(k)] (a_k^\dagger a_k + b_k^\dagger b_k) \\ & - 2S\textstyle\sum_k [J_{RS}(k) b_k^\dagger a_k + J_{SR}(k) a_k^\dagger b_k], \end{aligned} \quad (5)$$

and

$$\begin{aligned} \mathcal{H}_1 = \frac{1}{N} \sum_{1,2,3,4} & \delta(k_1 + k_2 - k_3 - k_4) \\ & \times \{ [J_{RR}(k_1) - J_{RR}(k_2 - k_1)] (a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3} a_{k_4} + b_{k_1}^\dagger b_{k_2}^\dagger b_{k_3} b_{k_4}) \\ & + J_{SR}(k_1) b_{k_1}^\dagger a_{k_2}^\dagger a_{k_3} a_{k_4} + J_{SR}(-k_1) a_{k_1}^\dagger b_{k_2}^\dagger b_{k_3} b_{k_4} \\ & - 2J_{SR}(k_2 - k_1) a_{k_1}^\dagger b_{k_2}^\dagger a_{k_3} b_{k_4} \} . \end{aligned} \quad (6)$$

$\mathcal{H}_0$ is diagonalized through the following transformation:

$$\begin{aligned} a_k &= (1/\sqrt{2}) e^{i\Theta_k} (\alpha_k + \beta_k) , \\ b_k &= (1/\sqrt{2}) e^{-i\Theta_k} (\alpha_k - \beta_k) , \end{aligned} \quad (7)$$

with

$$e^{4i\Theta_k} = J_{SR}(-k)/J_{SR}(k) . \quad (8)$$

The diagonal form of $\mathcal{H}_0$ is

$$\mathcal{H}_0 = E_0 + \textstyle\sum_k \{A(k) - [B(k)]^{1/2}\} \alpha_k^\dagger \alpha_k + \textstyle\sum_k \{A(k) + [B(k)]^{1/2}\} \beta_k^\dagger \beta_k , \quad (9)$$

with

$$A(k) = 2S[J_{SR}(0) + J_{RR}(0) - J_{RR}(k)] , \quad (10a)$$

$$B(k) = 4S^2 J_{SR}(k) J_{SR}(-k) . \quad (10b)$$

$\mathcal{H}_0$ represents the energy of Holstein-Primakoff spin waves which are the true collective excitations of the system at low temperatures; $\alpha_k^\dagger \alpha_k$ is the number of acoustical spin waves of wave number k, $\beta_k^\dagger \beta_k$ the number of optical spin waves. The above result was given in Ref. 6. Writing $\mathcal{H}_1$ in terms of the α's and β's and linearizing, which corresponds to a self-consistent first Born approximation to Dyson's theory,[10] we obtain the renormalized spin-wave energies (the upper sign holds for the optical branch and the lower for the acoustical one):

$$\begin{aligned}\epsilon^{\pm}_{k_1}(T) = {} & g\mu_B H_A(T) + 2S\{J_{SR}(0) + J_{RR}(0) - J_{RR}(k_1) \pm [J_{SR}(k_1)J_{SR}(-k_1)]^{1/2}\} \\ & - \frac{1}{N}\sum_{k_2}[J_{RR}(0) + J_{RR}(k_2-k_1) - J_{RR}(k_1) - J_{RR}(k_2)]\,(\langle \alpha^{\dagger}_{k_2}\alpha_{k_2}\rangle + \langle \beta^{\dagger}_{k_2}\beta_{k_2}\rangle) \\ & - \frac{1}{N}\sum_{k_2}\{J_{SR}(0) \mp \tfrac{1}{2}J_{SR}(k_2-k_1)\,e^{2i(\Theta_{k_2}-\Theta_{k_1})} \mp \tfrac{1}{2}J_{SR}(k_1-k_2)\,e^{2i(\Theta_{k_1}-\Theta_{k_2})} \pm [J_{SR}(k_1)J_{SR}(-k_1)]^{1/2} \\ & - [J_{SR}(k_2)J_{SR}(-k_2)]^{1/2}\}\langle \alpha^{\dagger}_{k_2}\alpha_{k_2}\rangle - \frac{1}{N}\sum_{k_2}\{J_{SR}(0) \pm \tfrac{1}{2}J_{SR}(k_2-k_1)\,e^{2i(\Theta_{k_2}-\Theta_{k_1})} \pm \tfrac{1}{2}J_{SR}(k_1-k_2)\,e^{2i(\Theta_{k_1}-\Theta_{k_2})} \\ & \pm [J_{SR}(k_1)J_{SR}(-k_1)]^{1/2} + [J_{SR}(k_2)J_{SR}(-k_2)]^{1/2}\}\langle \beta^{\dagger}_{k_2}\beta_{k_2}\rangle\,, \qquad (11)\end{aligned}$$

with

$$\langle \alpha^{\dagger}_k\alpha_k\rangle = \frac{1}{e^{\epsilon^{-}_k(T)/kT}-1}\,, \qquad \langle \beta^{\dagger}_k\beta_k\rangle = \frac{1}{e^{\epsilon^{+}_k(T)/kT}-1}\,. \qquad (12)$$

The symmetry properties of the crystal do not enable us to simplify the above expressions. As we shall see, however, since J_1 is much bigger than all the other exchange parameters, it is possible to introduce renormalization factors as Davis and Narath did.

First of all, we remark, as in Ref. 3, that

$$\begin{aligned}&\sum_{k_2}[J_{RR}(0) + J_{RR}(k_2-k_1) - J_{RR}(k_1) - J_{RR}(k_2)]\,\langle \alpha^{\dagger}_{k_2}\alpha_{k_2}\rangle \\ &\qquad \equiv \sum_{k_2}[J_{RR}(0) - J_{RR}(k_1)]\left[1 - \frac{J_{RR}(k_2)}{J_{RR}(0)}\right]\langle \alpha^{\dagger}_{k_2}\alpha_{k_2}\rangle\,. \qquad (13)\end{aligned}$$

The above result follows from the relation

$$J_{RR}(k) = J_3(k) + J_5(k)\,. \qquad (14)$$

We now observe that, since $J_1 \gg J_2$, J_4, then

$$\begin{aligned}[J_{SR}(k)J_{SR}(-k)]^{1/2} \simeq {} & [J_1(k)J_1(-k)]^{1/2} + \tfrac{1}{2}J_2(-k)[J_1(k)/J_1(-k)]^{1/2} \\ & + \tfrac{1}{2}J_2(k)[J_1(-k)/J_1(k)]^{1/2} + \tfrac{1}{2}J_4(-k)[J_1(k)/J_1(-k)]^{1/2} + \tfrac{1}{2}J_4(k)[J_1(-k)/J_1(k)]^{1/2}\,. \qquad (15)\end{aligned}$$

Using Eqs. (13) and (15), we obtain

$$\begin{aligned}\epsilon^{\pm}_k(T) = {} & g\mu_B H_A(T) + 2S\{J_1(0) \pm [J_1(k)J_1(-k)]^{1/2}\}\xi_1(T) + S\{J_2(0) \pm J_2(-k)[J_1(k)/J_1(-k)]^{1/2}\}\xi_2(T) + \text{c. c.} \\ & + 2S[J_3(0) - J_3(k)]\xi_3(T) + S\{J_4(0) \pm J_4(-k)[J_1(k)/J_1(-k)]^{1/2}\}\,\xi_4(T) + \text{c. c.} \\ & + 2S[J_5(0) - J_5(k)]\,\xi_5(T)\,, \qquad (16)\end{aligned}$$

where

$$\begin{aligned}\xi_1(T) &= 1 - \frac{1}{2SN}\sum_{k}\left[\left(1 - \frac{[J_1(k')J_1(-k')]^{1/2}}{J_1(0)}\right)\langle \alpha^{\dagger}_{k'}\alpha_{k'}\rangle + \left(1 + \frac{[J_1(k')J_1(-k')]^{1/2}}{J_1(0)}\right)\langle \beta^{\dagger}_{k'}\beta_{k'}\rangle\right], \\ \xi_2(T) &= 1 - \frac{1}{2SN}\sum_{k'}\left\{\left[1 - \frac{J_2(k')}{J_2(0)}\left(\frac{J_1(-k')}{J_1(k')}\right)^{1/2}\right]\langle \alpha^{\dagger}_{k'}\alpha_{k'}\rangle + \left[1 + \frac{J_2(k')}{J_2(0)}\left(\frac{J_1(-k')}{J_1(k')}\right)^{1/2}\right]\langle \beta^{\dagger}_{k'}\beta_{k'}\rangle\right\}, \qquad (17) \\ \xi_3(T) &= 1 - \frac{1}{2SN}\sum_{k'}\left(1 - \frac{J_3(k')}{J_3(0)}\right)(\langle \alpha^{\dagger}_{k'}\alpha_{k'}\rangle + \langle \beta^{\dagger}_{k'}\beta_{k'}\rangle)\,,\end{aligned}$$

and the expressions for $\xi_4(T)$ and $\xi_5(T)$ are similar to $\xi_2(T)$ and $\xi_3(T)$, respectively.

Introducing, as in Ref. 6 ($\vec{t}_1$, $\vec{t}_2$, $\vec{t}_3$ are the rhombohedral vectors defining the unit cell and are defined in Ref. 6) the quantities

$$\alpha = \vec{k}\cdot\vec{t}_1\,, \quad \beta = \vec{k}\cdot\vec{t}_2\,, \quad \gamma = \vec{k}\cdot\vec{t}_3\,, \qquad (18)$$

and

$$U=\cos(\alpha+\beta-\gamma)+\cos(\beta+\gamma-\alpha)+\cos(\gamma+\alpha-\beta)\,,$$
$$V=\cos\alpha+\cos\beta+\cos\gamma\,, \tag{19}$$
$$W=\cos(\alpha-\beta)+\cos(\beta-\gamma)+\cos(\gamma-\alpha)\,,$$

we may write

$$\begin{aligned}\epsilon_k^{\pm}(T)={}&g\mu_B H_A(T)+2SJ_1[3\pm(3+2W_k)^{1/2}]\xi_1\\&+2SJ_2\left(1\pm\frac{V_k}{(3+2W_k)^{1/2}}\right)\xi_2+4SJ_3(3-W_k)\xi_3\\&+6SJ_4\left(1\pm\frac{U_k+2V_k}{(3+2W_k)^{1/2}}\right)\xi_4+4SJ_5(3-V_k)\xi_5\,.\end{aligned} \tag{20}$$

In conclusion, we have taken advantage of the large value of J_1 compared to the other exchange parameters to write the expression

$$\epsilon_k(T)=\sum\nolimits_m \epsilon_k(J_m)\xi_m(T)\,, \tag{21}$$

which, for arbitrary values of J_i, is valid only for centrosymmetric ferromagnets.[11]

The above expression is valid everywhere in the Brillouin zone, except near the surface where $(3+2W)^{1/2}=0$. We note, however, that the magnons which are important for the thermodynamical properties of $CrBr_3$ have their $\vec{k}$ vector inside a cylinder of axis z, limited by two planes perpendicular to z, and of small radius compared to the distance between the planes. The surface $(3+2W)^{1/2}=0$ is well outside this cylinder, and consequently the expression above may be used to calculate the magnetization $M(T,H)$, as we shall see in the following.

III. COMPARISON TO NEUTRON-DIFFRACTION EXPERIMENTS

A. Noninteracting Spin Waves

The spin-wave spectrum of $CrBr_3$ was measured[6] along the directions Λ, Δ, Σ, F, and B at 6 °K, a temperature for which the renormalization effects are almost negligible. We therefore take $\xi_i=1$ in the preceding formulas when comparing to these experimental results. However, we first ask if these results may be explained with the two-parameter model.[2,3] According to this model, the acoustical Z branch has the energy

$$\epsilon_k^-(0)=g\mu_B H_A(0)+4SJ_L(1-\cos c'k)\,, \tag{22}$$

where c' is the interatomic distance along Z.

The experimental result[6] for the $\Lambda(Z)$ direction is

$$\epsilon_k^-(0)=\epsilon_0+\epsilon_1(1-\cos c'k)\,, \tag{23}$$

and consequently the Z branch is well described by the simplified model. The value of J_L obtained from ϵ_1 is also in good agreement with that deduced from the NMR experiments. Along Σ or Δ directions in the c plane, the simplified model gives

$$\epsilon_k^-(0)=g\mu_B H_A(0)+D(a'k)^2 \tag{24}$$

for small values of k, where $D=\frac{9}{4}J_T$ and a' is the interatomic distance in the c plane. For the Σ and Δ directions this describes the experiment[6] well and gives a reasonable value for J_T. The inadequacy of the two-parameter model appears when comparing the Δ, Σ and F, B branches. For F and B, which are directions parallel to Σ and Δ but displaced by a full reciprocal-lattice vector in Z, the result[6] is

$$\epsilon_k^-(0)=g\mu_B H_A(0)+4SJ_L+D'(a'k_{xy})^2\,. \tag{25}$$

According to the simplified model, the transverse part of the energy is independent of Z, so that one should have $D'=D$, which is not the case experimentally, where $D'/D\simeq 0.92\pm0.05$. This means that one has no additivity property of the type $\epsilon_k^-=\epsilon_{k_{xy}}^-+\epsilon_{k_z}^-$, as in the simple two-parameter model.[2,3]

Now we come back to the true model and see how the main features of the spin-wave spectrum may be explained. In the Z direction, from Eq. (18), $\alpha=\beta=\gamma$. Using the fact that $J_1\gg J_2$, J_3, J_4, J_5, we obtain

$$\epsilon_k^-(0)=g\mu_B H_A(0)+2S(J_2+3J_4+6J_5)(1-\cos\alpha)\,, \tag{26a}$$

$$\begin{aligned}\epsilon_k^+(0)={}&g\mu_B H_A(0)+2S\\&\times[6J_1+(J_2+3J_4)(1+\cos\alpha)+6J_5(1-\cos\alpha)]\,.\end{aligned} \tag{26b}$$

Consequently, as confirmed by experiment,[6] the energy of the acoustical Z branch is proportional to $1-\cos\alpha$ and independent of J_1. We note that, owing to the symmetry properties of the crystal, the phase difference between the spins R and S is identically zero at the zone center (Γ). Some phase difference which depends on the exchange parameters may appear between the two sublattices along the Z branch but it is almost zero between an R spin and an S spin belonging to the same plane. In fact, the J_1 contribution to $\epsilon_k^-(0)$ is very weak, since $W=3$ along Z. In addition, the optical Z branch is very flat, since $J_1\gg J_2$, J_4, J_5. Only in the simplified model[3] are the optical and acoustic Z branches parallel.

Comparing the two models, we find that the acoustical Z branch gives an effective longitudinal exchange parameter J_L, and we have the relation

$$2J_L=J_2+3J_4+6J_5\,. \tag{27}$$

Let us now consider the Σ acoustical branch. As a consequence of the fact that J_1 is by far the largest exchange parameter, we obtain from Eqs. (18)–(20), for small values of the wavenumber ($\beta=\gamma=\frac{1}{2}\alpha$),

$$\epsilon_k^-(0)=g\mu_B H_A(T)+D\alpha^2\,, \tag{28}$$

with

$$D=\tfrac{9}{4}(J_1+6J_3+J_4+2J_5)\ . \tag{29}$$

The same result holds for the Δ branch. For the F and B branches, we have from Eq. (18)

$$\alpha=\alpha_Z+\alpha'\ ,\quad \beta=\alpha_Z+\beta'\ ,\quad \gamma=\alpha_Z+\gamma'\ . \tag{30}$$

The same relations between α', β', γ' hold as between α, β, γ for the Σ and Δ directions, and we obtain

$$\epsilon_k^-(0)=g\mu_B H_A(T)+4SJ_L+D'\alpha'^2\ , \tag{31}$$

with

$$D'=\tfrac{9}{4}(J_1+6J_3-J_2-J_4-2J_5)\ . \tag{32}$$

The true model thus explains why $D'\neq D$. Moreover, we may consider that J_1+6J_3 is an effective transverse exchange parameter similar to J_T.

At this point, we note that the thermodynamics of $CrBr_3$ is determined by the acoustical Z branch and the quadratic part of the in-plane or out-of-plane branches, i.e., by J_L, D, and D'. Since this part of the spectrum is insensitive to the ratio J_3/J_1 (this is not true for the magnons of large k_{xy}), just one relation between J_2, J_4, and J_5 is necessary to describe the NMR results. We show below that the introduction of three parameters (J_T, J_L, and J_2 or J_L, D, and D') gives a good fit both to $M(T,0)$ and $M(T_0,H)$. Moreover, we show that this result is *not* a consequence of the increased number of parameters.

B. Interacting Spin Waves

For the acoustical Z branch, we have

$$\epsilon_k^-(T)=g\mu_B H_A(T)+2SJ_2(1-\cos\alpha)\xi_2+6SJ_4(1-\cos\alpha)\xi_4+12SJ_5(1-\cos\alpha)\xi_5\ . \tag{33}$$

As we have checked by numerical calculation (see Sec. IV), the renormalization factors ξ_2, ξ_4, and ξ_5 are almost the same. Consequently, in the first-order theory, even when using the true model, the renormalization of the acoustical Z branch is k independent. This result is also predicted by the simple model, and the magnitude of the ξ's (~ 0.12–0.15 at $0.6T_c$) is in good agreement with the experimental results,[6] within the accuracy of the measurement.

For the acoustical Σ branch, we have

$$\epsilon_k^-(T)=g\mu_B H_A(T)+\tfrac{9}{4}(J_1\xi_1+6J_3\xi_3)\alpha^2+\tfrac{9}{4}(J_4\xi_4+2J_5\xi_5)\alpha^2\ . \tag{34}$$

As J_1 is much larger than the other exchange parameters, the renormalization of this branch depends mainly on ξ_1, and consequently it is k independent from Γ to the edge of the Brillouin zone. The same result holds for the Δ branch, except just near the maximum value of the energy, for the F and B branches, and for the optical branches. Again, the value of ξ_1 (~ 0.03 at $0.6T_c$) is in good agreement with the experimental result[6] for small values of k_Σ and k_Δ. However, recent measurements[12] of $\Delta E/E$ for larger values of k_Δ suggest that the renormalization of the Δ branch is k dependent. This cannot be explained within the framework of the Hartree-Fock approximation.

IV. CALCULATION OF THE MAGNETIZATION $M(T,H)$

In the first-order spin-wave theory there are no lifetime effects, so that the magnetization is given by

$$M(T,H)=(1/V_{\mathrm{Bz}})\int_{\mathrm{Bz}}d^3k\,(e^{\beta[g\mu_B H_A(T)+\epsilon_k(T)]}-1)^{-1}\ , \tag{35}$$

where $\epsilon_k(T)$ is the renormalized energy spectrum given in Eq. (20), and Bz means Brillouin zone.

To calculate $M(T,H)$, we use the true reciprocal unit cell (α, β, γ varying from 0 to 2π). The integrations are performed by means of Simpson's rule. As a first approximation, we choose a reasonable set of the exchange parameters from the neutron-diffraction data. Knowing D, D', and J_L, we take $J_4=J_5$ and $J_3=0$ (the important spin waves have a quadratic energy in the plane, and consequently only the linear combination J_1+6J_3 has to be determined). Furthermore, we choose $J_2=J_4$, so that we are left with two parameters which may be called J_L and J_T. In fact, we know the true value of the ratio J_2/J_4 from J_L, D, and D', or more precisely from Ref. 6, but we do not expect it to give the best fit to $M(T,H)$; indeed, we are neglecting the correlations between spin waves, which, as we shall show, influence only slightly the magnetization, but much more the value of J_L, and consequently the ratio J_2/J_4.

In order to check the validity of the approximate form of the energy spectrum given in Eq. (16), we have calculated the reduced magnetization $M(T,0)$ without any renormalization using first the exact expression of $\epsilon_k^{\pm}(0)$ [Eq. (11)], then the approximate one (which is obtained from the condition $J_1\gg J_i$). The surface where this approximation is not valid is easily discarded by the integration procedure, and we have checked that the relative error introduced by the approximation is less than 10^{-4}, therefore negligible. In addition, we have found that the magnetization is almost insensitive to the existence of the optical branches (error 10^{-3}) and quite insensitive to the exact value of the spin-wave energies far from Γ in the xy plane. Finally, our result for the renormalization coefficients ξ_i at 18 °K is that ξ_2, ξ_4, and ξ_5 are similar and comparable to ξ_L of Ref. 3, while ξ_1 is comparable to ξ_T of Ref. 3 (ξ_3 has a roughly intermediate value). The theoretical values compare favorably with the experimental ones,[6] and we again have verified that in the first Born approximation, the renormalization is k independent along the Z, Δ, and Σ branches.

The results of our numerical computations of $M(T, H)$ are summarized in Fig. 1. It is possible to find a curve I in the (J_L, J_T) plane each point of which gives a good fit to $M(T, 0)$ versus T. Similarly one finds a curve II each point of which gives a good fit to the susceptibility at 18 °K. These two curves cross at P near the values

$$J_L = 0.414\ ^\circ\text{K}, \quad J_T = 8.60\ ^\circ\text{K} . \qquad (36)$$

A similar situation was found in the two-parameter model,[5] but the two curves did not cross. In the preceding calculation, we have neglected self-consistency and correlation effects; these effects have been evaluated for the simplified model.[5] Taking them into account would probably translate the curve I to I′. The quality of the fits described here is very similar to that found in Refs. 3 and 5. The theoretical values fall well within the scatter of the experimental points in all cases.

The values of the exchange parameters determined above do not agree very well with the neutron-diffraction values, even if we allow a correction for the self-consistency and correlation effects; particularly, the value of the effective J_L is too low. However, we have arbitrarily chosen the ratio J_2/J_4 to be 1, and we should allow this quantity to vary.

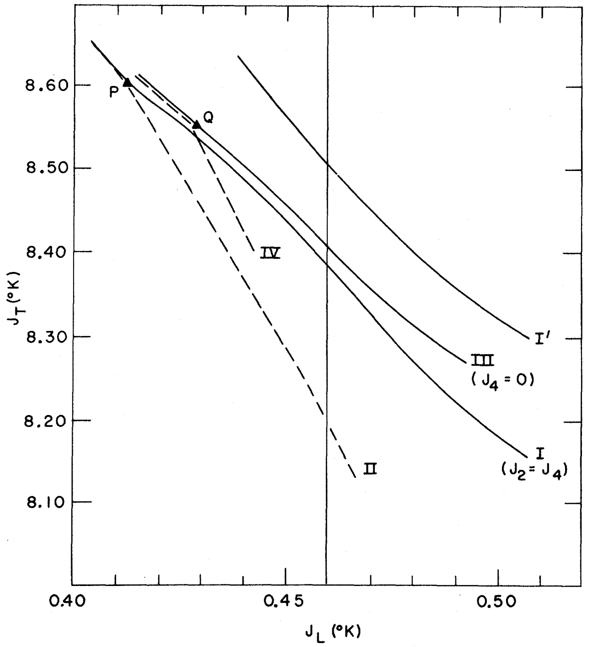

FIG. 1. Sets of exchange parameters (J_T, J_L) which give a good fit to the experimental observations: curves I and III fit to $M(T, 0)$, curves II and IV to $M(T_0, H)$, $T_0 = 18$ °K. For curves I and II, which cross at P, $J_2 = J_4$; for curves III and IV which cross at Q, $J_4 = 0$.

Increasing J_2/J_4 leads to a higher value for J_L (curves III and IV crossing in Q) and finally we get a good fit to $M(T, 0)$ and $M(T_0, H)$ for the following values of the parameters, which agree with the experimental value[6] of the difference $D - D'$:

$$J_4 = 0, \quad J_2 \equiv J_L = 0.43\ ^\circ\text{K}, \quad J_1 = 8.55\ ^\circ\text{K} . \qquad (37)$$

Taking into account the order of magnitude of self-consistency and correlation effects brings J_L to slightly more than 0.46 °K, which is compatible with the acoustical Z branch of the magnon spectrum. The exchange constants given in Eq. (37) then represent well the low-energy (thermodynamically important) spin waves.

To get a good fit to the Σ and Δ branches far from the center of the Brillouin zone, we must increase the value of J_1 to about 8.80 °K, therefore we introduce $J_3 < 0$. This implies no significant change of J_L, since the part of the spectrum which is modified is only slightly populated even at 20 °K, and the final values of the parameters J_2, J_4, and J_5 differ slightly from those determined by Samuelsen *et al.*[6] from the neutron data.[13] This slight difference is to be expected, since we have used a first-order spin-wave theory, and the effects of higher-order interactions on $M(T, H)$ have been shown[4,5] to be nonnegligible. At any rate, we wish to stress here that the motivation of the present work is not to put precise limits on the exchange constants, but to show that with the use of the true crystal structure one may formulate a spin-wave theory which describes equally well $M(T, 0)$ and $M(T_0, H)$. Of course, we expect the exchange constants used to give a reasonable agreement with the measured spectrum,[6] and they do.

Finally, we compare the two models: the two-parameter model of Gossard, Jaccarino, and Remeika[2] (two exchange interactions J_T and J_L) and the true model with J_1 and J_2 being the only nonzero exchange parameters (Fig. 2).

In the first model, we have identical planes stacked along z, two atoms in each cell, in a plane; above and below an R atom are R atoms. In the second model, we have three kinds of planes packed along z according to the sequence *ABCABC*. The symmetry is in fact rhombohedral, with two atoms in each cell, and consequently no low-energy optical branch appears along Z, since any optical mode involves J_1 (if the packing was *ABAB*, the crystal would be hexagonal with four atoms per cell and low-energy optical magnons would appear).

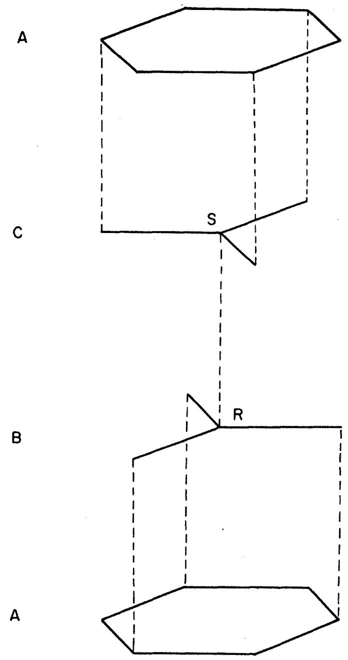

FIG. 2. True lattice of Cr^{3+} ions in $CrBr_3$. The heavy lines connect RS atoms with the exchange parameter J_1, and the dashed lines RS atoms with the exchange parameter J_2.

Although the two models have very different structural properties, they give rise to very similar magnon spectra since in each case there are two atoms per cell and the limit of the Brillouin zone along z is the same (two planes translated by $\vec{c}$ are identical irrespective of translation xy). The Σ and Δ branches depend only on J_T or J_1 (the dispersion law is almost isotropic in the two models), and the Z acoustical branch only on J_L or J_2 (in a k_z mode, the spins of one plane remain in phase). The optical z branches are slightly different because one finds RRR or RS atoms along z. Because $J_1 \gg J_2$, the acoustical (R and S in phase) or optical (R and S opposite in phase) character of the z branches remains from Γ to Z.

The limits of the Brillouin zone in directions perpendicular to z are different in the two models, but this difference is not meaningful for the thermodynamical properties, since the corresponding magnons have very high energies. The main difference between the models lies in the packing of the planes (only RS links along z) which explains the difference between D and D', easily detected by neutron diffraction. This difference may be thought of as arising from the mixing of J_1 and J_2 in J_{RS} (see Fig. 2) in the true model, whereas in the simplified model, $J_{RR} \sim J_2$, and $J_{RS} \sim J_1$. As a final point, we note that both models exhibit a strong directional dependence of the renormalization, and that in the five-parameter model, as well as the simplified one, two effective renormalization coefficients ξ_L and ξ_T, which are k independent in the first-order theory, may be introduced. Our calculations show that this result is valid also for the optical modes.

V. CONCLUSION

Previous authors[5] were unable to explain simultaneously the magnetization and the susceptibility of $CrBr_3$ in the frame of the two-parameter model introduced in Ref. 2, even when correlation effects were taken into account. We have shown that this is not due to some failure of the spin-wave theory, and that explanation by means of this theory becomes possible when the true crystallographic structure is used. The values we have obtained for the different exchange parameters agree with the values deduced from the spin-wave spectrum as determined by neutron diffraction.

The above result is not a consequence of the large number of exchange parameters introduced in the true model, since in fact just two nonzero parameters, J_1 and J_2, are sufficient to explain the low-temperature thermodynamical properties (but not the entire spin-wave spectrum), but is due to the introduction of the true crystal structure (and its main consequence: $D' \neq D$). $CrBr_3$ is not a simple stacking of identical planes, as considered in the simplified model, and the propagation of any spin wave implies both longitudinal and transverse exchange parameters.

Finally, we note that Narath and Davis were able to explain simultaneously the magnetization and the susceptibility of $CrCl_3$[14] in the ferromagnetic phase with the simplified two-parameter model. According to the preceding discussion, this is due to the fact that J_L is almost zero in $CrCl_3$ and then $D' \cong D$. It is suspected, on the contrary, that the simplified model would be unable to explain the thermodynamics of CrI_3,[15] whose two-dimensional character is less marked.

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Ferromagnetic Stability and Elementary Excitations in a $S=\frac{1}{2}$ Heisenberg Ferromagnet with Dipolar Interaction by the Green's-Function Method

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The temperature dependence of the elementary excitation energy in a dipolar Hamiltonian has been studied by the Green's-function equation-of-motion method. The result differs from that given by Charap and reduces to it for $B=F=0$. The magnetization calculated at zero order of an iterative procedure coincides with the Holstein-Primakoff result. The criterion of stability of the ferromagnetic state has been found to be temperature dependent.

I. INTRODUCTION

Several models have been introduced in order to explain the properties of $3d$ electrons in magnetic solids in connection with the concept of itinerant versus localized nature of these electrons. Considerable interest has also been shown in recent years by experimentalists on the reliability of these models. In particular an extensive effort has been made in two directions: measuring the magnon dispersion law for a wide range of momentum transfer and studying their temperature dependence.[1]

The dispersion law has been studied as a function of the temperature for various materials with different experimental methods such as spin-wave resonance,[2] neutron diffraction, energy analysis by triple-axis spectrometry, and small-angle scattering techniques.[3] Since the temperature dependence is different in different models, we can distinguish among them.

Theoretically the temperature dependence of the magnon dispersion law in the Heisenberg model has been found by a perturbation method[4] and by solving temperature-dependent Green's-function equations of motion in some approximation.[5]

In this paper we consider anisotropy effects. We assume a more general Hamiltonian as a sum of the Heisenberg and dipolar terms:

$$H=H_e+H_d\ ,$$

where

$$\begin{aligned} H_e&=-\sum_{l m} J_{lm}\vec{S}_l\cdot\vec{S}_m+2\mu_B\mathcal{H}\sum_l S_l^z\ ,\\ H_d&=\sum_{l>m}\tfrac{1}{2}d_{lm}[\vec{S}_l\cdot\vec{S}_m-3r_{lm}^{-2}(\vec{S}_l\cdot\vec{r}_{lm})(\vec{S}_m\cdot\vec{r}_{lm})]\ . \end{aligned} \tag{1}$$

Magnetic quadrupole interactions, also invoked to explain ferromagnetic anisotropy, are neglected; in fact their ratio to the dipole-dipole interaction need only be $\sim\frac{1}{10}$.[6] We can express H_d in terms of raising and lowering operators:

$$H_d=H_d^0+H_d^++H_d^-+H_d^{++}+H_d^{--}\ ,$$

where

$$H_d^0=\sum_{l\neq m}E_{lm}(\vec{S}_l\cdot\vec{S}_m-3S_l^zS_m^z)\ ,$$

$$H_d^+=\sum_{l\neq m}F_{lm}S_l^+S_m^z\ ,$$

$$H_d^-=\sum_{l\neq m}F_{lm}^*S_l^-S_m^z\ ,$$