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Classification

Physics Abstracts

1.650 — 8.534 — 8.536

# QUANTUM THEORY OF ONE- AND TWO-DIMENSIONAL FERRO- AND ANTIFERROMAGNETS WITH AN EASY MAGNETIZATION PLANE

## I. IDEAL 1-D OR 2-D LATTICES WITHOUT IN-PLANE ANISOTROPY

J. VILLAIN

Institut Max-von-Laue-Paul-Langevin, BP 156, 38042 Grenoble-Cedex, France

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**Résumé.** — Nous introduisons une représentation dite « semi-polaire » des opérateurs de spin, qui permet, dans l'approximation harmonique, de définir des magnons de n'importe quelle longueur d'onde à basse température dans les systèmes magnétiques à 1 ou 2 dimensions dépourvus d'ordre à longue distance, à condition qu'ils aient un plan de facile aimantation (système « planaire »).

Nous utilisons la représentation semi-polaire pour calculer à basse température la fonction de corrélation de spins. Sa transformée de Fourier spatio-temporelle (directement observable par diffusion des neutrons), comporte un pic relativement large dû aux fluctuations des spins dans le plan de facile aimantation, et un pic plus étroit dû aux fluctuations hors-plan. Nous calculons, pour toutes valeurs du transfert d'impulsion, l'intensité, la largeur et la forme des 2 pics dans le cas à une dimension aussi bien qu'à 2 dimensions, ainsi que le déplacement de la fréquence avec la température.

**Abstract.** — A « semi-polar » representation of the spin operators is introduced, which makes possible, in the harmonic approximation, the definition of magnons for any wavelength at low temperature in one-dimensional (= 1-D) or two-dimensional (= 2-D) magnetic systems without long-range order, provided they are of the « planar » type, i. e. they have an easy magnetization plane.

The semi-polar representation is used to calculate the spin pair correlation function at low temperature. Its space-time Fourier transform (directly observable by neutron scattering) consists of a relatively broad peak due to spin fluctuations inside the easy plane, plus a narrower peak due to out-of-plane fluctuations. The intensity, width and lineshape of both peaks are calculated in both 1-D and 2-D cases for all momentum transfers, as well as the frequency shift as a function of temperature.

### Sommaire

1. INTRODUCTION. — Le modèle ; exemple expérimental.

2. LA TRANSFORMATION. — Dans un système magnétique avec plan de facile aimantation, une transformation du genre Holstein-Primakoff peut être faite même en l'absence d'ordre à grande distance. Il en résulte une extension aux cas quantiques des calculs faits par d'autres auteurs dans le cas classique à  $t = 0$ .

3. L'APPROXIMATION HARMONIQUE. — On montre que le concept de magnon peut avoir un sens même dans certains milieux magnétiquement désordonnés.

4. DOMAINE DE VALIDITÉ DE L'APPROXIMATION HARMONIQUE. — Nous n'avons été capable de justifier l'approximation harmonique que pour des valeurs  $s$  du spin très supérieures à 1 ; il y a cependant des raisons de croire qu'elle est encore acceptable pour

### Summary

1. INTRODUCTION. — The model ; experimental example.

2. THE TRANSFORMATION. — In a magnetic system with an easy magnetization plane, a transformation in the Holstein-Primakoff style can be carried out, even if there is no long range order. This allows for a quantum extension of the classical, zero-time calculations of other authors.

3. THE HARMONIC APPROXIMATION. — It is shown that magnons can be defined even in certain magnetically disordered media.

4. DOMAIN OF VALIDITY OF THE HARMONIC APPROXIMATION. — We have only been able to justify the harmonic approximation for large values of the spin  $s$  ; however, there are arguments to believe that it is still acceptable for lower values ; on the other hand, cal-

$s$  petit ; d'autre part, dans la limite des grands spins, notre méthode a l'intérêt de fournir une vérification de calculs basés sur d'autres techniques.

5. APPROXIMATION HARMONIQUE SELF-CONSISTENTE (SCHA). — C'est l'amélioration la plus simple de l'approximation harmonique. Elle ne donne pas d'amortissement, mais seulement un déplacement de raie de type Hartree.

6. FONCTION DE CORRÉLATION SPIN-SPIN. — Des formules générales sont données dans le cadre de l'approximation harmonique.

7. PROPRIÉTÉS DANS L'ÉTAT FONDAMENTAL. — Pour  $D = 2$  il n'y a qu'une réduction de spin dont le calcul ne présente pas de difficulté. Pour  $D = 1$ , il n'y a pas d'ordre à grande distance dans l'état fondamental, sauf pour  $s = \infty$  ou pour les ferromagnétiques isotropes qui ne sont pas pris en considération dans ce travail. Cependant, dans la plupart des cas usuels, on trouve une fonction de corrélation statique qui décroît très lentement avec la distance ; un cas extrême est le modèle XY avec  $s = 1/2$ , où on trouve un taux de décroissance très proche de la valeur exacte.

8. FRÉQUENCE DES MAGNONS A TEMPÉRATURE FINIE. — Dans l'approximation SCHA, le déplacement à basse température est proportionnel à  $T^{D+1}$ .

9. LA CHAÎNE LINÉAIRE A BASSE TEMPÉRATURE (FLUCTUATIONS DE GRANDE LONGUEUR D'ONDE). — La section efficace de diffusion inélastique des neutrons est donnée dans la limite classique par une formule analytique : c'est une fonction très simple de la fréquence, qui présente 4 pôles. Les magnons sont visibles dans le spectre de neutrons si  $q > \kappa$ . La principale correction quantique à la fonction de corrélation statique se présente comme une érosion des ailes de la Lorentzienne d'Ornstein et Zernike.

10. LA CHAÎNE LINÉAIRE A BASSE TEMPÉRATURE : COURTES LONGUEURS D'ONDE. — Il apparaît une différence essentielle avec le cas isotrope : outre la contribution à  $S(q, \omega)$  due aux fluctuations dans le plan, dont la largeur devient importante à température relativement basse, il existe des corrélations hors-plan, qui ne s'élargissent qu'à température plus élevée. La largeur du pic dans le plan doit avoir un minimum relatif en bord de zone.

11. SYSTÈMES MAGNÉTIQUES BI-DIMENSIONNELS DANS L'APPROXIMATION DES GRANDES DISTANCES OU DES TEMPS LONGS. — La principale correction quantique à la fonction de corrélation statique classique se réduit à une constante multiplicative. Les magnons apparaissent dans le spectre de neutrons pour toutes les longueurs d'onde à basse température.

12. SYSTÈMES MAGNÉTIQUES BI-DIMENSIONNELS DANS L'APPROXIMATION DES COURTES DISTANCES ET DES TEMPS COURTS. — On montre que  $S(q, \omega)$  a un pic

calculations in the large  $s$  limit provide a check of other theoretical calculations.

5. SELF-CONSISTENT HARMONIC APPROXIMATION (SCHA). — It is the simplest improvement to the harmonic approximation ; it gives a Hartree shift, but no damping.

6. SPIN-PAIR CORRELATION FUNCTION. — General formulae are given within the harmonic approximation.

7. GROUND STATE PROPERTIES. — For  $D = 2$  there is just a spin reduction which is easily calculated. For  $D = 1$ , there is no long range order in the ground state, except for  $s = \infty$  or for isotropic ferromagnets (which are not considered here). In most usual cases, however, the static correlation function is found to have a very slow decay in space ; an extreme case is the XY model with  $s = 1/2$ , where the decay rate is found to be very close to the exact value.

8. MAGNON FREQUENCY AT FINITE TEMPERATURE. — It is found in the SCHA approximation to have a shift proportional to  $T^{D+1}$ .

9. THE LINEAR CHAIN AT LOW TEMPERATURE (FLUCTUATIONS OF LONG WAVELENGTH). — An explicit formula for the neutron inelastic scattering cross section is given in the classical limit ; it is a very simple analytic function with 4 poles. Magnons show up in the neutron spectrum if  $q > \kappa$ . The main quantum correction to the classical correlation function is calculated : in the 1- $D$  case it can be described as an erosion of the wings of the Ornstein-Zernike lorentzian function.

10. THE LINEAR CHAIN AT LOW TEMPERATURE : SHORT WAVELENGTH CASE. — Here a basic difference with the isotropic case appears : besides the in-plane contribution, which becomes broad at fairly low temperatures, there is an out-of-plane correlation, which broadens at higher temperature. The width of the in-plane peak is predicted to have a relative minimum at the zone boundary.

11. TWO-DIMENSIONAL MAGNETS AT LOW TEMPERATURE IN THE LONG DISTANCE OR LONG TIME APPROXIMATION. — The first order quantum correction to the classical result is found to be a mere multiplicative constant.

Magnons are found to show up in the neutron spectrum at any wavelength, at low temperature.

12. TWO-DIMENSIONAL MAGNETS IN THE SHORT DISTANCE, SHORT TIME APPROXIMATION. — The neutron scattering function is found to have an infinite peak

infini à  $\omega = \omega_q$  à basse température, et une singularité avec dérivée infinie à température plus élevée ; ces singularités sont en fait émoussées par l'amortissement des magnons.

L'amortissement des magnons est calculé dans l'Appendice A. Il coïncide avec la largeur du pic *hors-plan*. Un calcul approximatif de cette largeur est également donné au paragraphe 10.5. Les Appendices B et C contiennent des calculs relatifs au paragraphe 11.

La présente théorie explique certains faits expérimentaux mais seules des recherches expérimentales ultérieures pourront indiquer si l'accord est tout à fait satisfaisant. La forme de raie que nous donnons dans le cas unidimensionnel semble être en désaccord avec les résultats expérimentaux et théoriques relatifs à des systèmes *isotropes* (le bord externe n'est pas abrupt selon nos calculs), et on voit mal comment ce désaccord pourrait être expliqué par l'anisotropie.

**Table of symbols.** — The reader interested in only a part of this paper can meet difficulties in understanding the symbols, when defined in another part of the paper. For this reason, most of non-standard symbols, which are used in more than one chapter, are listed below.

- $A$  : anisotropy constant, eq. (2).
- $G_{RR'}^\perp(t)$  : angular correlation function, eq. (24).
- $\tilde{G}^\perp(\mathbf{q}, t)$  : spatial Fourier transform of  $G_{RR'}^\perp(t)$ .
- $\tilde{G}^\perp(\mathbf{q}, \omega)$  : space-time Fourier transform of  $G_{RR'}^\perp(t)$ .
- $g_{RR'}^0(t), \gamma_{RR'}(t)$  : see eq. (25), (26), (27).
- $\mathcal{G}(k)$  : see eq. (13).
- $J_{RR'}$  : coupling constant, eq. (2).
- $J$  defined by  $\mathcal{G}(0) - \mathcal{G}(k) \simeq Ja^2 k^2$  for small  $k$  ;

1. **Introduction.** — In this paper will be investigated the static and dynamical properties at low temperature of a system of spins  $S_R$  localized at Bravais lattice points  $R$  and submitted to a Hamiltonian of the general form :

$$\mathcal{H} = - \sum_{RR'} J_{RR'} (S_R^x S_{R'}^x + S_R^y S_{R'}^y) - \sum_{RR'} K_{RR'} S_R^z S_{R'}^z \quad (1)$$

where the  $xOy$  plane is assumed to be an *easy* magnetization plane. This clearly implies some inequality between the  $J$ 's and the  $K$ 's. The case generally considered in this paper is :

$$J_{RR'} = K_{RR'} \quad (R \neq R'), \quad J_{RR} = 0, \quad -K_{RR} = A > 0$$

so that the Hamiltonian becomes :

$$\mathcal{H} = - \sum_{RR'} J_{RR'} S_R \cdot S_{R'} + A \sum_R S_R^z{}^2 \quad (2)$$

In this case the modulus  $s$  of the spins must of course be different from  $1/2$ .

Another case is the « anisotropic Heisenberg model » corresponding to

$$J_{RR} = K_{RR} = 0,$$

at the magnon frequency at low temperature, and a singularity with an infinite derivative at higher temperature.

In Appendix A is calculated (in the ferromagnetic case) the damping of the magnons, which coincides with the width of the *out-of plane* peak. A rough calculation is also given in paragraph 10.5. Appendices B and C contain calculations related to paragraph 11.

The present theory explains certain experimental facts but further experimental work is necessary to decide whether there is a good agreement or not. Our lineshape in the 1- $D$  case disagrees with both experimental and theoretical results in the *isotropic* case (the outer edge is not sharp according to our calculation), and it is not clear that this can be an effect of the anisotropy.

$J > 0$  for ferromagnets,  $J < 0$  for antiferromagnets.

$\mathfrak{J}(k)$  : FT of  $J_{RR'}$ , eq. (13).

$f_{RR'}(t)$  : see eq. (28) and (29).

$1/\kappa$  : correlation length.

$\mathbf{R}$  : lattice site.

$\mathbf{r} = \mathbf{R}' - \mathbf{R}$ .

$S_R$  : spin operator at  $\mathbf{R}$ .

$s$  :  $(S_R)^2 = s(s+1)$ .

$\varphi_R$  : semi-polar angle of  $S_R$ , eq. (4).

$\varepsilon_R, \psi_R$  : see following eq. (10).

$\tau$  = characteristic time defined by (58), when dealing with 1- $D$  systems.

$\tau$  = reduced temperature defined by (74), when dealing with 2- $D$  systems.

a special case of which is, for  $K_{RR'} = 0$ , the XY model :

$$\mathcal{H} = - \sum_{RR'} J_{RR'} (S_R^x S_{R'}^x + S_R^y S_{R'}^y). \quad (3)$$

The models considered here have no long range order [1], [2], [3] in the case of a lattice of dimensionality  $D = 1$  or 2. For  $D = 1$  as well, there is no long range order in the ground state, except for  $A = 0$  or  $s = \infty$  (classical limit). However, the correlation length is large in both cases [2], [5], [6], [7] at low temperature. Moreover for  $D = 2$ , there is a phase transition without long range order [2], [7], [8], [9], below which the spin pair correlation decays as some power of the distance  $r$ , rather than exponentially.

These properties are true for the isotropic case  $A = 0$  as well. However, the mathematical treatment is much easier for  $A > 0$ . A very good example of model (2) is  $\text{CsNiF}_3$ , which has been recently investigated by Steiner and Dorner [10], [11] and has a linear structure (i. e.  $D = 1$ ).

In the present paper will be considered ideal one-dimensional (= 1- $D$ ) or two-dimensional (= 2- $D$ )

systems with no in-plane anisotropy. Real systems will be treated in a subsequent paper.

2. **The transformation.** — Conventional representations of the spin operators are not adequate in the absence of long range order.

Therefore, use will be made of the following representation of the spin operators :

$$\begin{cases} S_{\mathbf{R}}^+ = e^{i\varphi_{\mathbf{R}}} \sqrt{(s + \frac{1}{2})^2 - (S_{\mathbf{R}}^z + \frac{1}{2})^2} \\ S_{\mathbf{R}}^- = \sqrt{(s + \frac{1}{2})^2 - (S_{\mathbf{R}}^z + \frac{1}{2})^2} e^{-i\varphi_{\mathbf{R}}} \end{cases} \quad (4)$$

This representation is a generalization of an obvious equality for classical spins ( $s = \infty$ ) ; then the angle  $\varphi_{\mathbf{R}}$  is a semi-polar coordinate. For finite  $s$ ,  $\varphi_{\mathbf{R}}$  is a hermitian operator defined by :

$$\begin{cases} [\varphi_{\mathbf{R}}, S_{\mathbf{R}'}^z] = i\delta_{\mathbf{R}\mathbf{R}'} \\ [\varphi_{\mathbf{R}}, \varphi_{\mathbf{R}'}] = 0 \\ \varphi_{\mathbf{R}} = \varphi_{\mathbf{R}}^* \end{cases} \quad (5)$$

A solution of (5) is :

$$\varphi_{\mathbf{R}} = \varphi_{\mathbf{R}}^* = i \frac{\partial}{\partial S_{\mathbf{R}}^z} \quad (6)$$

From (5) can be deduced the following commutation relation :

$$[S_{\mathbf{R}}^z, e^{\pm i m \varphi_{\mathbf{R}}} ] = \pm m e^{\pm i m \varphi_{\mathbf{R}}} \quad (7)$$

Using (7), it is easy to check that the operators defined by (4) verify the following 3 relations :

$$\begin{aligned} [S_{\mathbf{R}}^z, S_{\mathbf{R}'}^{\pm}] &= \pm \delta_{\mathbf{R}\mathbf{R}'} S_{\mathbf{R}'}^{\pm}, \quad [S_{\mathbf{R}}^+, S_{\mathbf{R}'}^-] = 2 \delta_{\mathbf{R}\mathbf{R}'} S_{\mathbf{R}}^z \\ \{ S_{\mathbf{R}}^+, S_{\mathbf{R}}^- \} + 2 S_{\mathbf{R}}^z &= 2 s(s + 1). \end{aligned}$$

And this proves that (4) is a representation of the spin operators. A quite similar representation has been used by Berezinskii [15] for Bose operators.

Making use of (4), the Hamiltonian (2) can be written as follows :

$$\begin{aligned} \mathcal{H} = - \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \sqrt{(s + \frac{1}{2})^2 - (S_{\mathbf{R}}^z + \frac{1}{2})^2} e^{i(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})} \times \\ \times \sqrt{(s + \frac{1}{2})^2 - (S_{\mathbf{R}'}^z + \frac{1}{2})^2} \\ - \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} S_{\mathbf{R}}^z S_{\mathbf{R}'}^z + A \sum_{\mathbf{R}} S_{\mathbf{R}}^z \end{aligned} \quad (8)$$

whereas the XY Hamiltonian is obtained if the last 2 terms of (8) are omitted.

It is important to note that eq. (5) and (6) only make sense if  $\varphi_{\mathbf{R}}$  and  $S_{\mathbf{R}}^z$  are continuous variables, similar to the abscissa and momentum of a harmonic oscillator. However,  $\varphi_{\mathbf{R}}$  appears in (4) through its exponential  $e^{i\varphi_{\mathbf{R}}}$ , which, in contrast with  $\varphi_{\mathbf{R}}$  itself, conserves the discrete set of eigenvectors of  $S_{\mathbf{R}}^z$  for integer (or half-integer) eigenvalues. Eq. (4) and (8) make sense inside the subspace  $|S_{\mathbf{R}}^z| \leq s$  of this set.

This is related to the « kinematic » interaction of spin waves [16].

3. **The harmonic approximation.** — Similar to the Holstein-Primakoff transformation, expression (8) is rigorous, but not very useful unless approximations are made. A good approximation is easy to find at low temperature if  $s$  is large and if  $A$  lies in some interval, as is shown in paragraph 4.

As  $A$  is positive, classical spins clearly satisfy at low temperature the inequality :

$$S_{\mathbf{R}}^z \ll s. \quad (9)$$

What about the  $\varphi$ 's ? Assume a nearest-neighbour interaction  $J$  (although this is not very important) ; then, if  $J > 0$  (ferromagnetic case), one clearly has :

$$\begin{aligned} J_{\mathbf{R}\mathbf{R}'} e^{i(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})} &\simeq \\ &\simeq J_{\mathbf{R}\mathbf{R}'} [1 + i(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'}) - \frac{1}{2}(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})^2]. \end{aligned}$$

We shall restrict our investigation of antiferromagnets to those cases where the lattice naturally splits into 2 sublattices (determined by the condition that the first neighbour interaction  $J_{\mathbf{R}\mathbf{R}'}$  is zero if  $\mathbf{R}$  and  $\mathbf{R}'$  belong to the same sublattice) ; this excludes, for instance, the triangular lattice. Then :

$$\begin{aligned} J_{\mathbf{R}\mathbf{R}'} e^{i(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})} &\simeq \\ &\simeq J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} [1 + i(\psi_{\mathbf{R}} - \psi_{\mathbf{R}'}) - \frac{1}{2}(\psi_{\mathbf{R}} - \psi_{\mathbf{R}'})^2] \end{aligned} \quad (10)$$

where  $\varepsilon_{\mathbf{R}} = 1$  on one sublattice,  $-1$  on the other,  $\psi_{\mathbf{R}} = \varphi_{\mathbf{R}}$  on one sublattice and  $\pi + \varphi_{\mathbf{R}}$  on the other.

Inserting (10) into (8), expanding the square root according to (9) and retaining the terms of second degree, one obtains a harmonic Hamiltonian :

$$\begin{aligned} \mathcal{H} = -s^2 \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} + \frac{1}{2} s^2 \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} (\psi_{\mathbf{R}} - \psi_{\mathbf{R}'})^2 + \\ + \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} S_{\mathbf{R}}^z S_{\mathbf{R}'}^z - \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} S_{\mathbf{R}}^z S_{\mathbf{R}'}^z + A \sum_{\mathbf{R}} S_{\mathbf{R}}^z \end{aligned} \quad (11)$$

Only the terms of highest degree in  $s$  have been retained, since approximations (9) and (10) are only correct in the classical limit  $s = \infty$ .

This expression also holds for ferromagnets with  $\varepsilon_{\mathbf{R}} = 1$  and  $\psi_{\mathbf{R}} = \varphi_{\mathbf{R}}$ . If there are non-nearest-neighbor interactions, the appropriate division into sublattices can be determined from the classical (Néel) ground state or from experiment.

The Hamiltonian (11) can easily be diagonalized by a Fourier transformation <sup>(1)</sup> :

$$\begin{aligned} \mathcal{H} = s^2 \sum_{\mathbf{k}} [\mathcal{G}(0) - \mathcal{G}(\mathbf{k})] \psi_{\mathbf{k}}^* \psi_{\mathbf{k}} + \\ + \sum_{\mathbf{k}} [A + \mathcal{G}(0) - \mathcal{J}(\mathbf{k})] S_{-\mathbf{k}}^z S_{\mathbf{k}}^z + C^t \end{aligned} \quad (12)$$

(1) All  $k$ -summations are over the first Brillouin zone ; therefore each term in (12) appears twice ( $+k$  and  $-k$ ). From this fact arises the factor 2 in (16).

with :

$$\psi_k = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} \psi_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}}, \quad S_k^z = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} S_{\mathbf{R}}^z e^{i\mathbf{k} \cdot \mathbf{R}}$$

$$\mathfrak{G}(\mathbf{k}) = \sum_{\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} e^{i\mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}, \quad \mathfrak{J}(\mathbf{k}) = \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} e^{i\mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}. \quad (13)$$

It is easy [17] to derive from (12) the following correlation functions :

$$\begin{aligned} & \langle \psi_k^* \psi_k(t) \rangle = \\ & = \frac{1}{s} \sqrt{\frac{A + \mathfrak{G}(0) - \mathfrak{J}(\mathbf{k})}{\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})}} \left( \frac{1}{2} e^{i\omega_k t} + \frac{\cos \omega_k t}{e^{\beta \hbar \omega_k} - 1} \right) \quad (14) \end{aligned}$$

$$\begin{aligned} & \langle S_{-k}^z S_k^z(t) \rangle = \\ & = s \sqrt{\frac{\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})}{A + \mathfrak{G}(0) - \mathfrak{J}(\mathbf{k})}} \left( \frac{1}{2} e^{i\omega_k t} + \frac{\cos \omega_k t}{e^{\beta \hbar \omega_k} - 1} \right) \quad (15) \end{aligned}$$

with the following dispersion relation :

$$\hbar \omega_k = 2s \sqrt{[\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})] [A + \mathfrak{G}(0) - \mathfrak{J}(\mathbf{k})]}. \quad (16)$$

**4. Domain of validity of the harmonic approximation.** — The approximations made in paragraph 3 amount to three :

i) The condition  $S_{\mathbf{R}}^z \leq s$  has been relaxed. This approximation is also made in the Holstein-Primakoff formalism. The present case implies :

$$\langle (S_{\mathbf{R}}^z)^2 \rangle \ll s^2. \quad (17a)$$

ii) The Taylor expansions (10) and (11). Approximation (10) is correct if :

$$\frac{1}{2} \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} \langle (\psi_{\mathbf{R}} - \psi_{\mathbf{R}'})^2 \rangle \ll \mathfrak{G}(0). \quad (17b)$$

This condition has been written without taking into account linear terms in (10), as they do not contribute to (11).

The approximate Hamiltonian (11) is correct if both (17a) and (17b) are satisfied.

iii) The condition that  $S_{\mathbf{R}}^z$  should be integer (or half integer) has been relaxed.

Consider  $S_{\mathbf{R}}^z$  as the momentum of a particle labelled by  $\mathbf{R}$ , and  $\psi_{\mathbf{R}}$  as its abscissa. The condition  $S_{\mathbf{R}}^z = \text{integer}$  means that the wave function (as a function of  $\psi_{\mathbf{R}}$ ) should be periodic, which means physically that the particle moves in a circle. The case  $S_{\mathbf{R}}^z = \text{half-integer}$  can be investigated in a similar way (for instance, by translating the origin in the momentum space).

Now, if the condition  $S_{\mathbf{R}}^z = \text{integer}$  is relaxed, the eigenfunctions of (12) are square integrable and therefore not periodic. However, for values of  $T$  and  $s$  such that physically significant (i. e., with an appreciable probability  $e^{-\beta E/Z}$ ) wave functions satisfy

(17b), there is a one-to-one correspondence between physically significant square integrable functions and periodic functions : indeed, to any periodic function  $F(\psi_1, \psi_2, \dots, \psi_N)$  satisfying  $|\psi_{\mathbf{R}} - \psi_{\mathbf{R}'}| \approx 2\pi n$  for  $\mathbf{R}, \mathbf{R}'$  neighbours, can be associated a square integrable function equal to  $F$  if  $|\psi_{\mathbf{R}} - \psi_{\mathbf{R}'}| < \pi$  for  $\mathbf{R}, \mathbf{R}'$  neighbours, and to zero otherwise. This is clearly a one-to-one correspondence and it can be seen that physically significant eigenfunctions of (12) correspond to approximate eigenfunctions of (8).

To summarize, the harmonic approximation is valid if both conditions (17) are satisfied.

Insertion of (14), (15) and (16) into (17) yields at  $T = 0$  :

$$\begin{cases} \frac{1}{2Ns^2} \sum_k \hbar \omega_k \ll \mathfrak{G}(0) \\ \frac{1}{2Ns} \sum_k \sqrt{\frac{\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})}{A + \mathfrak{G}(0) - \mathfrak{J}(\mathbf{k})}} \ll 1. \end{cases}$$

Strictly speaking, this leads to the very disappointing condition (ferromagnetic case) :

$$J/s^2 \ll A \ll Js^2 \quad (18)$$

which is rarely satisfied, as very large values of  $s$  are not physically available.

The case  $A > Js^2$ , which is of interest for rare earths, will be considered first. This condition means that the exchange energy is a perturbation with respect to the anisotropy. i) If  $s$  is an integer, the unperturbed ground state is non-degenerate and therefore the true ground state is not ordered. ii) If  $s$  is half-integer, the unperturbed ground state is  $2^N$  times degenerate, because it is defined by  $S_{\mathbf{R}}^z = \pm 1/2$ . It is then easy to show that the Hamiltonian (1) reduces to an anisotropic Heisenberg Hamiltonian with  $s = 1/2$ . The conclusion is that the case  $A > Js^2$  can be discarded and must be treated by completely different methods.

If  $A < Js^{-2}$ , it turns out that many of the results derived below remain correct to a good approximation, although it is not easy to explain why. Our theory only gives the quantum correction of first order in  $1/s$  ; as this correction generally turns out to be small (see § 7), it is expected that higher corrections are also small, and the foregoing theory will therefore be applicable to materials which do not satisfy (18), and especially to  $\text{CsNiF}_3$ , where  $A \approx Js^2/2$ .

It is however of interest to improve the harmonic approximation ; this is done in the next chapter, but the resulting theory is so cumbersome that we shall in many cases be content with the harmonic approximation.

**5. Self consistent harmonic approximation (SCHA).**

— The Hamiltonian (8) will be approximated by a temperature-dependent hamiltonian :

$$\tilde{\mathcal{H}} = \sum_k [a_k S_{-k}^z S_k^z + b_k \psi_{-k} \psi_k] \quad (19)$$

determined by means of the following property :

Let  $F(\xi_1, \xi_2, \xi_3, \xi_4)$  be any analytic function of

$$\xi_1 = \psi_{\mathbf{R}}, \quad \xi_2 = \psi_{\mathbf{R}'}, \quad \xi_3 = S_{\mathbf{R}}^z, \quad \xi_4 = S_{\mathbf{R}'}^z.$$

If the system is large and if the Hamiltonian has the form (13'), the function

$$\tilde{F}(\{\xi_i\}) = F(0) + \sum_i \xi_i \left\langle \frac{\partial F}{\partial \xi_i} \right\rangle + \frac{1}{2} \sum_{ij} \left\langle \frac{\partial^2 F}{\partial \xi_i \partial \xi_j} \right\rangle \xi_i \xi_j \quad (20)$$

satisfies the following relations, where  $\xi, \xi' = S_{\mathbf{R}''}^z$  or  $\psi_{\mathbf{R}''}$  and  $\mathbf{R}''$  is any lattice point :

$$\left\{ \begin{array}{l} \langle \tilde{F} \rangle = \langle F \rangle \\ \langle \xi \tilde{F} \rangle = \langle \xi F \rangle \\ \langle \xi \xi' \tilde{F} \rangle = \langle \xi \xi' F \rangle. \end{array} \right. \quad (21)$$

Thus,  $\tilde{F}$  can be considered as a good approximation of  $F$ .

*Proof.* — Since  $F$  is analytic, it is sufficient to prove the property for a product of powers of the  $\xi_i$ 's :

$$F(\{\xi_i\}) = \xi_1^{n_1} \xi_2^{n_2} \xi_3^{n_3} \xi_4^{n_4}.$$

This function can be written as a sum of products of  $n$  operators  $S_k^z$  and  $\psi_k$ , where

$$n = \sum_i n_i \cdot \xi F, \quad \xi \xi' F, \quad \xi \tilde{F} \quad \text{and} \quad \xi \xi' \tilde{F}$$

are also the sums of such products of  $(n+1)$  or  $(n+2)$  operators ; when taking the mean value, the dominant contribution for a large system comes from terms where  $\mathbf{k}$  takes the values  $\mathbf{k}_1, -\mathbf{k}_1, \mathbf{k}_2, -\mathbf{k}_2, \dots$ , with all  $\mathbf{k}_i$ 's different. Closer inspection shows that eq. (21) follow from this fact. Of course the second term of (20) vanishes if  $n$  is even, and the 3rd term vanishes if  $n$  is odd.

The approximate Hamiltonian (19) can now be determined from (20) with  $F = \mathcal{H}$ , defined by (8), and where all mean values  $\langle A \rangle$  are replaced by

$$\langle A \rangle \sim \frac{1}{\tilde{Z}} \text{Tr } A e^{-\beta \tilde{\mathcal{H}}}$$

with

$$\tilde{Z} = \text{Tr } e^{-\beta \tilde{\mathcal{H}}}.$$

It is readily seen that :

$$\left\langle \frac{\partial \mathcal{H}}{\partial \xi_i} \right\rangle \sim 0.$$

Indeed both  $\mathcal{H}$  and  $\tilde{\mathcal{H}}$  are invariant through the transformation ( $S^z \rightarrow -S^z, \varphi \rightarrow -\varphi$ ) which therefore changes  $\partial \mathcal{H} / \partial \xi_i$  into  $-\partial \mathcal{H} / \partial \xi_i$ .

It is somewhat more difficult to prove that

$$\left\langle \frac{\partial^2 \mathcal{H}}{\partial S_{\mathbf{R}}^z \partial \varphi_{\mathbf{R}'}} \right\rangle \sim 0 \quad (22)$$

The left hand side is a sum of terms of the form :

$$\langle S_{\mathbf{R}}^{z_p} \psi_{\mathbf{R}'}^m \psi_{\mathbf{R}}^n S_{\mathbf{R}'}^{z_q} - (-1)^{m+n} S_{\mathbf{R}'}^{z_q} \psi_{\mathbf{R}}^n \psi_{\mathbf{R}'}^m S_{\mathbf{R}}^{z_p} \rangle$$

times a coefficient.

i) Terms with  $m+n+p+q$  odd give a negligible contribution proportional to  $1/\sqrt{N}$ .

ii) Terms with  $(p+q)$  and  $(m+n)$  even vanish because  $\mathcal{H}$  is invariant under the transformation ( $S^z \rightarrow -S^z, \psi \rightarrow -\psi$ ).

iii) Terms with  $(p+q)$  and  $(m+n)$  odd are (neglecting contributions proportional to  $1/N$ ) sums of terms of the form :

$$\begin{aligned} & \langle S_{-\mathbf{k}_1}^z S_{\mathbf{k}_1}^z \rangle \langle S_{-\mathbf{k}_2}^z S_{\mathbf{k}_2}^z \rangle \cdots \langle S_{-\mathbf{k}_\mu}^z S_{\mathbf{k}_\mu}^z \rangle \times \\ & \quad \times \langle \psi_{-\mathbf{k}_1} \psi_{\mathbf{k}_1} \rangle \cdots \langle \psi_{-\mathbf{k}'_\nu} \psi_{\mathbf{k}'_\nu} \rangle \\ & \langle S_{-\mathbf{k}'_1}^z \psi_{\mathbf{k}'_1} \rangle \cdots \langle S_{-\mathbf{k}'_\rho}^z \psi_{\mathbf{k}'_\rho} \rangle \langle \psi_{-\mathbf{k}'_1} S_{\mathbf{k}'_1}^z \rangle \cdots \\ & \quad \cdots \langle \psi_{-\mathbf{k}'_\sigma} S_{\mathbf{k}'_\sigma}^z \rangle + h.c \end{aligned}$$

times an imaginary constant.  $(\rho + \sigma)$  should be odd because  $p+q = 2\mu + \rho + \sigma$ . Now :

$$\langle S_{-\mathbf{k}}^z \psi_{\mathbf{k}} \rangle = - \langle \psi_{-\mathbf{k}} S_{\mathbf{k}}^z \rangle$$

so that terms of class (iii) also vanish.

The formal calculation of the remaining derivatives,

$$\frac{\partial^2 \mathcal{H}}{\partial \varphi_{\mathbf{R}} \partial \varphi_{\mathbf{R}'}} \quad \text{and} \quad \frac{\partial^2 \mathcal{H}}{\partial S_{\mathbf{R}}^z \partial S_{\mathbf{R}'}^z},$$

is straightforward, but the explicit calculation of their mean values as functions of  $a_k$  and  $b_k$  is difficult, unless additional approximations are made ; these approximations consist of a few factorizations, and finally the self-consistent equations which determine  $a_k$  and  $b_k$  are (dropping the tildes) :

$$b_k = \sum_{\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} \langle (s + \frac{1}{2})^2 - (S_{\mathbf{R}}^z + \frac{1}{2})^2 \rangle \langle \cos(\psi_{\mathbf{R}'} - \psi_{\mathbf{R}}) \rangle [1 - \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})] \quad (23a)$$

$$a_k = A - \mathcal{J}(\mathbf{k}) + \sum_{\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} \frac{\langle (s + \frac{1}{2})^2 - \frac{1}{2}(S_{\mathbf{R}}^z + \frac{1}{2})^2 \rangle}{\langle (s + \frac{1}{2})^2 - (S_{\mathbf{R}}^z + \frac{1}{2})^2 \rangle} \langle \cos(\psi_{\mathbf{R}'} - \psi_{\mathbf{R}}) \rangle \quad (23b)$$

$$\langle S_{\mathbf{R}}^{z^2} \rangle = \frac{1}{2N} \sum_{\mathbf{k}} \sqrt{\frac{b_k}{a_k}} \coth \frac{1}{2} \beta \hbar \omega_k \quad (23c)$$

$$\langle \cos(\psi_{\mathbf{R}'} - \psi_{\mathbf{R}}) \rangle = \exp - \frac{1}{2N} \sum_{\mathbf{k}} [1 - \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})] \sqrt{\frac{a_k}{b_k}} \coth \frac{1}{2} \beta \hbar \omega_k \quad (23d)$$

$$\hbar \omega_k = 2 \sqrt{a_k b_k}. \quad (23e)$$

To establish (23d), use has been made of the fact that the probability distribution for  $S_k^z$ ,  $\psi_k$  is gaussian [18].

The method indicated above can be shown to be equivalent to the use of the Bogoljubov variational principle, which determines  $\tilde{\mathcal{H}}$  by minimizing

$$- \text{Log Tr } e^{-\beta \tilde{\mathcal{H}}} + \beta \langle \mathcal{H} - \tilde{\mathcal{H}} \rangle \sim .$$

**6. Spin pair correlation function.** — The longitudinal part is readily given by (15). The most important part, however, is the transverse part, which according to (4) can be written as follows :

$$\begin{aligned} \langle S_{\mathbf{R}}^x S_{\mathbf{R}'}^x(t) \rangle + \langle S_{\mathbf{R}}^y S_{\mathbf{R}'}^y(t) \rangle &= \\ &= \frac{1}{2} \langle S_{\mathbf{R}}^+ S_{\mathbf{R}'}^-(t) + S_{\mathbf{R}}^- S_{\mathbf{R}'}^+(t) \rangle \simeq \langle (s + \frac{1}{2})^2 - (S_{\mathbf{R}}^z + \frac{1}{2})^2 \rangle \langle \cos(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'}(t)) \rangle . \end{aligned}$$

This expression results from a factorization approximation combined with a maximum simplification of the factor depending on  $S^z$ . Clearly the second factor is most interesting ; using the fact the probability distribution is gaussian, it can be written [18] as :

$$\begin{aligned} G_{\mathbf{R}\mathbf{R}'}^{\perp}(t) &\equiv \langle \cos(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'}(t)) \rangle = \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} \langle \cos[\psi_{\mathbf{R}} - \psi_{\mathbf{R}'}(t)] \rangle = \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} \exp - \frac{1}{2} \langle [\psi_{\mathbf{R}} - \psi_{\mathbf{R}'}(t)]^2 \rangle \\ &= \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} \exp - \frac{1}{N} \sum_{\mathbf{k}} [\langle \psi_{\mathbf{k}}^* \psi_{\mathbf{k}} \rangle - \langle \psi_{\mathbf{k}}^* \psi_{\mathbf{k}}(t) \rangle \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})] . \end{aligned} \quad (24)$$

It turns out that the quantitative application of the SCHA approximation is difficult ; the explicit form of (24) will therefore be given in the harmonic approximation of paragraph 3 :

$$G_{\mathbf{R}\mathbf{R}'}^{\perp}(t) = \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} g_{\mathbf{R}\mathbf{R}'}^0(t) \gamma_{\mathbf{R}\mathbf{R}'}(t) \quad (25)$$

with :

$$g_{\mathbf{R}\mathbf{R}'}^0(t) = \exp - \frac{1}{2sN} \sum_{\mathbf{k}} [1 - e^{i\omega_{\mathbf{k}}t} \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})] \sqrt{\frac{A + \mathcal{G}(0) - \mathcal{J}(\mathbf{k})}{\mathcal{G}(0) - \mathcal{G}(\mathbf{k})}} \quad (26)$$

$$\gamma_{\mathbf{R}\mathbf{R}'}(t) = \exp - \frac{1}{sN} \sum_{\mathbf{k}} \frac{1 - \cos \omega_{\mathbf{k}} t \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}{e^{\beta \hbar \omega_{\mathbf{k}}} - 1} \sqrt{\frac{A + \mathcal{G}(0) - \mathcal{J}(\mathbf{k})}{\mathcal{G}(0) - \mathcal{G}(\mathbf{k})}} . \quad (27)$$

The quantity of interest in many cases, especially in neutron scattering, is the Fourier transform of (25) :

$$\tilde{G}^{\perp}(\mathbf{q}, t) = \sum_{\mathbf{R}} e^{-i\mathbf{q} \cdot \mathbf{R}} e^{-f_{\text{OR}}(t)} \varepsilon_{\mathbf{R}} \quad (28)$$

with :

$$f_{\mathbf{R}\mathbf{R}'}(t) = \frac{1}{sN} \sum_{\mathbf{k}} \left[ \frac{1 - e^{i\omega_{\mathbf{k}}t} \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}{2} + \frac{1 - \cos \omega_{\mathbf{k}} t \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}{e^{\beta \hbar \omega_{\mathbf{k}}} - 1} \right] \sqrt{\frac{A + \mathcal{G}(0) - \mathcal{J}(\mathbf{k})}{\mathcal{G}(0) - \mathcal{G}(\mathbf{k})}} . \quad (29)$$

It is relevant to express  $\tilde{G}^{\perp}(\mathbf{q}, t)$  in the following manner :

$$\tilde{G}^{\perp}(\mathbf{q}, t) = \frac{1}{2\mathcal{G}(0)} \sum_{\mathbf{R}\mathbf{R}'} \varepsilon_{\mathbf{R}} e^{-i\mathbf{q} \cdot \mathbf{R}} J_{\mathbf{R}\mathbf{R}'} [e^{-i\mathbf{q} \cdot (\mathbf{R}' - \mathbf{R})} e^{-f_{\text{OR}'}(t)} + \varepsilon_{\mathbf{R}'} e^{-f_{\text{OR}}(t)}] . \quad (30)$$

We now replace  $e^{i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}')}$  in the bracket by :

$$e^{i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}')} = -\varepsilon_{\mathbf{R}'} + \varepsilon_{\mathbf{R}} + e^{i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}')} .$$

When inserted into (30), the last 2 terms of this identity give  $\tilde{G}^{\perp}(\mathbf{q}, t)$ , times something which does not depend on  $t$  ; the term  $-\varepsilon_{\mathbf{R}'}$ , together with the second term in the bracket of (30), yields a term which contains the difference  $e^{-f_{\text{OR}}(t)} - e^{-f_{\text{OR}'}(t)}$ . The result is :

$$\tilde{G}^{\perp}(\mathbf{q}, t) = \frac{1}{\mathcal{G}(0) - \mathcal{J}(\mathbf{q})} \sum_{\mathbf{R}\mathbf{R}'} e^{-i\mathbf{q} \cdot \mathbf{R}} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} [e^{-f_{\text{OR}}(t)} - e^{-f_{\text{OR}'}(t)}] .$$

The advantage of this expression is that at low temperature the variation of  $f$  is slow in space, so that the bracket can be replaced by a Taylor expansion limited to 2nd order :

$$\begin{aligned} \tilde{G}^{\perp}(\mathbf{q}, t) &= \frac{1}{\mathcal{G}(0) - \mathcal{J}(\mathbf{q})} \sum_{\mathbf{R}} e^{-i\mathbf{q} \cdot \mathbf{R}} \sum_{\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} [f_{\text{OR}'}(t) - f_{\text{OR}}(t)] e^{-f_{\text{OR}}(t)} - \frac{1}{2} \frac{1}{\mathcal{G}(0) - \mathcal{J}(\mathbf{q})} \times \\ &\quad \times \sum_{\mathbf{R}} e^{-i\mathbf{q} \cdot \mathbf{R}} \sum_{\mathbf{R}'} \varepsilon_{\mathbf{R}} \varepsilon_{\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} [f_{\text{OR}'}(t) - f_{\text{OR}}(t)]^2 e^{-f_{\text{OR}}(t)} . \end{aligned} \quad (31)$$



7. **Ground state properties.** — They are contained in the factor  $g_{RR'}^0(t)$  given by (26).

The 2- $D$  case will not be considered : (26) describes zero-point fluctuations plus a spin reduction which can be calculated by other methods as well (Dyson transformation, etc.).

Much more interesting is the 1- $D$  case : the summation in (26) exhibits a divergence at  $k = 0$  for

$$R' - R = \infty \quad \text{or} \quad t = \infty .$$

Thus long range order is destroyed in 1- $D$  systems even in the ground state for finite  $s$  and  $A \neq 0$ , or even for  $A = 0$  in the antiferromagnetic case (this last point does not result from the present paper but was already known [4]).

The expression of  $g_{RR'}^0(t)$  is given below, using (26) and (16), neglecting the imaginary part, which is finite, and using the following long wavelength approximation :

$$\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k}) \simeq |J| a^2 k^2 \quad (32)$$

where  $a$  is the interatomic distance. Then for ferromagnets :

$$g_{RR'}^0(t) \approx \left[ \sup \left( \frac{|r|}{a}, \frac{2s|t|}{\hbar} \sqrt{AJ} \right) \right]^{-\gamma} \quad (33)$$

with

$$\gamma = \frac{1}{2\pi s} \sqrt{\frac{A}{J}} \quad (34)$$

and  $r = |R' - R|$ .  $\gamma$  is generally small. For instance if  $A = J/2$ , and  $s = 1$ , then  $\gamma = 1/9$  and  $g_{RR'}^0 = 1/2$  for  $r = 510 a$ .

For antiferromagnets,  $A$  must be replaced by  $[A + \mathfrak{G}(0) - \mathfrak{J}(0)]$  and  $\gamma$  can be larger.

The 1- $D$  XY model is of special interest as a check of our method, because its static properties can be calculated exactly for  $s = 1/2$  [4], [19]. The XY model is obtained from (12) with  $A = \mathfrak{J}(k) = 0$ ; the frequency spectrum is :

$$\begin{aligned} \hbar\omega_k &= 2s \sqrt{\mathfrak{G}(0) [\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})]} = \\ &= 4s \sqrt{2} \left| J \sin \frac{ka}{2} \right|. \end{aligned} \quad (35)$$

The static correlation function at  $T = 0$  is :

$$\begin{aligned} g_{RR'}^0(0) &= \exp - \frac{1}{2Ns} \sum_{\mathbf{k}} \sqrt{\frac{\mathfrak{G}(0)}{\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})}} [1 - \cos kr] \\ &= \exp - \frac{a}{\pi s \sqrt{2}} \int_0^{\pi/a} \frac{\sin^2 kr/2}{\sin ka/2} dk \\ &\approx \left( \frac{r}{a} \right)^{-1/\pi s \sqrt{2}} . \end{aligned} \quad (36)$$

For  $s = 1/2$  the correlation function decays as  $r^{-0.45}$  in very good agreement with the rigorous

result [4], [20]  $r^{-1/2}$ , which is rather unexpected after paragraph 4.

The longitudinal correlation at  $T = t = 0$  in the linear chain decays as  $r^{-2}$  for large  $r$ , as can be seen from (15). For instance for the XY model ( $A = \mathfrak{J}(k) = 0$ ) :

$$\langle S_{-k}^z S_k^z \rangle = \frac{s}{\sqrt{2}} \left| \sin \frac{ka}{2} \right|$$

or :

$$\begin{aligned} \langle S_{\mathbf{R}}^z S_{\mathbf{R}'}^z \rangle &= \frac{a}{\pi \sqrt{2}} s \int_0^{\pi/a} \sin \frac{ka}{2} \cos kr dk = \\ &= - \frac{s \sqrt{2}}{4 \frac{r^2}{a^2} - 1} \quad (r \neq 0) . \end{aligned} \quad (37)$$

The rigorous result for  $s = 1/2$  is [4] :

$$\langle S_{\mathbf{R}}^z S_{\mathbf{R}'}^z \rangle = - \frac{1 - (-1)^{r/a}}{2 \pi^2 r^2 / a^2} \quad (r \neq 0) . \quad (38)$$

Comparison of (37) and (38) shows that for *small* spins our description of long wavelength phenomena is qualitatively correct but our method fails to account for short wavelength features.

8. **Magnon frequency at finite temperature.** — In this chapter the SCHA approximation (§ 5) is used to show that the magnon frequency in the long wavelength limit is of the form :

$$\omega_k \simeq C(T) k \simeq (B - B' T^{D+1}) k \quad (39)$$

in agreement with 3-dimensional results [21], [22] for antiferromagnets (of course eq. (39) does not apply to isotropic ferromagnets) but in disagreement with the classical result [12] that  $C \simeq B - B' T$ .

To prove this, eq. (23c) and (23d) will be approximated as follows :

$$\langle S_{\mathbf{R}}^{z^2} \rangle \simeq \frac{K_B T}{2N} \sum_{k < k_c} \frac{1}{a_k} + \frac{1}{2N} \sum_k \sqrt{\frac{b_k}{a_k}} \quad (40)$$

$$\begin{aligned} \langle \cos(\psi_{\mathbf{R}'} - \psi_{\mathbf{R}}) \rangle &\simeq \\ &\simeq \left( \exp - \frac{K_B T}{2N} \sum_{k < k_c} \frac{1 - \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}{b_k} \right) \times \\ &\quad \times \exp - \sum_k \frac{1 - \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}{2N} \sqrt{\frac{a_k}{b_k}} \end{aligned} \quad (41)$$

where the cut-off  $k_c$  is defined at low temperature by :

$$\hbar\omega_{k_c} = K_B T . \quad (42)$$

The summations in (40) and (41) run over the Brillouin zone when not specified otherwise. The case of nearest-neighbour interactions only will be consi-

dered ; if  $\mathbf{R}$  and  $\mathbf{R}'$  are nearest-neighbours, (41) can be written as follows :

$$x \equiv \langle \cos(\psi_{\mathbf{R}'} - \psi_{\mathbf{R}}) \rangle = \left( \exp - \frac{K_B T}{2 \mathfrak{G}(0) y} \frac{\delta}{s^2 x} \right) \times \exp - \sum_k \frac{1 - \cos \mathbf{k} \cdot (\mathbf{R}' - \mathbf{R})}{2 N} \sqrt{\frac{a_k}{b_k}} \quad (43)$$

where

$$y = \frac{1}{s^2} \left\langle \left( s + \frac{1}{2} \right)^2 - \left( S_{\mathbf{R}}^z + \frac{1}{2} \right)^2 \right\rangle \quad (44)$$

and  $\delta$  is the fraction of the Brillouin zone which satisfies (42) ; at low temperature :

$$\delta = \left( \frac{a k_c}{\pi} \right)^D = \left( \frac{K_B T}{2 \pi_s \sqrt{AJ}} \right)^D. \quad (45)$$

This formula holds for the anisotropic ferromagnet ; for the antiferromagnet,  $A$  should be replaced by  $[A + \mathfrak{G}(0) - \mathfrak{J}(0)]$ .

It is easily checked from (23a), (23b), (40), (43) and (45) that the relative decrease of all quantities  $a_k$ ,  $b_k$ ,  $\langle S_{\mathbf{R}}^z \rangle^2$  and  $x$  is proportional to  $T^{D+1}$ , and the same property for  $\hbar \omega_k$  then follows from (23e). A rough order of magnitude for large  $s$  can be obtained if the second factor of (34) is neglected as well as the temperature variation of  $y$  :

$$x \simeq \exp - \frac{K_B T \delta}{2 s^2 \mathfrak{G}(0) x}. \quad (46)$$

This quantity defines the relative variation of  $a_k$  and therefore (if the variation of  $b_k$  is neglected) of the frequency. The curve (Fig. 1) exhibits a part in

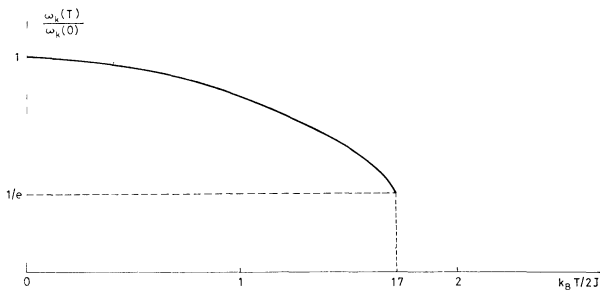


FIG. 1. — Temperature dependent frequency of magnons in  $\text{CsNiF}_3$  according to eq. (46).

$T^{D+1}$  followed (for  $s$  large enough) by a part linear in  $T$ , for  $K_B T > 2 s \sqrt{AJ}$ . There is a first-order transition at some temperature  $T_c$ , but this is clearly an artefact (as in ref. [21] and [22]) : the SCHA approximation is only correct at low temperature. In fact there is no transition for  $D = 1$ , and probably a second-order transition for  $D = 2$  [23].

According to (46), the transition temperature is defined by

$$K_B T \delta = 2 e s^2 \mathfrak{G}(0) \quad (47)$$

and at the transition :

$$x = 1/e. \quad (48)$$

**9. The linear chain at low temperature (fluctuations of long wavelength).** — In this chapter the time-dependent correlation is investigated and the quantum correction of first order in  $1/s$  to the classical result [24] is calculated. The temperature variation of the frequency is neglected (which is correct at low temperature and seems experimentally justified) and a long wavelength approximation is used, namely :

$$\begin{aligned} \mathfrak{G}(0) - \mathfrak{G}(k) &\approx |J| a^2 k^2 \\ A + \mathfrak{G}(0) - \mathfrak{J}(k) &\approx A \quad (\text{ferromagnets}) \\ A + \mathfrak{G}(0) - \mathfrak{J}(0) &\approx A + \mathfrak{G}(0) - \mathfrak{J}(0) \quad (\text{antiferromagnets}). \end{aligned} \quad (49)$$

Moreover the Bose factor in (27) will be approximated by a cut off  $k_c$  defined by (42) at low temperatures ; at large temperatures, where the solution of (42) would be greater than  $\pi/a$ ,  $k_c$  is defined by  $k_c = \pi/a$ , and the classical results of [24] are recovered. The approximate form of (27) is, using (16) :

$$\gamma(r, t) \simeq \exp - \frac{a}{2 \pi s^2} \int_0^{k_c} K_B T \frac{1 - \cos \omega_k t \cos kr}{|J| a^2 k^2} dk.$$

For large  $t$  ( $\omega_{k_c} t \gg 1$ ), or long distance ( $k_c r \gg 1$ ), this can be approximated as :

$$\begin{aligned} \gamma(r, t) &\simeq \exp - \frac{a}{2 \pi s^2} \frac{K_B T}{|J|} \times \\ &\times \left[ \int_0^\infty \frac{1 - \cos \omega_k t \cos kr}{a^2 k^2} dk - \int_{k_c}^\infty \frac{dk}{a^2 k^2} \right] \end{aligned}$$

or

$$\begin{aligned} \gamma(r, t) &\simeq \exp \left( \frac{K_B T}{2 \pi |J| s^2 a k_c} \right) \times \\ &\times \exp - \frac{a}{2 \pi s^2} \frac{K_B T}{|J|} \int_0^\infty \frac{1 - \cos \omega_k t \cos kr}{a^2 k^2} dk. \quad (50) \end{aligned}$$

The second factor is the classical expression [24] and the first factor is the quantum correction, a factor greater than 1 ; do not forget, however, that eq. (25) also contains a correction  $g_{\text{RR}}^0(t)$  independent of temperature, and which is smaller than 1. At low temperature where  $k_c$  is defined by (42) the quantum correction in (50) is also independent of  $T$ . The evaluation of the integral in (50) is straightforward [25] and the final result is :

$$\begin{aligned} \gamma(r, t) &= \exp \left( \frac{\hbar c}{2 \pi |J| s^2 a} \right) \times \\ &\times \exp - \frac{K_B T}{8 |J| s^2} \left( \left| \frac{r + ct}{a} \right| + \left| \frac{r - ct}{a} \right| \right) \quad (51) \end{aligned}$$

where the magnon velocity  $c$  is defined by :

$$\omega_k \simeq ck \quad (52)$$

$$\left. \begin{aligned} c &= \frac{2s}{\hbar} a \sqrt{AJ} && \text{(ferromagnets)} \\ c &= \frac{2s}{\hbar} a \sqrt{|J| [A + 2g(0)]} && \text{(antiferromagnets with nearest-neighbour interactions).} \end{aligned} \right\} (53)$$

*Neutron scattering.* — The neutron scattering cross section is essentially proportional to the Fourier transform  $\check{G}^\perp(k, \omega)$  of (25). The formulae given below hold for ferromagnets *in the classical limit*. The space transform after some elementary calculation is found to be :

$$\check{G}^\perp(k, t) = \frac{2e^{-|t|/\tau}}{a} \times \left\{ \frac{\kappa}{\kappa^2 + k^2} \cos \omega_k t + \left[ \frac{1}{k} - \frac{k}{\kappa^2 + k^2} \right] \sin \omega_k |t| \right\} \quad (54)$$

and the space-time Fourier transform is easily found to be :

$$a\check{G}^\perp(k, \omega) = 2f(k, \omega) + 2f(k, -\omega) \quad (55)$$

with

$$f(k, \omega) = \frac{\kappa}{\kappa^2 + k^2} \frac{1/\tau}{\frac{1}{\tau^2} + (\omega_k + \omega)^2} + \left( \frac{1}{k} - \frac{k}{\kappa^2 + k^2} \right) \frac{\omega_k + \omega}{\frac{1}{\tau^2} + (\omega_k + \omega)^2} \quad (56)$$

$$\kappa = \frac{K_B T}{4 J a s^2} \quad (57)$$

$$\frac{1}{\tau} = c\kappa. \quad (58)$$

All formulae hold for ferromagnets, but can be extended to antiferromagnets if  $k$  is replaced by  $\frac{\pi}{a} + k$ , with  $k$  small.

An alternative form of (56) is :

$$\check{G}^\perp(k, \omega) = \frac{1}{a} \frac{8c\kappa^2}{\kappa^2 + k^2} \times \frac{\omega_k^2 + \frac{1}{\tau^2}}{\left( \omega^2 + \omega_k^2 + \frac{1}{\tau^2} \right)^2 - 4\omega_k^2 \omega^2}. \quad (59)$$

For small wavelengths, formula (56) is most convenient ( $\kappa \ll k \ll 1/a$ ). The spectrum essentially consists of 2 Lorentzians represented by the first term of (56),

whereas the second term is a small correction except at high frequency where it truncates the Lorentzians.

For  $k < \kappa$ , expression (59) clearly shows that spin waves do *not* show up in the neutron spectrum (Fig. 2).

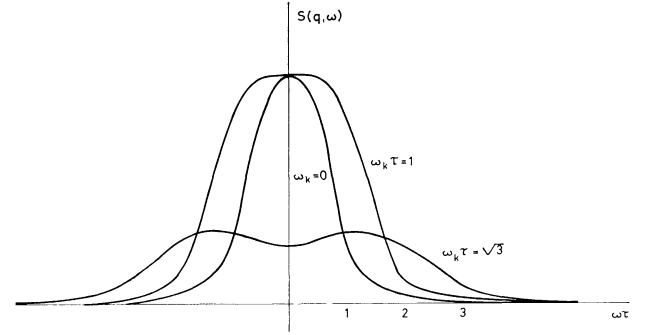


FIG. 2. — In-plane lineshapes in the classical linear chain. The ordinates are in different units for the 3 curves.

An alternative condition for the observability of spin waves is :

$$\lambda \lesssim 2\pi\xi \quad (60)$$

where  $\xi = 1/\kappa$  is the correlation length. Thus our formula (59) confirms the existence of a factor  $2\pi$  in the condition (60) as suspected by other authors [26]. This point is essentially confirmed by the quantum calculation in chapter 11 ; however, the concept of correlation length is not clear for finite  $s$  : one can imagine the correlation length as something smaller than  $1/\kappa$  (defined by eq. (57)) at small temperature because of  $g_{RR}^0$  (see §§ 6 and 7), but larger than  $1/\kappa$  at higher temperature because of the first factor in eq. (51).

Let us compare our result (59) with analytic formulae obtained by other authors [13], [14]. The derivation is of course quite different and involves a new method of calculation of the second moment (a method which is not applicable to the isotropic case). Now our formula has only 4 poles and satisfies fewer moment relations than in [13] and [14]. The 4th and higher moments are infinite because of the replacement of the upper bound  $k_c$  by  $+\infty$  in an integral at the beginning of the calculation. This approximation seems correct for *small* frequencies, and thus, if one believes eq. (27), the effect of the finite 4th moment would be to cut large frequencies off, rather than to add intensity at zero frequency, as seems to be the case in the continued fraction method used in [13] and [14].

**10. The linear chain at low temperature : short wavelength case.** — 10.1 STATIC CORRELATION. — In what follows, it will be shown that the static correlation function is *gaussian* at short distances, instead of exponential, as in the classical case, represented by the second factor of eq. (51). For  $t = 0$  and

$$|\mathbf{R}' - \mathbf{R}| = na$$

small with respect to  $k_c^{-1} = 2\beta$  as  $(AJ)^{1/2}$  (this formula holds for ferromagnets), eq. (27) reads :

$$\gamma_{RR}(0) \equiv \gamma_n(0) = \exp - \frac{1}{s} \frac{a}{2\pi} \int_{-\infty}^{\infty} dk \frac{\frac{1}{2} k^2 a^2 n^2}{e^{2\beta s |k| a \sqrt{AJ}} - 1} \frac{\sqrt{AJ}}{|ka|}.$$

This expression can be calculated by means of the formula :

$$\int_0^{\infty} \frac{x dx}{e^x - 1} = \int_0^1 \frac{\text{Log } y}{1 - y} dy = \frac{\pi^2}{6}$$

yielding :

$$\gamma_n(0) = e^{-(\kappa' na)^2} \quad (61)$$

with :

$$\kappa' a = \kappa a \sqrt{\frac{\pi s}{3}} \left(\frac{J}{A}\right)^{1/4}. \quad (62)$$

The extension to antiferromagnets involves the replacement of  $A$  by  $\mathcal{G}(0) - \mathcal{J}(0) + A$ , thus obtaining smaller values of  $\kappa'$ .

The static correlation function, resulting from (51) and (61), is plotted in figure 3 for  $\kappa' = \kappa$  (which is

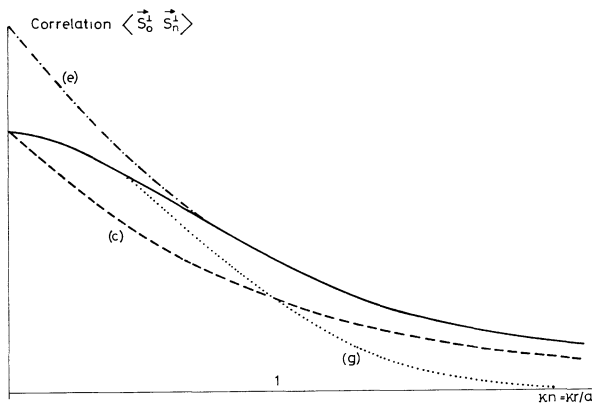


FIG. 3. — Static correlation function in CsNiF<sub>3</sub> (full curve), as obtained by interpolation between the exponential form (e) (eq. (51)) and the gaussian form (g) (eq. (61)). The result is higher than the classical curve (59) and this effect would be increased in an antiferromagnet. The factor (26) has been neglected. The ordinate is in « arbitrary units »: clearly  $\langle |S_0|^2 \rangle$  is of order  $s^2$ , but was not accurately calculated.

approximately the case for CsNiF<sub>3</sub>). The Fourier transform is given by figure 4. The classical, Ornstein-Zernike law is not modified for small  $k$ , but the wings are gaussian-like.

Expressions (61) and (62) are correct whenever  $k_c$  lies in the linear part of the spectrum, even if  $\pi/na$  does not.

10.2 INELASTIC NEUTRON SCATTERING IN-PLANE AND OUT-OF-PLANE CORRELATIONS. — Although magnon peaks are already present in the classical formula (59), this formula is quite unable to describe the experiments

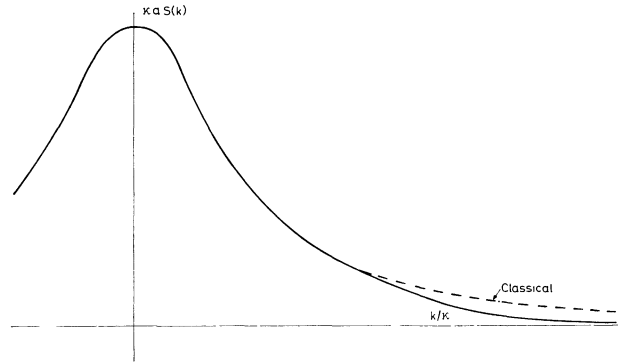


FIG. 4. — Fourier transform of figure 3. The dashed curve is classical.

on CsNiF<sub>3</sub>, where quantum effects, and especially dissymmetry between energy gain and energy loss are present. Moreover, beside the in-plane correlation calculated in paragraph 6, the out-of-plane correlation given by eq. (15) can also be observed for large momentum transfer, which gives rise to a very narrow peak whereas the in-plane contribution is broad except at very low temperature. The presence of a narrow peak at fairly high temperature is characteristic of an anisotropic system: it was observed in CsNiF<sub>3</sub> [29] but not in TMMC [26], [12].

A hand-waving description of the situation is given below: locally, the system looks like a helimagnet, with a pitch described by a vector  $\mathbf{Q}$ , the direction of which is very close to  $Oz$ , whereas its magnitude varies between  $-\kappa$  and  $\kappa$  (for a ferromagnet). The magnon frequency is [27], [28]

$$\begin{aligned} \hbar\omega_k = \\ = 2s \sqrt{[\mathcal{J}(\mathbf{Q}) - \mathcal{J}(\mathbf{k}) + A] \left[ \mathcal{J}(\mathbf{Q}) - \frac{\mathcal{J}(\mathbf{Q} + \mathbf{k}) + \mathcal{J}(\mathbf{Q} - \mathbf{k})}{2} \right]} \end{aligned} \quad (63)$$

(note that this formula reduces to eq. (16) for  $\mathbf{Q} = 0$  or half a reciprocal lattice vector). A neutron scattering experiment with a momentum transfer  $\mathbf{q}$  « sees » magnons of wave vector  $\mathbf{q}$  through the out-of-plane correlation  $\langle S^z S^z \rangle$ , and magnons of wave vectors  $\mathbf{q} \pm \mathbf{Q}$  through the in-plane correlation. It is seen from (63) that the correction to  $\omega_{\mathbf{q} \pm \mathbf{Q}}$  is proportional to  $Q$ , and the correction to  $\omega_{\mathbf{q}}$  is only proportional to  $Q^2$ . Therefore the spread in  $\omega_{\mathbf{q} \pm \mathbf{Q}}$  is proportional to  $\kappa$  whereas the spread in  $\omega_{\mathbf{q}}$  is proportional to  $\kappa^2$ , as see below, paragraph 10.5.

Of course, correlation functions of the type  $\langle S_R^x S_R^z(t) \rangle$  vanish because of the invariance under the transformation

$$S_R^x \rightarrow -S_R^x, \quad S_R^y \rightarrow -S_R^y.$$

Clearly the existence of a narrow peak due to out-of-plane correlations is *typical of systems with an easy magnetization plane*, whereas the in-plane component

is expected to be similar to the spectrum of isotropic systems.

10.3 IN-PLANE CORRELATION. — The problem is to evaluate the 2 terms at the right hand side of (31). The first term is readily obtained from (29)

$$\begin{aligned} \sum_{R'} J_{RR'} \varepsilon_R \varepsilon_{R'} (f_{OR'} - f_{OR}) &= \\ &= \frac{1}{sN} \sum_k e^{-ikR} \frac{\hbar\omega_k}{2s} \left( \frac{e^{i\omega_k t}}{2} + \frac{\cos \omega_k t}{e^{\beta\hbar\omega_k} - 1} \right) \end{aligned} \quad (64)$$

and after summation over R appears  $\tilde{G}^\perp(q+k, t)$ .

$$\tilde{G}^\perp(q, t) = \frac{1}{2s^2 N} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(q) + |J| \kappa^2 a^2} \sum_k \hbar\omega_k \left( \frac{e^{i\omega_k t}}{2} + \frac{\cos \omega_k t}{e^{\beta\hbar\omega_k} - 1} \right) \tilde{G}^\perp(q+k, t). \quad (66)$$

The magnon peak clearly comes from those values of  $k$  which are close to  $-q$ , because  $\tilde{G}^\perp(q+k, t)$  is then large. Then the variation with time of the bracket is much faster than the variation of  $\tilde{G}^\perp(q+k, t)$ ; therefore a satisfactory description of the magnon peak is obtained by replacing  $\tilde{G}^\perp(q+k, t)$  by  $\tilde{G}^\perp(q+k, 0)$

$$\tilde{G}^\perp(q, t) = \frac{1}{2s^2 N} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(q) + |J| \kappa^2 a^2} \sum_k \hbar\omega_k \left( \frac{e^{i\omega_k t}}{2} + \frac{\cos \omega_k t}{e^{\beta\hbar\omega_k} - 1} \right) \tilde{G}^\perp(q+k, 0). \quad (67)$$

The Fourier transform of this function is :

$$\check{G}^\perp(q, \omega) = \frac{1}{4s^2} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(q) + |J| \kappa^2 a^2} \int \frac{a dk}{2\pi} \hbar\omega_k \left[ 2\pi\delta(\omega - \omega_k) + 2\pi \frac{\delta(\omega - \omega_k) + \delta(\omega + \omega_k)}{e^{\beta\hbar\omega_k} - 1} \right] \tilde{G}^\perp(q+k, 0)$$

or :

$$\check{G}^\perp(q, \omega) = \frac{a}{4s^2} \frac{\hbar\omega}{\mathfrak{G}(0) - \mathfrak{J}(q) + |J| \kappa^2 a^2} \frac{\tilde{G}^\perp(q+k(|\omega|), 0) + \tilde{G}^\perp(q-k(|\omega|), 0)}{1 - e^{-\beta\hbar\omega}} \frac{d|\omega|}{d|\omega|}. \quad (68)$$

Here  $k(\omega)$  is the inverse function of  $\omega(k)$ , chosen to be positive in order to have a single-valued function. The result is shown in figure 5 for CsNiF<sub>3</sub> at 10 K

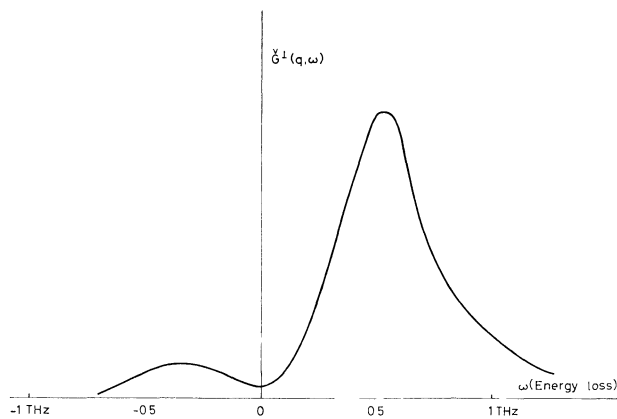


FIG. 5. — In-plane scattering function in CsNiF<sub>3</sub> at 10 K for a momentum transfer  $0.3 \pi\hbar/a$ .

with  $q = 0.3 \pi/a$ . Numerically, our curve seems to agree with what might be obtained by the McLean-Blume method [12] for an isotropic system; however the outer edge of the line is not as sharp. Our curve

The second term at the right hand side of (31) is small at low temperature except for small values of  $q$ ; indeed, for  $q = 0$ , the second term is a sum of positive terms, which is not the case for the first one. An approximation is obtained as follows from (51) and (28), neglecting  $g^0$  in (25) :

$$|f_{OR}(t) - f_{OR}(0)| \simeq |f_{OR}(0) - f_{OR}(0)| \approx \kappa a. \quad (65)$$

This is clearly an overestimation because the left hand side decreases with  $t$ . The final result obtained by insertion of (64) and (65) into (31) is for first neighbour interactions :

was also compared with a calculation using the method of reference [13], which was kindly communicated to us by S. W. Lovesey : it turns out that : i) Lovesey's spectrum is somewhat narrower, which can be due to underestimation of the quantum correction in (50) for  $s = 1$ ; ii) Lovesey finds a larger intensity at small frequency. As stated before (§ 9), we do not see any reason for a strong scattering at  $\omega \simeq 0$  and, although there is some experimental evidence for such an effect [26], it is not excluded that it is an artefact of the continued fraction method.

In CsNiF<sub>3</sub>, no broad spectrum similar to figure 5 has been observed, which is not surprising, due to statistical errors. Instead, a narrow peak has been observed [11], [29], which can be attributed to the out-of-plane correlation (see 10.1 and 10.2). Further experiments are necessary to check : i) that a broader peak appears at lower temperatures; ii) that the narrow peak vanishes when  $q$  becomes close to the  $z$ -axis, in agreement with general neutron scattering formulae.

There is a much simpler way to obtain eq. (67) for  $q \gg \kappa$  : the equation

$$\frac{d^2}{dt^2} \langle e^{i(\psi_R - \psi_{R'}(t))} \rangle = - \langle \dot{\psi}_R \dot{\psi}_{R'}(t) e^{i(\psi_R - \psi_{R'}(t))} \rangle$$

(which is a classical approximation, but a quantum version can easily be given) is factorized into :

$$\frac{d^2}{dt^2} \langle e^{i(\psi_R - \psi_{R'}(t))} \rangle = - \langle \dot{\psi}_R \dot{\psi}_{R'}(t) \rangle \langle e^{i(\psi_R - \psi_{R'}(t))} \rangle$$

Then  $\psi_{R'}(t)$  is replaced by  $\psi_{R'}(0)$  in the exponent of the right hand side, the equation is integrated and Fourier transformed in space and one recovers (67); the sophisticated proof has the advantage of giving the range of validity of (67).

It is possible to check the approximation (67) for  $t = 0$  for very large frequencies where the Boltzmann factor  $e^{\beta \hbar \omega_k}$  is large; one has :

$$\begin{aligned} \tilde{G}^\perp(q, 0) &\simeq \frac{1}{2sN} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(q)} \frac{\hbar \omega_{q+Q}}{2s} \sum_k \tilde{G}^\perp(k, 0) = \\ &= \frac{1}{2s} \sqrt{\frac{A + \mathfrak{G}(0) - \mathfrak{G}(q)}{\mathfrak{G}(0) - \mathfrak{J}(q)}}} \end{aligned}$$

in agreement with (20) and the Taylor expansion of (26).  $Q$  is a superlattice point ( $Q = 0$  in a ferromagnet). Use has been made of  $G(Q + k) = J(k)$ .

In the classical limit, eq. (67) reads :

$$\tilde{G}^\perp(q, 0) = \frac{1}{2s^2} \frac{K_B T}{\mathfrak{G}(0) - \mathfrak{J}(q) + |J| \kappa^2 a^2}.$$

It is of interest to note that in the ordered case the function  $\tilde{G}(k + q, 0)$  contains a  $\delta(k + q - Q)$ , so that in this case eq. (67) describes undamped magnons.

**10.4 ZONE BOUNDARY NARROWING.** — The « magnon damping » calculated in Appendix A is not included in this expression. Neglecting this damping, the linewidth of the neutron line is roughly given by :

$$\delta \omega_k = \frac{\partial \omega_k}{\partial k} \delta k \quad (69)$$

where  $\delta k$  is the width of the static correlation function  $\tilde{G}^\perp(k, 0)$ . For classical spins,  $k = \kappa$ , given by (57). According to (69), the linewidth vanishes at the zone boundary; this is actually not the case because of higher order terms neglected in (69), and because of the « magnon damping » of Appendix A. However, the magnon damping is expected to be *minimal at the zone boundary* in a linear chain (or at the superzone boundary in an antiferromagnet). This *zone boundary narrowing* can also be obtained (from a decoupling approximation) in one-dimensional Heisenberg (isotropic) systems. Its existence is clear for systems with an easy magnetization plane, at least in the classical case where (69) is proportional to  $T$ , whereas the magnon damping is proportional to  $T^2$ . In quantum systems, however, the effect might be killed by the magnon damping.

**10.5 OUT-OF-PLANE CORRELATIONS.** — It has been seen above (§ 10.1) that at low temperature the out-of-plane correlations give rise to a very narrow peak.

The basic questions are : i) what is the intensity of this peak compared with the broad, « in-plane » peak ? ii) what is the damping ? iii) up to which temperature will the narrow peak persist ?

The intensities follow at once from (15) and (68). The last two questions will be answered by means of (63), although more sophisticated calculations of the damping can be made (see Appendix).

To simplify the formulae, a long wavelength approximation :

$$\mathfrak{J}(Q + k) \equiv \mathfrak{G}(q + k) \simeq \mathfrak{G}(0) - |J| a^2 (q + k)^2$$

will be used inside (63), yielding :

$$\begin{aligned} \hbar \omega_k &\simeq 2s \sqrt{[\mathfrak{G}(0) - \mathfrak{J}(k) + A - |J| a^2 q^2] [\mathfrak{G}(0) - \mathfrak{G}(k)]} \\ &\simeq \hbar \omega_k(T=0) \left[ 1 - \frac{1}{2} \frac{|J| a^2 q^2}{\mathfrak{G}(0) - \mathfrak{J}(k) + A} \right]. \end{aligned}$$

Here  $q$  is a small vector which describes the local pitch of the local helix; in the classical approximation :

$$-\kappa \lesssim q \lesssim \kappa.$$

This gives both the shift  $\delta \omega_k$  and the relaxation time  $\tau_k$  :

$$\frac{\delta \omega_k}{\omega_k} \simeq \frac{1}{\tau_k \omega_k} \simeq \frac{1}{2} \frac{|J| a^2 \kappa^2}{\mathfrak{G}(0) - \mathfrak{J}(k) + A}. \quad (70)$$

To find the domain of validity of the theory, one can remark that the spins are aligned inside a region containing about  $(\kappa a)^{-1}$  spins, which therefore has a total spin of about  $(s/\kappa a)$ , and an anisotropy energy of about  $A(s/\kappa a)^2$ , which has to be much larger than  $K_B T$  for the theory to be valid :

$$A \left( \frac{s}{\kappa a} \right)^2 \gg K_B T \quad \text{or} \quad \left( \frac{K_B T}{2|J|s^2} \right)^3 \ll \frac{2A}{J}. \quad (71)$$

Note that the  $k$ -dependent shift that appears in (70) is different from the one that results from the SCHA approximation. The relaxation time, however, is qualitatively consistent with the calculation in Appendix A, at least in the linear region of the spectrum.

**11. Two-dimensional magnets at low temperature in the long distance or long time approximation ( $k_c r \gg 1$  or  $ck_c t \gg 1$ ).** —

**11.1 GENERAL FORMULAE.** — Only the in-plane correlation will be considered here. As in the linear case, the out-of-plane correlation is observable at large momentum transfer by neutron scattering, and gives rise to a very narrow magnon peak even at reasonably high temperature.

Using the same approximations as in paragraph 9, it is shown in Appendix B that the in-plane correlation function is defined by :

$$\left\{ \begin{aligned} \gamma(r, t) &= \alpha^\tau (k_c r)^{-\tau} & (ct \leq r) \\ \gamma(r, t) &= \alpha^\tau (t \omega_T + \sqrt{t^2 \omega_T^2 - k_c^2 r^2})^{-\tau} & (ct \geq r) \end{aligned} \right. \quad (72)$$

where  $\alpha$  is a constant and :

$$k_c \approx \inf\left(\frac{K_B T}{\hbar c}, \frac{\pi}{a}\right) \quad (73)$$

$$\omega_T = ck_c, \quad \tau = \frac{K_B T}{4\pi |J| s^2}; \quad (74)$$

$c$  is defined by (53).

11.2 EQUAL TIME CORRELATION. — The space Fourier transform at  $t = 0$  is obtained from (72) :

$$\begin{aligned} \tilde{\gamma}(q, 0) &\equiv \frac{1}{a^2} \int \gamma(\mathbf{r}, 0) e^{i\mathbf{q}\cdot\mathbf{r}} d^2r \\ &= \frac{\alpha^\tau}{a^2} \text{Lim}_{\varepsilon \rightarrow 0} \int \frac{e^{i\mathbf{q}\cdot\mathbf{r}}}{(k_c r)^\tau} e^{-\varepsilon r} d^2r \\ &= \frac{\alpha^\tau}{k_c^\tau a^2} \text{Lim}_{\varepsilon \rightarrow 0} 2\pi \int_0^\infty J_0(qr) r^{1-\tau} e^{-\varepsilon r} dr \\ &= A \frac{\alpha^\tau}{(k_c a)^\tau} (qa)^{-2+\tau} \end{aligned} \quad (75)$$

with

$$A = 2\pi \text{Lim}_{\varepsilon \rightarrow 0} \int_0^\infty x^{1-\tau} J_0(x) e^{-\varepsilon x} dx. \quad (76)$$

Without  $\varepsilon$  the integral would be meaningless at low temperature. Nevertheless, the Fourier transform of  $\gamma(r, 0)$  is quite well defined as a distribution [30].

11.3 NEUTRON INELASTIC SCATTERING. — Function (72) has the very remarkable feature that, for finite  $t$ , it increases with  $r$  for  $r < ct$  (Fig. 6). It has

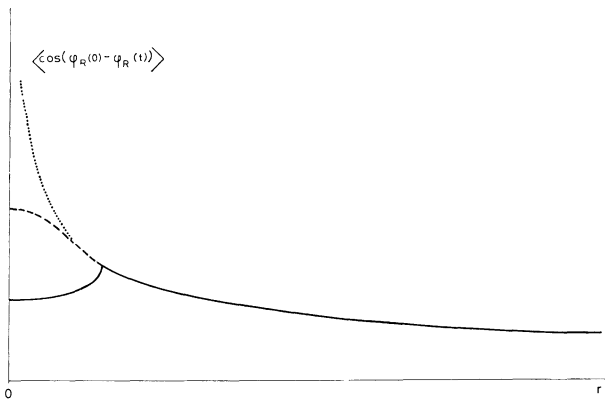


FIG. 6. — In-plane correlation function in a 2-D system as a function of  $r$  for given  $t$  (for  $\tau = 1/2$ ).

a square root singularity at  $r = ct$ , and this results, as shown in Appendix C, in a singularity of the scattering function  $\tilde{\gamma}(q, \omega)$  at  $\omega = \pm \omega_q$  (Fig. 7). Namely :

If  $\tau < 1$ ,  $\tilde{\gamma}(q, \omega)$  diverges as  $|\omega \pm \omega_q|^{\tau-1}$ .

If  $\tau = 1$ ,  $\tilde{\gamma}(q, \omega)$  diverges logarithmically at  $\pm \omega_q$ .

If  $1 < \tau < 2$ ,  $\tilde{\gamma}(q, \omega)$  is peaked at  $\pm \omega_q$  and its derivative diverges as  $|\omega - \omega_q|^{\tau-2}$ .

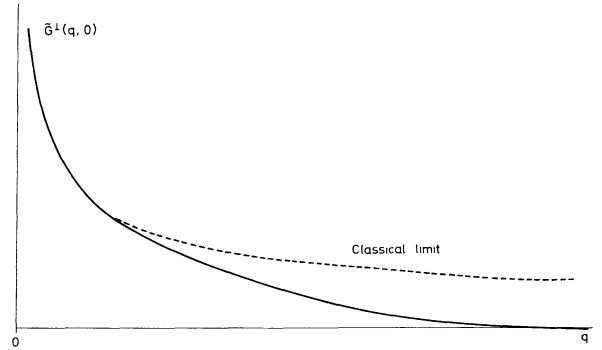


FIG. 7. — Fourier transform of figure 6 at  $t = 0$ .

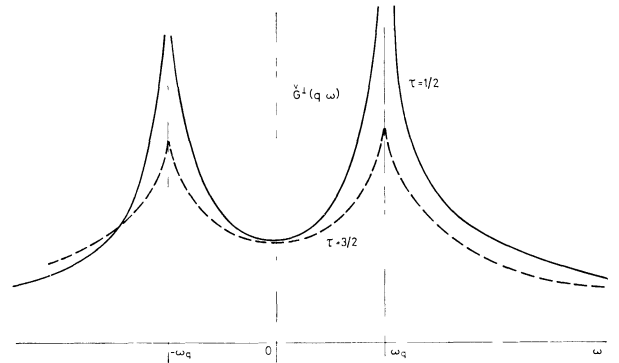


FIG. 8. — Typical curves for the in-plane scattering function in a 2-D system.

The case  $\tau > 2$  is beyond the range of validity of the harmonic approximation.

The above results are stated for ferromagnets; for antiferromagnets,  $\omega_q$  must be replaced by  $\omega_{q+\mathbf{Q}}$ , with  $\varepsilon_{\mathbf{R}} = e^{i\mathbf{Q}\cdot\mathbf{R}}$  (see § 3).

11.4 CONCLUSION. — In agreement with other authors [2], the equal time correlation function is found to diverge at  $q = 0$  for  $\tau < 2$ ; the quantum correction to the classical formula [2], [7], [9] is contained in  $k_c$  in eq. (72).

It is found that spin waves are visible by neutron scattering (neglecting instrumental resolution) even at very small  $q$ ; in reference [24] we were unable to prove this, and a sufficient (but not necessary) condition was given, which is therefore meaningless. Of course, the peak is rounded by the damping (see Appendix A) but should be observable at least at small  $q$ , where the damping is small; a remarkable conclusion of the theory is that, at moderate temperature, spin waves might be observable in the in-plane correlation at low momentum transfer, but not for larger  $q$ ! This is based, of course, on a perturbation expansion whose convergence is questionable (see Appendix A); it surely diverges above some temperature, perhaps the Stanley Kaplan transition temperature [23].

In the next paragraph, the time dependent correlation is calculated in the short wavelength limit;

the results are identical to those demonstrated in the present section, and this is not surprising because the long wavelength region and the short wavelength region turn out to overlap.

Mikeska has obtained results quite similar to ours in a phonon problem which is mathematically equivalent to our problem [31].

**12. Two dimensional magnets in the short distance, short time approximation.** — 12.1 THE STATIC CORRELATION ( $t = 0$ ) at short distance is obtained from (27) by a calculation quite similar to that of paragraph 10, and is again described by a *gaussian* law :

$$\gamma(r, 0) = e^{-(\kappa' r)^2} \quad (77)$$

with

$$\kappa' = \frac{(K_B T)^{3/2}}{4 \pi \hbar c s \sqrt{J}} \sqrt{\int_0^\infty \frac{x^2 dx}{e^x - 1}}. \quad (78)$$

Formula (77) holds for  $k_c r < 1$ , or  $r < \hbar c / K_B T$ , in agreement with (73).

12.2 The time dependent correlation function is still given by (31). Neglecting the second term on the right hand side, one obtains an equation similar to (67) :

$$\begin{aligned} \tilde{G}^\perp(q, t) &= \frac{1}{2 s^2 N} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(q)} \times \\ &\times \sum_k \hbar \omega_k \left( \frac{e^{i\omega_k t}}{2} + \frac{\cos \omega_k t}{e^{\beta \hbar \omega_k} - 1} \right) \tilde{G}^\perp(q + k, 0). \quad (79) \end{aligned}$$

Here the  $a^2 \kappa^2$  term of (67) is lacking in the denominator. Now the second term of the right hand side of (31) will be evaluated at  $t = 0$ ; using (72), it is found that if R and R' are neighbours :

$$|f_{OR} - f_{OR'}| \lesssim \frac{\tau a}{r}.$$

Hence the order of magnitude of the second term on the right hand side of (31) is :

$$\begin{aligned} \frac{\mathfrak{G}(0)}{2[\mathfrak{G}(0) - \mathfrak{J}(\mathbf{q})]} \int_0^\infty \frac{d^2 r}{a^2} e^{i\mathbf{q}r} \frac{a^2 \tau^2}{r^2} \left[ \frac{r}{a} \inf \left( 1, \frac{a K_B T}{\hbar c} \right) \right]^{-\tau} < \\ < \frac{\mathfrak{G}(0) \tau}{\mathfrak{G}(0) - \mathfrak{J}(\mathbf{q})} \left[ \sup \left( a, \frac{\hbar c}{K_B T} \right) \right]^\tau. \end{aligned}$$

$$\tilde{G}^\perp(\mathbf{q}, t) = \frac{1}{2 s^2} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(\mathbf{q})} \frac{a^2}{4 \pi^2} \int_0^\infty \frac{d\omega}{|\nabla_q \omega_q|} \hbar \omega \left( \frac{e^{i\omega t}}{2} + \frac{\cos \omega t}{e^{\beta \hbar \omega} - 1} \right) \int_{-\infty}^\infty dk_\perp \tilde{G}^\perp(\mathbf{q} + \mathbf{k}_\omega + \mathbf{k}^\perp, 0)$$

where, according to (75) :

$$\begin{aligned} \int_{-\infty}^\infty dk_\perp \tilde{G}^\perp(\mathbf{q} + \mathbf{k}_\omega + \mathbf{k}^\perp, 0) &\approx \left( \frac{\alpha}{k_c} \right)^\tau \int_{-\infty}^\infty \frac{dk^\perp}{\{a^2 [(\mathbf{q} + \mathbf{k}_\omega)^2 + k_1^2]\}^{2-\tau}} = \frac{(\alpha/k_c)^\tau}{a^{2-\tau} |\mathbf{q} + \mathbf{k}_\omega|^{1-\tau}} = \\ &= \left( \frac{\alpha}{k_c} \right)^\tau a^{\tau-2} \frac{|\nabla_q \omega_q|^{1-\tau}}{|\omega - \omega_q|^{1-\tau}}. \end{aligned}$$

Therefore :

$$\tilde{G}^\perp(\mathbf{q}, t) = \frac{1}{8 \pi^2 s^2} \frac{1}{\mathfrak{G}(0) - \mathfrak{J}(\mathbf{q})} \left( \frac{\alpha a}{k_c |\nabla_q \omega_q|} \right)^\tau \int_0^\infty \frac{d\omega \hbar \omega}{|\omega - \omega_q|^{1-\tau}} \left( \frac{e^{i\omega t}}{2} + \frac{\cos \omega t}{e^{\beta \hbar \omega} - 1} \right). \quad (81)$$

The upper bound has been obtained by replacing  $e^{i\mathbf{q} \cdot \mathbf{r}}$  by 1.

A sufficient (but not necessary) condition for (79) to be correct is that this upper bound be much smaller than the left hand side of (31) (for  $t = 0$ ), which is given by (75), namely :

$$\frac{\mathfrak{G}(0) \tau}{2 |J| a^2 q^2} \ll (qa)^{\tau-2} \quad \text{or} \quad aq \gg \tau^{1/\tau}. \quad (80)$$

12.3 THE CASE  $\tau < 1$ . — It will now be shown that eq. (79) defines 2 peaks with divergences at  $\omega = \pm \omega_q$ .

The vectors  $k$  such that  $\omega_k$  lies between  $\omega$  and  $\omega + d\omega$  are defined, in the vicinity of  $k = -q$ , by :

$$\omega < \omega_q + (\mathbf{k} + \mathbf{q}) \cdot \nabla_q \omega_q < \omega + d\omega.$$

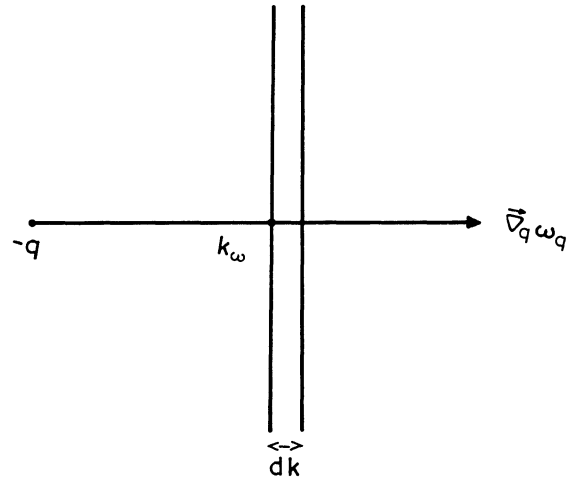


FIG. 9. — Equal frequency surfaces in the reciprocal space.

This is the equation of a strip of width :

$$dk = \frac{d\omega}{|\nabla_q \omega_q|}$$

centered at a point  $\mathbf{k}_\omega$  lying on the parallel to  $\nabla_q \omega_q$  through point  $-\mathbf{q}$  (Fig. 9). Defining  $\mathbf{k} = \mathbf{k} - \mathbf{k}_\omega$ , eq. (79) can be written as :



The scattering function  $\check{G}^\perp$  follows at once ; 2 corrections are to be made :

i) The wings should be cut off at  $|\omega - \omega_q| = \Delta\omega$  with :

$$\Delta\omega = |\nabla_q \omega_q| \Delta k_\omega \approx |\nabla_q \omega_q| \frac{k_c}{\alpha}.$$

Indeed the integration must be carried out on  $-k_c < k < k_c$ , where  $k_c$  is given by (73), and not from  $-\infty$  to  $\infty$ , as done above.

ii) The divergence is smeared out by the damping, which in the appendix is found to be proportional to  $T^5 q^2$ . As this is small at low temperature, it is seen that the magnon peaks can be observed if condition (80) holds.

At low temperature, condition (80) is extremely weak and its domain of validity overlaps with the domain of validity of chapter 11. Thus magnons show up in the neutron spectrum for *all* momentum transfer. Note that the results obtained in paragraphs 11.3 and 12.3 are the same.

12.4 THE CASE  $1 < \tau < 2$ . — A similar calculation shows that  $\check{G}^\perp(q, \omega)$  has an infinite derivative at  $\pm \omega_q$ , in agreement with the result of the previous section. It is believed, however, that the inelastic scattering neutron spectrum will be essentially structureless, due to the damping as well as the instrumental resolution.

12.5 Below are summarized the results obtained for the space Fourier transform  $\check{\gamma}(k, 0)$  of the function (27) at  $t = 0$  :

12.5.1 *Quantum case* ( $K_B T < \hbar c/a$ ) :

$$\check{\gamma}(k, 0) \approx \frac{1}{2 s^2 [\mathfrak{G}(0) - \mathfrak{G}(\mathbf{k})]} \frac{\hbar \omega_k}{2 \operatorname{th} \frac{1}{2} \beta \hbar \omega_k} \times \times \operatorname{inf} \left[ 1, \left( \frac{\mathfrak{G}(0) - \mathfrak{G}(k)}{\hbar \omega_{\max}} \right)^{\tau/2} \right].$$

12.5.2 *Classical case* :

$$\check{\gamma}(k, 0) \approx \frac{K_B T}{2 s^2 |J|^{\tau/2} [\mathfrak{G}(0) - \mathfrak{G}(k)]}.$$

Comparison of our results with experiment is difficult : in  $K_2\text{CuF}_4$  [32], [33] the planar anisotropy is too weak (1 %) to use the present theory ; moreover it is likely that even a very weak uniaxial anisotropy or 3-D interaction is sufficient to spoil the 2-D character [31], [34], [35].

**Acknowledgments.** — I am very grateful to B. Dorner and M. Steiner for communication of their experimental results and fruitful discussions, to G. Sarma and H. J. Mikeska for information about their theoretical work, to W. B. Yelon and G. Parisot for helpful discussions. The author is happy to thank S. W. Lovesey, who pointed out the analogy and the differences of the present method with the continued fraction method.

## APPENDIX A

1. **Damping of magnons (ferromagnets).** — In this Appendix will be calculated the damping of « magnons » (i. e. the quasi-particles which appear in the harmonic approximation) ; The Hartree renormalization will not be considered, for the sake of simplicity : one has to assume that it is already performed. Only the fourth-order interaction between magnons will be considered, and a formula due to Mori and Kubo will be used, together with a decoupling approximation, to obtain a self-consistent damping. Because of the decoupling, this formula is equivalent to a perturbation theory without vertex renormalization. It is of interest to ask the dangerous question : « what would happen in a perfectly correct theory ? » Our answer is :

- 1) « The perturbation series would diverge ;
- 2) However, the foregoing treatment is expected to be correct for a sufficiently large  $k$ . »

The complete perturbation series is expected to diverge because the interaction  $-\cos(\psi_i - \psi_j)$  has a negative fourth order term ; to understand the meaning of this fact, consider a solid with a first neighbour interaction consisting of a harmonic

attraction plus a fourth order repulsion : clearly, it will explode after some time (which can be long at low temperature, if the solid is finite), and thus the perturbation expansion has to diverge. Of course, the explosion is prevented by higher order attractive terms in the hamiltonian, but this is probably not sufficient to prevent the divergence of the perturbation expansion : the physical meaning of this divergence is that at any finite temperature an atom has some finite probability (even though perhaps very small) of leaving its site and diffusing across the crystal ; one can argue that this divergence has no physical meaning because particles are indistinguishable and sooner or later a new particle will come and occupy the vacancy, whereas the old particle will fall into another vacancy, etc... ; but in conventional phonon theory a divergence must appear in the calculation of the phonon propagator. It is the same in our case, but we do not know the effect of this divergence. At least in the 1-D case, a dramatic increase of the damping at very low wave vector is expected ; indeed, the result derived in this Appendix is that magnons are well defined for small  $k$ . On the other hand we do not expect this at high temperature and there must

be a transition temperature, but this is not very likely in one dimension, so the results derived below are expected to be wrong.

Nevertheless, they are probably correct for large wave vectors  $k$ . Indeed if we assume that the breakdown of the perturbation theory is due to the possibility of thermally excited jumps over a barrier of height  $4Js^2$ , the number of atoms within a period being about  $1/ka$  and the number of attempts made by each atom to jump over the barrier during a period of time being about  $1/ka$ , we see that the probability of jumping is negligible if

$$ka \gg \exp - \frac{4\beta Js^2}{D+1}.$$

This result suggests that at temperatures well below

the mean field Curie point, the foregoing calculation is good except in a very small region around  $k = 0$ . It is also of interest to note that this calculation is in qualitative agreement with the hand-waving calculation of paragraph 10.4.

We emphasize that the divergence of  $\langle \varphi_{\mathbf{R}}^2 \rangle$ , inherent to the 1- $D$  or 2- $D$  character of the system, has no influence on the perturbation theory at least at lower orders, as only the (finite) fluctuations of  $(\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})$  for first neighbours are relevant.

The quantitative calculation of damping is extremely tedious; our purpose in this appendix is only to show that damping is small at low temperature, especially at large wavelength, and to describe qualitatively how the damping varies with  $T$  and  $k$  in a few limiting cases.

The fourth-order terms of (8) are in the classical limit ( $s \rightarrow \infty$ ):

$$\begin{aligned} \mathcal{H}^{(4)} &= - \sum_{\mathbf{R}\mathbf{R}'} \frac{J_{\mathbf{R}\mathbf{R}'}}{4s^2} S_{\mathbf{R}}^z S_{\mathbf{R}'}^z + \frac{1}{4s^2} \mathfrak{J}(0) \sum_{\mathbf{R}} S_{\mathbf{R}}^z - \frac{1}{4} \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} (S_{\mathbf{R}}^z + S_{\mathbf{R}'}^z) (\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})^2 - \frac{s^2}{24} \sum_{\mathbf{R}\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'} (\varphi_{\mathbf{R}} - \varphi_{\mathbf{R}'})^4 \\ &= \frac{1}{4s^2 N} \sum_{kq q'} [\mathfrak{J}(0) - \mathfrak{J}(\mathbf{k})] S_q^z S_{k-q}^z S_{-q'}^z S_{q'-k}^z - \frac{J}{2N} \sum_{akq q'} \gamma_q^a \gamma_{k-q}^a \varphi_q \varphi_{k-q} S_{-q'}^z S_{q'-k}^z - \\ &\quad - \frac{Js^2}{24N} \sum_{akq q'} \gamma_q^a \gamma_{-q'}^a \gamma_{k-q}^a \gamma_{q'-k}^a \varphi_q \varphi_{-q'} \varphi_{k-q} \varphi_{q'-k} \end{aligned} \quad (\text{A.1})$$

where  $a$  is the vector distance between 2 nearest neighbours and

$$\gamma_k^a = 1 - e^{i\mathbf{k} \cdot \mathbf{a}}.$$

Henceforth a long wavelength approximation will be made:

$$\mathfrak{J}(0) - \mathfrak{J}(\mathbf{k}) \simeq Ja^2 k^2, \quad \gamma_k^a \simeq i\mathbf{k} \cdot \mathbf{a}.$$

It is convenient to introduce the magnon destruction operator:

$$c_k = \frac{1}{\sqrt{2}} \left\{ \frac{s^2[\mathfrak{J}(0) - \mathfrak{J}(\mathbf{k})]}{A + \mathfrak{J}(0) - \mathfrak{J}(\mathbf{k})} \right\}^{1/4} \varphi_k + \frac{i}{\sqrt{2}} \left\{ \frac{A + \mathfrak{J}(0) - \mathfrak{J}(\mathbf{k})}{s^2[\mathfrak{J}(0) - \mathfrak{J}(\mathbf{k})]} \right\}^{1/4} S_k^z \quad (\text{A.2})$$

in terms of which the hamiltonian reads:

$$\begin{aligned} \mathcal{H}^{(4)} &= \frac{1}{16AN} \sum_{kq q'} Ja^4 k^2 \sqrt{qq' |\mathbf{k} - \mathbf{q}| |\mathbf{k} - \mathbf{q}'|} (c_q - c_{-q}^+) (c_{k-q} - c_{q'-k}^+) (c_{-q'} - c_{q'}^+) (c_{q'-k} - c_{k'-q'}^+) - \\ &\quad - \frac{Ja}{8N} \sum_{akq q'} \frac{(\mathbf{q} \cdot \mathbf{a})}{\sqrt{aq}} \frac{\mathbf{a} \cdot (\mathbf{k} - \mathbf{q})}{\sqrt{a|\mathbf{k} - \mathbf{q}|}} \sqrt{q' |\mathbf{q}' - \mathbf{k}|} (c_{-q}^+ + c_q) (c_{q'-k}^+ + c_{k-q}) (c_{q'}^+ - c_{-q'}) (c_{k-q}^+ - c_{q'-k}) \\ &\quad - \frac{A}{96N} \sum_{aq q' k} \frac{\mathbf{q} \cdot \mathbf{a}}{\sqrt{aq}} \frac{\mathbf{q}' \cdot \mathbf{a}}{\sqrt{aq'}} \frac{(\mathbf{q} - \mathbf{k}) \cdot \mathbf{a}}{\sqrt{a|\mathbf{q} - \mathbf{k}|}} \frac{(\mathbf{q}' - \mathbf{k}) \cdot \mathbf{a}}{\sqrt{a|\mathbf{q}' - \mathbf{k}|}} (c_{-q}^+ + c_q) (c_{q'}^+ + c_{-q'}) (c_{q'-k}^+ + c_{k-q}) (c_{k'-q'}^+ + c_{q'-k}). \end{aligned} \quad (\text{A.3})$$

The equations of motion are:

$$\dot{c}_k^+ = \frac{i}{\hbar} [c_k^+, (\mathcal{H} + \mathcal{H}^{(4)})] = i(\omega_k + \Delta\omega_k) c_k^+ + if_k^+.$$

Here the Hartree shift  $\Delta\omega_k$  comes from the commutation of  $c_k^+$  with the terms of  $\mathcal{H}^{(4)}$  which contain twice the same wave vector;  $f_k^+$  comes from the

commutation of  $c_k^+$  with the other terms of  $H(4)$  and is a sum of products of 3 creation or destruction operators with 3 different wave vectors.  $f_k^+$  will be identified with Mori's operator  $f_1^+(t)$  [36]. Thus, the Laplace transform

$$(c_k, c_k^+) \Xi_k(z) \text{ of } (c_k, c_k^+(t))$$

is given by Mori's eq. (3.6 in ref. [36]) :

$$\bar{\Xi}_k(z) = \frac{1}{z - i(\omega_k + \Delta\omega_k) + \Sigma_k(z)}$$

with :

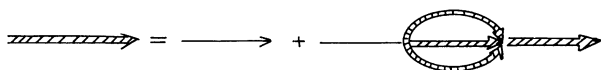
$$\Sigma_k(z) = \frac{1}{(c_k^+, c_k)} \int_0^\infty e^{-zt} (f_k, f_k^+(t)) dt .$$

The relaxation time is therefore given by :

$$\frac{1}{\tau_k} = \text{Re} \Sigma_k(i\omega_k) = \frac{1}{(c_k^+, c_k)} \text{Re} \int_0^\infty e^{-i\omega_k t} (f_k, f_k^+(t)) dt$$

$(f_k, f_k^+(t))$  can then be evaluated by a factorization approximation.

**2. The linear chain.** — 2.1 DAMPING IN THE GROUND STATE. — It is directly given by the Dyson equation :



In the linear part of the spectrum, the energy-momentum conservation is identically satisfied, provided that :

$$0 < q, q', k - q - q' < k .$$

The relaxation time is given in order of magnitude by :

$$\frac{1}{\tau_k} \approx \frac{1}{\hbar^2} \left( \frac{J^2}{4A} a^2 k^2 + \frac{J}{2} + \frac{A}{24} \right)^2 \times \frac{4}{4\pi^2} ka \int_0^{ka} dx \int_0^{ka} dy \frac{xy(ka-x-y)}{1/\tau_k}$$

$$\frac{1}{\tau_k} \approx \frac{1}{2\pi\hbar} (ka)^3 \left( \frac{J^2}{4A} a^2 k^2 + \frac{J}{2} + \frac{A}{24} \right) .$$

The relaxation time is independent of  $s$ , whereas the frequency is proportional to  $s$ .

Ions of the damping for larger  $q$  have not been made. In view of the fact that no ground-state damping has ever been observed in CsNiF<sub>3</sub>, this is not too surprising considering the results of paragraph 7.

**2.2 DAMPING OF SUB-THERMAL MAGNONS.** —  $(f_k, f_k^+(t))$  can be approximated by  $\beta \langle f_k f_k^+(t) \rangle$ , which can be evaluated by means of (A.3) and (A.4) as a sum of 6-fold correlation functions which can be evaluated by a decoupling approximation, for instance :

$$\langle c_{k_1}^+ c_{k_2} c_{k_3} c_{k_1'}(t) c_{k_2'}^+(t) c_{k_3'}^+(t) \rangle = (\delta_{k_2 k_2'} \delta_{k_3 k_3'} + \delta_{k_2 k_3'} \delta_{k_3 k_2}) \delta_{k_1 k_1'} \langle c_{k_1}^+ c_{k_1}(t) \rangle \langle c_{k_2} c_{k_2}(t) \rangle \langle c_{k_3} c_{k_3}(t) \rangle .$$

The inverse relaxation time is found to be proportional to :

$$\frac{1}{N^2} \frac{ka}{\langle n_k \rangle} \sum_{qq'} \text{Re} \frac{qq' |k - q - q'| \langle n_q \rangle \langle n_{q'} \rangle \langle n_{k-q-q'} \rangle}{i(\omega_q + \omega_{q'} + \omega_{k-q-q'} - \omega_k) + \frac{1}{\tau_q} + \frac{1}{\tau_{q'}} + \frac{1}{\tau_{k-q-q'}}} . \quad (\text{A.4})$$

In the linear region of the spectrum, energy-momentum conservation is again identically satisfied and the relaxation time is found to be proportional to  $(Tk^2)^{-1}$  ; this contribution is only relevant if it is larger than the zero-point contribution of the last paragraph.

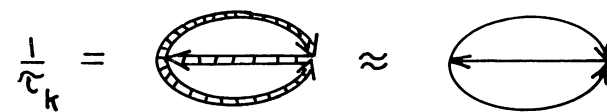
**2.3 DAMPING OF SUPER-THERMAL MAGNONS.** — It turns out that the formula above is still correct. In the linear part of the spectrum, the relaxation time is found to be proportional to  $T^{-2}/\omega_k$ , in agreement with (70).

**3. Two-dimensional case.** — As a rule, the damping is weaker at low temperature than in the linear case.

**3.1 GROUND STATE DAMPING.** — The Dyson equation is the same as for  $D = 1$ . Energy-momentum conservation is satisfied if  $q$  and  $q'$  lie inside a cigar of axis  $\mathbf{k}$  and of thickness  $(k/\tau_k)^{1/2}$ . The inverse relaxation time is proportional to a factor  $k^4$ , due to the conservation law, times a factor  $k^3/\tau_k$ , due to integration over  $\mathbf{q}$  and  $\mathbf{q}'$ , times a factor  $\tau_k$ , due to integration over time. The resulting relaxation time is proportional to  $k^{-7}$  in the linear region of the spectrum.

Here again the damping might be appreciable at the zone boundary for small spin, especially in the antiferromagnetic case.

**3.2 HYDRODYNAMIC REGION,  $h\omega_k < K_B T$ .** — The contribution calculated above (due to the decay of a magnon into 3 magnons) would be proportional to  $k^{-5} T^{-2}$ . The dominant contribution comes from the absorption of a magnon with creation of 2 magnons :



The energy-momentum conservation is ensured (in the linear part of the spectrum) if, for given  $\mathbf{q}$ ,  $\mathbf{q}'$  lies on the ellipse through  $\mathbf{k}$ , with foci 0 and  $(\mathbf{k} + \mathbf{q})$ . In eq. (A.4), the conservation law provides a factor  $k$ , the factor  $\langle n_k \rangle^{-1}$  provides a factor  $k/T$ , the 3 factors  $q \langle n_q \rangle$  yield a factor  $T^3$ , and it turns out that the integration over  $q$  and  $q'$  yield another factor  $T^3$ , so that finally the relaxation time is proportional to  $T^{-5} k^{-2}$ .

## APPENDIX B

Function (27) can be written in the small  $k$  approximation as :

$$\begin{aligned} \gamma_{\text{RR}}(t) &\equiv \gamma(r, t) = \exp - f(\mathbf{r}, t) \\ f(\mathbf{r}, t) &= \frac{1}{8 \pi^2 s^2} \frac{\hbar c}{|J|} \int_0^{\pi/a} \frac{dk}{e^{\beta \hbar c k} - 1} \int_0^{2\pi} d\varphi [1 - \cos ckt \cos(kr \cos \varphi)] \\ &= \frac{\hbar c}{4 \pi |J| s^2} \int_0^{\pi/a} \frac{dk}{e^{\beta \hbar c k} - 1} [1 - J_0(kr) \cos ckt]. \end{aligned}$$

We now choose an arbitrary distance  $ct_0 \gg a$ , and write :

$$f(r, t) = \tau [g_0(T) + g(c, r, t)] \quad (\text{B.1})$$

$$g_0(T) = \int_0^{\pi/a} \frac{dk}{k} [1 - \cos ckt_0] \frac{\beta \hbar c k}{e^{\beta \hbar c k} - 1} \quad (\text{B.2})$$

$$g(c, r, t) = \int_0^{\pi/a} \frac{dk}{k} [\cos ckt_0 - J_0(kr) \cos ckt] \frac{\beta \hbar c k}{e^{\beta \hbar c k} - 1}$$

$$g(c, r, t) \simeq \int_0^{\infty} \frac{dk}{k} [\cos ckt_0 - J_0(kr) \cos ckt]. \quad (\text{B.3})$$

Using eq. (11.4.39) of reference [37], it is possible to evaluate the derivative :

$$\frac{\partial}{\partial c} g(c, r, t) \simeq \int_0^{\infty} [t \sin ckt J_0(kr) - t_0 \sin ckt_0] dk \begin{cases} = \frac{1}{\sqrt{c^2 - r^2/t^2}} - \frac{1}{c} & (ct > r) \\ = -\frac{1}{c} & (ct < r). \end{cases} \quad (\text{B.4})$$

For  $c = \infty$ ,  $J_0(kr)$  can be replaced by 1 in (B.3) and  $g(\infty, r, t)$  is found from eq. (858.51) of reference [25] :

$$g(\infty, r, t) = \int_0^{\infty} (\cos ckt_0 - \cos ckt) \frac{dk}{k} = \text{Log} \frac{t}{t_0}.$$

The calculation of  $g(c, r, t)$  from this equation and (B.4) is straightforward :

$$g(c, r, t) = \text{Log} \frac{ct + \sqrt{c^2 t^2 - r^2}}{2 ct_0} \quad (ct \geq r) \quad (\text{B.5})$$

$$g(c, r, t) = \text{Log} \frac{r}{2 ct_0} \quad (ct \leq r).$$

$g_0(T)$  will now be calculated in the low temperature limit, where (B.2) reads :

$$\begin{aligned} g_0(T) &= \int_0^{\pi/a} \beta \hbar c dk \frac{1 - \cos ckt_0}{e^{\beta \hbar c k} - 1} \simeq \beta \hbar c \int_0^{\infty} \frac{1 - \cos ckt_0}{e^{\beta \hbar c k} - 1} dk = \int_0^{\infty} \frac{1 - \cos t_0 x / \beta \hbar}{e^x - 1} dx \\ &= \int_0^{\infty} \left( \frac{1}{e^x - 1} - \frac{\cos x}{x} \right) dx + \int_0^{\infty} \frac{\cos x - \cos xt_0 K_B T / \hbar}{x} dx + \int_0^{\infty} \left( \frac{1}{x} - \frac{1}{e^x - 1} \right) \cos xt_0 K_B T / \hbar dx \\ &= C + \frac{1}{2} \psi(iK_B T t_0 / \hbar) + \frac{1}{2} \psi(-iK_B T t_0 / \hbar). \end{aligned} \quad (\text{B.6})$$

Use has been made of eq. (3.951.6) and (3.951) of references [38], [25] and eq. (858.51) of reference [25].  $C = 0.577$  is the Euler constant and if  $t_0$  is large, one has [39], eq. (1.7.1) and (1.18.1) :

$$\psi(z) \simeq \text{Log} z.$$

Eq. (72) follows from (B.1, B.5, B.6, B.7) with

$$\alpha = 2 e^{-C}.$$

It is easily checked that (72) is still correct (but only qualitatively) at higher temperature (classical region).

## APPENDIX C

As we did not succeed in Fourier transforming (72), we shall deduce the long time behaviour of the Fourier transform from general properties of the Fourier transformation. These are more likely available in textbooks, but we could neither find them in elementary books, nor understand advanced treatises. Therefore, the relevant properties are proved below :

One possible method is to write  $\gamma(r, t)$  as

$$\gamma(r, t) = \varphi(r, t) + f(r, t) \quad (\text{C. 1})$$

where  $f(r, t)$  has all its derivatives continuous up to  $f^{(2n)}(r, t)$  and

$$\varphi(r, t) = \sum_{m=1}^n a_m \frac{(c^2 t^2 - r^2)^{m/2}}{(ct)^{m+\tau}} \quad (r < ct)$$

$$\varphi(r, t) = 0 \quad (r > ct).$$

The existence of the identity (C.1) is easily checked from the Taylor expansions of  $\gamma(r, t)$  near  $r = ct$ . For instance :

$$a_1 = -\tau, \quad a_2 = \frac{1}{2}(\tau - 1)^2$$

$\gamma(r, t)$ ,  $\varphi(r, t)$  and  $f(r, t)$  are of the form

$$\frac{1}{r^\tau} \times \text{a function of } \frac{r}{ct}.$$

Therefore their FT is of the form

$$\frac{1}{q^{2-\tau}} \times \text{a function of } cqt.$$

Note that the FT are defined in the continuous plane, e. g. :

$$\tilde{\gamma}(q, t) = \frac{1}{a^2} \int \gamma(r, t) e^{iq \cdot r} d^3 r.$$

Thus  $q$  can go to infinity, and the behaviour of  $\tilde{\gamma}(q, t)$  at large  $t$  for given  $q$  can be deduced from the behaviour of  $\tilde{\gamma}(q, t)$  at large  $q$  for given  $t$ , because

$$\tilde{\gamma}(q, t) = \frac{1}{q^{2-\tau}} g(cqt).$$

Now the integration of the expansion

$$\nabla^{2n}(f e^{iq \cdot r}) = (\nabla^{2n} f) e^{iq \cdot r} + \dots + (-q^2)^n f e^{iq \cdot r}$$

shows that

$$\int f(r, t) e^{iq \cdot r} d^2 r$$

vanishes at least as fast as  $1/q^{2n}$  as  $q$  goes to  $\infty$  at finite  $t$ .

Therefore the large  $q$  behaviour of  $\gamma(q, t)$  is determined by that of :

$$\begin{aligned} \tilde{\varphi}(q, t) &= \sum_{m=1}^{2n} \frac{a_m}{(ct)^{m+\tau}} \int_0^{ct} dx \cos qx \int_0^{\pi/2} (c^2 t^2 - x^2)^{(m+1)/2} \cos^{m+1} \theta d\theta \\ &= \sum_{m=1}^{2n} \frac{\sqrt{\pi}}{2} \frac{\Gamma\left(\frac{m}{2} + 1\right)}{\Gamma\left(\frac{m+3}{2}\right)} \frac{a_m}{(ct)^{m+\tau}} \int_0^{ct} dx \cos qx (c^2 t^2 - x^2)^{(m+1)/2}. \end{aligned}$$

It is easily seen, for instance, by calculation of these integrals, that the asymptotic behaviour of  $\varphi(q, t)$  is dominated by the term  $m = 1$ . From eq. (1.3.2) of reference [38] one has :

$$\int_0^{ct} dx (\cos qx) (c^2 t^2 - x^2) = \frac{1}{2} [\Phi(1, 2, iy) + \Phi(1, 2, -iy)] - \frac{1}{6} [\Phi(3, 4, iy) + \Phi(3, 4, -iy)].$$

From the asymptotic expansion of Kummer's hypergeometric series ([39], § 6.13) one obtains the long-time expression

$$\gamma(q, t) \simeq \tilde{\varphi}(q, t) \simeq \frac{3 \tau \sqrt{\pi}}{2} \frac{\Gamma(3/2)}{\Gamma(2)} \frac{\cos cqt}{q^2 (ct)^\tau}.$$

Taking the Fourier transform of this function ([38], eq. (1.3.2)), it is found that for  $\omega \simeq \omega_q$  :

$$S(q, \omega) \simeq \frac{C^\tau}{|\omega - \omega_q|^{1-\tau}} \quad (\tau < 1)$$

$$\frac{d}{d\omega} S(q, \omega) \simeq \frac{C^\tau}{|\omega - \omega_q|^{2-\tau}} \frac{qc - \omega}{|qc - \omega|} \quad (1 < \tau < 2)$$

$$S(q, \omega) \simeq C^\tau \cdot \text{Log} \frac{1}{|\omega - \omega_q|} \quad (\tau = 1)$$

where all 3 constants are positive.

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