

Subject classification: 2 ; 18 . 2

MAGNETIC EXCITATIONS IN AMORPHOUS FERROMAGNETS

by

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ABSTRACT

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~~We study the~~ propagation of magnetic excitations in amorphous  
ferromagnets from the point of view of the theory of random frequency  
modulation. *H.U.* ~~We show~~ that the spin waves in the hydrodynamic limit are well  
described by perturbation theory while the roton-like magnetic excitations  
with wavevector about the peak in the structure factor are not. A criterion  
of validity of perturbation theory is found which is identical to a  
narrowing condition in magnetic resonance.

Nous étudions la propagation des excitations magnétiques dans les so-  
lides amorphes ferromagnétiques du point de vue de la théorie de modulation  
aléatoire de fréquence. Nous montrons que les ondes de spins dans le li-  
mite hydrodynamique sont bien décrites par la théorie de perturbation, et  
que les excitations magnétiques rotoniques, de vecteurs d'onde près du  
maximum du facteur de structure, ne le sont pas. On obtient une condition  
de validité de la théorie de perturbation qui est identique à la condition  
de diminution de la largeur de la ligne ("narrowing condition") dans la  
théorie de résonance magnétique.

## INTRODUCTION

The study of amorphous ferromagnets has been intense in the last years, both theoretically and experimentally.

The early theories of amorphous ferromagnets took essentially a mean field approach to the problem [1][2], and neglected dynamical effects. Later however the equation of motion method has been used to investigate the elementary excitations in a ferromagnetic glass and the stability of its ground state [3][4][5][6].

From the experimental point of view the existence of spin waves in these glasses was confirmed both directly through inelastic neutron scattering [7][8] and indirectly through the observation of a  $T^{3/2}$  term in the decrease of the magnetization at low temperatures [9][10]. Unexpectedly the neutron scattering experiments revealed the existence of roton-like magnetic excitations with wave vector about the peak in the structure factor in the amorphous ferromagnets CoP [7] and FePC [11].

The question of the existence of spin waves in an amorphous ferromagnet is not academic. This solid possesses a different kind of translation invariance, namely every site in the glass share with the others the property, at least ideally, that they don't have anything in common. The full complexity of the dynamics of amorphous ferromagnets is revealed by the discrepancy on the values of the spin wave stiffness as obtained by different kind of measurements [12], and in the anomalously large coefficient of the  $T^{3/2}$  term in the magnetization which extends beyond half the Curie point [10].

In this paper we study the dynamics of an amorphous ferromagnet from the point of view of the theory of random frequency modulation. A preliminary approach to this problem was made by Continentino and Rivier [13].

The Hamiltonian and Green's function

The Hamiltonian describing the amorphous ferromagnet is the standard Heisenberg Hamiltonian

$$H = - 1/2 \sum_{i,j} J_{ij} \underline{S}_i \cdot \underline{S}_j \quad (1)$$

where the spin  $\underline{S}_i$  are distributed in a Bernal structure [14]. The interaction  $J_{ij}$  between magnetic moments may extend beyond nearest neighbours and oscillate due to the metallic nature of the glass.

We now introduce, for a given configuration of the magnetic glass, the double time, temperature dependent Green's function

$$G_{ij}^{+-}(t, t') = \langle\langle S_i^+(t) | S_j^-(t') \rangle\rangle = i\theta(t-t') \langle [S_i^+(t), S_j^-(t')] \rangle \quad (2)$$

where the spin operators are given in a Heisenberg representation and  $\theta(t)$  is the step function. The symbol  $\langle \quad \rangle$  stands for a thermal average.

Within the Random Phase Approximation (R.P.A.) the Green's function satisfies the following equation of motion

$$i \frac{d}{dt} G_{ij}^{+-}(t) = -2\sigma\delta_{ij} \delta(t) - \sigma \sum_{\ell} J_{i\ell} G_{\ell j}^{+-}(t) + \sigma \sum_{\ell} J_{i\ell} G_{ij}^{+-}(t) \quad (3)$$

The magnetization  $\langle S^2 \rangle \equiv \sigma$  is assumed to be uniform in the spirit of the R.P.A. This is a reasonable approximation at sufficient low temperatures.

A formal solution of the equation of motion (3) can be immediately found. It is given by [15]

$$G_{ij}^{+-}(t) = 2i\sigma \theta(t) \{ \exp[i\sigma t \hat{\Omega}] \}_{ij} \quad (4)$$

where the matrix  $\hat{\Omega}$  is defined by

$$(\hat{\Omega})_{ij} = J_{ij} - \delta_{ij} \sum_l J_{il} \quad (5)$$

As will become clear below it is more convenient to deal with the Fourier transform of the Green's function  $G_{ij}^{+-}(t)$ , that is

$$G_{\underline{k}\underline{k}'}^{+-}(t) = (1/N) \sum_i \sum_j e^{-i\underline{k}\cdot\underline{R}_i} e^{i\underline{k}'\cdot\underline{R}_j} G_{ij}^{+-}(t) \quad (6)$$

This transformation can be applied to each term on the series represented by the exponential on the right hand side of equation (4) and one finally obtains

$$G_{\underline{k}\underline{k}'}^{+-}(t) = 2i\sigma\theta(t)\rho(\underline{k}-\underline{k}') \langle \underline{k} | \exp[-i\sigma t \sum_{\substack{\underline{k}_1 \underline{k}_2 \\ -1 -2}} \Omega_{\underline{k}_1 \underline{k}_2} | \underline{k}_1 \rangle \langle \underline{k}_2 | ] | \underline{k}' \rangle \quad (7)$$

where

$$\rho(\underline{k} - \underline{k}') = (1/N) \sum_i e^{-i(\underline{k} - \underline{k}')\cdot\underline{R}_i} \quad (8)$$

and

$$\Omega_{\underline{k}_1 \underline{k}_2} = (1/N) \sum_i e^{-i(\underline{k}_1 - \underline{k}_2)\cdot\underline{R}_i} \sum_j J_{ij} (1 - e^{-i\underline{k}_2\cdot(\underline{R}_i - \underline{R}_j)}) \quad (9)$$

with  $N$  the number of magnetic ions. We have chosen to write the Fourier transformed Green's function as the expectation value of an operator instead of as a matrix element. The kets  $|\underline{k}\rangle$  span an abstract Hilbert space and are orthonormalised. In (6) and (7),  $G_{\underline{k}\underline{k}'}^{+-}(t)$  corresponds to a given configuration. The ensemble averaged Green's

function will be diagonal in the  $\underline{k}$ -representation, which therefore imposes itself naturally at this stage.

The Green's function (7) obeys the standard equation of motion for disordered Heisenberg ferromagnets [5][16].

We should point out that in the amorphous metal due to the absence of structural periodicity there is not a well defined Brillouin zone and one must therefore use the concept of extended zone scheme. Symmetry considerations however lead to the conclusion that the surfaces of constant energy in wave vector space are sphericals.

The allowed values of  $\underline{k}$  are determined by imposing periodic boundary conditions to apply over the volume  $V$  of the amorphous sample so that the density of allowed  $\underline{k}$  values in wave vector space remains uniform and equal to  $V/(2\pi)^3$ .

#### AVERAGING PROCEDURE

The Green's function  $G_{\underline{k}\underline{k}}^{+-}(t)$  involves a summation over the magnetic ions and therefore must be averaged over all possible configurations. This average may be evaluated directly and in order to do that it is useful to rewrite the evolution operator in (7) in an "interaction" representation. For this purpose we define a diagonal operator  $\hat{\Omega}_0$  by

$$\hat{\Omega}_0 = \sum_{\underline{k}_1 \underline{k}_2} \langle \Omega_{\underline{k}_1 \underline{k}_2} \rangle_{AV} |\underline{k}_1\rangle \langle \underline{k}_2| \quad (10)$$

with  $\Omega_{\underline{k}_1 \underline{k}_2}$  given by (9). The symbol  $\langle \rangle_{AV}$  means a configurational average. As we will show later  $\langle \Omega_{\underline{k}_1 \underline{k}_2} \rangle_{AV} = \langle \Omega_{\underline{k}_1} \rangle_{AV} \delta_{\underline{k}_1 \underline{k}_2}$ , then

$$\hat{\Omega}_0 = \sum_{\underline{k}_1} \omega(\underline{k}_1) |\underline{k}_1\rangle \langle \underline{k}_1| \quad (11)$$

where we defined  $\omega(\underline{k}) = \langle \Omega_{\underline{k}_1} \rangle_{AV}$ . Finally one gets in the "interaction" representation the following operator identity

$$\exp(-i\sigma \hat{\Omega} t) = \exp(-i\sigma \hat{\Omega}_0 t) T \exp\{-i\sigma \int_0^t d\tau \hat{\Omega}_1(\tau)\} \quad (12)$$

In (12)  $T$  is the time ordering operator [17] and the operator  $\hat{\Omega}_1 = \hat{\Omega} - \hat{\Omega}_0$  has a time dependence given by

$$\hat{\Omega}_1(\tau) = e^{-i\sigma \hat{\Omega}_0 \tau} (\hat{\Omega} - \hat{\Omega}_0) e^{i\sigma \hat{\Omega}_0 \tau} \quad (13)$$

with  $\hat{\Omega} = \sum_{\underline{k}_1 \underline{k}_2} \Omega_{\underline{k}_1 \underline{k}_2} |\underline{k}_1\rangle \langle \underline{k}_2|$  and  $\hat{\Omega}_0$  as defined in (11).

We then have for the average Green's function

$$\langle G_{\underline{k} \underline{k}'}^{+-}(t) \rangle_{AV} = 2i\sigma \theta(t) e^{-i\sigma \omega(\underline{k})t} \langle \rho(\underline{k} - \underline{k}') \langle \underline{k} | T \exp\{-i\sigma \int_0^t d\tau \hat{\Omega}_1(\tau)\} | \underline{k}' \rangle \rangle_{AV} \quad (14)$$

In order to calculate this average we introduce the following decoupling:

$$\begin{aligned} \langle \rho(\underline{k} - \underline{k}') \langle \underline{k} | T \exp\{-i\sigma \int_0^t d\tau \hat{\Omega}_1(\tau)\} | \underline{k}' \rangle \rangle_{AV} &= \\ \langle \rho(\underline{k} - \underline{k}') \rangle_{AV} \langle \langle \underline{k} | T \exp\{-i\sigma \int_0^t d\tau \hat{\Omega}_1(\tau)\} | \underline{k}' \rangle \rangle_{AV} &= \end{aligned} \quad (15)$$

$$= \delta_{\underline{k}\underline{k}'} \langle\langle \underline{k} | T \exp\{-i\sigma \int_0^t \hat{\Omega}_1(\tau) d\tau\} | \underline{k}' \rangle\rangle_{AV}$$

We rewrite the average Green's function as

$$\langle G_{\underline{k}}^{+-}(t) \rangle_{AV} = 2i\sigma\theta(t) e^{-i\sigma\omega(k)t} \langle\langle \underline{k} | T \exp\{-i\sigma \int_0^t \hat{\Omega}_1(\tau) d\tau\} | \underline{k} \rangle\rangle_{AV} \quad (16)$$

The perturbation  $\hat{\Omega}_1(\tau)$  represents a modulation of the spin wave frequency  $\omega(k)$  and can be viewed as a stochastic process, due to the structural disorder of the amorphous, which may be averaged over an ensemble. The study of spin wave propagation in a ferromagnetic glass has then been reduced to a random frequency modulation problem.

Let us define an averaging operation for operators according to

$$((\hat{A})) = \langle\langle \underline{k} | \hat{A} | \underline{k} \rangle\rangle_{AV} \quad (17)$$

Notice that

$$((\hat{1})) = 1 \quad (18)$$

and

$$((a\hat{A} + b\hat{B})) = a((\hat{A})) + b((\hat{B})) \quad (19)$$

is a linear operation. Then  $((\dots))$  is indeed a proper average.

We rewrite once again the average spin wave Green's function, now

as

$$\langle G_{\underline{k}}^{+-}(t) \rangle_{AV} = 2i\sigma\theta(t) e^{-i\sigma\omega(k)t} ((T \exp\{-i\sigma \int_0^t \hat{\Omega}_1(\tau) d\tau\})) \quad (20)$$

The average of the operator on the right hand side of the above equation may be evaluated using a generalized cumulant expansion introduced by Kubo [18]. We shall consider the simplest random sequence of operators which includes correlation, namely a Gaussian ensemble. Since a Gaussian average corresponds to taking terms up to second order in the cumulant expansion, it yields

$$\begin{aligned} & \left( \left( T \exp \left\{ -i\sigma \int_0^t d\tau \hat{\Omega}_1(\tau) \right\} \right) \right) = \\ & \exp \left\{ -i\sigma \int_0^t d\tau_1 \left( \hat{\Omega}_1(\tau_1) \right) \right\}_c - \frac{1}{2} \sigma^2 \int_0^t d\tau_1 \int_0^t d\tau_2 \left( \left( T [\hat{\Omega}_1(\tau_1) \hat{\Omega}_1(\tau_2)] \right) \right)_c \end{aligned} \quad (21)$$

where  $T[\hat{\Omega}_1(\tau_1) \hat{\Omega}_1(\tau_2)]$  means the time ordered product of these operators. The cumulant averages  $(( \quad ))_c$  are defined in the usual way [18]

$$\begin{aligned} \left( \left( \hat{\Omega}_1(\tau_1) \right) \right)_c &= \left( \left( [\hat{\Omega}_1(\tau_1) - \hat{\Omega}_0(\tau_1)] \right) \right)_c = \left( \left( [\hat{\Omega}_1(\tau_1) - \hat{\Omega}_0(\tau_1)] \right) \right) = \left( \left( \hat{\Omega}_1(\tau_1) \right) \right) = 0 \end{aligned} \quad (22)$$

The last equality follows from the definition of  $\hat{\Omega}_0$ . The second order term is given by

$$\begin{aligned} & \left( \left( T [\hat{\Omega}_1(\tau_1) \hat{\Omega}_1(\tau_2)] \right) \right)_c = \left( \left( T [\hat{\Omega}_1(\tau_1) - \hat{\Omega}_0(\tau_1)] [\hat{\Omega}_1(\tau_2) - \hat{\Omega}_0(\tau_2)] \right) \right)_c = \\ & \left( \left( T [\hat{\Omega}_1(\tau_1) - \hat{\Omega}_0(\tau_1)] [\hat{\Omega}_1(\tau_2) - \hat{\Omega}_0(\tau_2)] \right) \right) - \left( \left( [\hat{\Omega}_1(\tau_1) - \hat{\Omega}_0(\tau_1)] \right) \right) \\ & \left( \left( [\hat{\Omega}_1(\tau_2) - \hat{\Omega}_0(\tau_2)] \right) \right) = \left( \left( T [\hat{\Omega}_1(\tau_1) - \hat{\Omega}_0(\tau_1)] [\hat{\Omega}_1(\tau_2) - \hat{\Omega}_0(\tau_2)] \right) \right) \\ & = \left( \left( T [\hat{\Omega}_1(\tau_1) \hat{\Omega}_1(\tau_2)] \right) \right) \end{aligned} \quad (23)$$

where we have used (22), and the average  $((\dots))$  is defined by (17).

Finally one has for the average expectation value of the time ordered operator in equation (16)

$$\begin{aligned} ((T \exp\{-i\sigma \int_0^t d\tau \hat{\Omega}_1(\tau)\}) &= \exp\{-\frac{1}{2}\sigma^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 ((T[\hat{\Omega}_1(\tau_1)\hat{\Omega}_1(\tau_2)])\}) \\ &= \exp(-\gamma(t)) \end{aligned} \quad (24)$$

in the Gaussian approximation. The last equality defines  $\gamma(t)$ .

Evaluating the time ordering operation it can be written as

$$\gamma(t) = \sigma^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 ((\hat{\Omega}_1(\tau_1)\hat{\Omega}_1(\tau_2))) \quad (25)$$

If the correlation function  $F(\tau_1 - \tau_2) = ((\hat{\Omega}_1(\tau_1)\hat{\Omega}_1(\tau_2)))$  depends only on  $\tau = \tau_1 - \tau_2$ , which actually is the case as can be seen using the  $|k\rangle$  representation for the operators  $\hat{\Omega}_1(\tau)$ , then one has

$$\gamma(t) = \sigma^2 \int_0^t (t - \tau)F(\tau)d\tau \quad (26)$$

The quantity  $\gamma(t)$  contains besides other parts, the results of perturbation theory in the second order, that is the energy corrections and transition probability calculated in this approximation. Let us look at it however from the point of view of the stochastic theory of frequency modulation. In this context it represents essentially the modulation of the spin wave propagation (given by the term  $\exp(-i\omega(k)t)$  in (16)) by the Gaussian random process  $\hat{\Omega}_1(\tau)$ .

The correlation function  $F(\tau)$  of the stochastic process  $\hat{\Omega}_1(\tau)$  can be described by two parameters [19]

An amplitude (k-dependent) defined by

$$\Delta^2(k) = F(\tau = 0) \quad (27)$$

which can be obtained using the  $|k\rangle$  representation for the operators  $\hat{\Omega}_1(\tau)$  and the definition (17) for the averaging operation represented by  $(\dots)$ . One then gets

$$\Delta^2(k) = \sum_{\underline{k}_1} \langle \Omega_{\underline{k}\underline{k}_1} \Omega_{\underline{k}_1\underline{k}} \rangle_{AV} - (\omega(k))^2 \quad (28)$$

with  $\omega(k) = \langle \Omega_{\underline{k}\underline{k}_1} \rangle_{AV} = \langle \Omega_{\underline{k}} \rangle_{AV} \delta_{\underline{k}\underline{k}_1}$  as before. We mention here and will show later that  $\Delta(k) \propto k$  in the limit  $k \rightarrow 0$ .

The other parameter associated with  $F(\tau)$  is a k-dependent correlation time  $\tau_c(k)$  that we define as

$$\tau_c(k) = \frac{\int_{-\infty}^{+\infty} (\langle \hat{\Omega}_1(\tau) \hat{\Omega}_1(0) \rangle) d\tau}{2\Delta^2(k)} \quad (29)$$

A calculation using the  $|k\rangle$  representation for the operators  $\hat{\Omega}_1(\tau)$  yields

$$\tau_c(k) = \frac{2\pi \sum_{\underline{k}_1} \delta(\omega(k) - \omega(k_1)) [\langle \Omega_{\underline{k}\underline{k}_1} \Omega_{\underline{k}_1\underline{k}} \rangle_{AV} - (\omega(k))^2]}{2\Delta^2(k)}$$

(30)

The correlation time  $\tau_c(k)$  is a measure of the speed of modulation [19] and is defined so that the correlation function  $(\langle \hat{\Omega}_1(\tau) \hat{\Omega}_1(0) \rangle) \sim 0$  for  $\tau \gg \tau_c$ .

The correlation time  $\tau_c(k) \propto k^3$  in the limit  $k \rightarrow 0$  as will be shown below.

We can distinguish two different behaviours of  $\gamma(t)$  corresponding to two regions of time

1)  $t \ll \tau_c$

In this region the correlation function  $F(\tau)$  may be considered constant and approximately equal to  $F(\tau = 0)$ . Hence for  $t \ll \tau_c$ ,  $\gamma(t)$  behaves like

$$\gamma(t) = \frac{1}{2} \sigma^2 \Delta^2(k) t^2 \quad (31)$$

where  $\Delta^2(k)$  is given by (18). In the short time regime then the average spin wave Green's function is a Gaussian

$$\langle G_k^{+-}(t) \rangle_{AV} = 2i\sigma\theta(t) \exp[-i\sigma\omega(k)t - \frac{1}{2} \sigma^2 \Delta^2(k) t^2] \quad (32)$$

and the corresponding spectral density will also be Gaussian.

However, in this regime the damping rate  $\Delta(k) \propto k$  in the limit  $k \rightarrow 0$ , implying that the spin wave excitations with energy  $\sigma\omega(k \rightarrow 0) = \sigma D k^2$  are overdamped and consequently not well defined in the long wavelength limit. This is an unphysical result since we have enough direct and indirect evidence confirming the existence of spin waves in the amorphous ferromagnet.

2)  $t \gg \tau_c$

The correct description of the hydrodynamics limit is given by the long time behaviour of the Green's function.

Since  $F(\tau) \sim 0$  for  $\tau \gg \tau_c$  one can push the limit of the integral in (26) to infinity and obtain

$$\gamma(t) = \sigma^2 |t| \int_0^\infty F(\tau) d\tau + \underline{cte} \quad (33)$$

The first term in  $\gamma(t)$  contains essentially the results of perturbation theory [20]. The integral of  $F(\tau)$  has an imaginary part which gives the correction of the spin wave energy to second order. It's real part provides the relaxation of these excitations, and can be written as

$$\gamma_R(t) = \sigma^2 |t| \Delta^2(k) \tau_c(k) \quad (34)$$

where  $\Delta$  and  $\tau_c$  have been defined previously in (28) and (29) respectively. Then in the long time region the spin wave Green's function is given by

$$\langle G_k^{+-}(t) \rangle_{AV} = 2i\sigma\theta(t) \exp[-i\sigma\omega(k)t - \sigma^2\Gamma(k)t] \quad (35)$$

and the corresponding spectral density will be a Lorentzian. The damping term  $\Gamma(k) = \Delta^2(k) \tau_c(k)$  is proportional to  $k^5$  (see below) in the limit  $k \rightarrow 0$ . This confirms the existence of well defined spin wave excitations in the hydrodynamic limit since their damping  $\Gamma(k) \propto k^5$  is much less than their energy  $\sigma\omega(k) \propto k^2$

in this limit. This fact as we show later will guarantee the validity of Bloch's  $T^{3/2}$  law associated with the decrease of the magnetization at low temperatures. In (35) we have neglected the energy correction which comes from the imaginary part of  $\gamma(t)$ .

The constant that appears in (33) is related to virtual transitions and has nothing to do with real propagation or relaxation.

If we pursue our analogy with the random frequency modulation problem we may distinguish two typical situations depending on the relative magnitudes of  $\Delta$  and  $\tau_c$  [19].

a) slow modulation ( $\Delta\tau_c \gg 1$ )

In this case the correlation time  $\tau_c(k)$  is large compared with  $1/\Delta(k)$ . The Green's function may be represented by the Gaussian propagator, and only in the tail corresponding to very large values of time the exponential character sets in. The spectral density in this regime is essentially a Gaussian.

b) fast modulation ( $\Delta\tau_c \ll 1$ )

The correlation time now is small compared with  $1/\Delta$ . The modulation  $\hat{\Omega}_1(\tau)$  lasts for very short times and is hardly effective, so that its effect is averaged out and the spin wave propagator becomes sharp and centered around the spin wave energy. The spectral density is taken as a Lorentzian with centre at  $\sigma\omega(k)$  and half-width  $\sigma^2\Gamma(k) = \sigma^2\Delta^2(k)\tau_c(k)$ . Since this is the regime for which

$$\Delta(k) \tau_c(k) \ll 1$$

(36)

one has therefore

$$\Gamma(k) \ll \Delta(k) \quad (37)$$

so that the spectral density is much narrowed. The condition (36) is indeed called the narrowing condition.

Since the spectral density, obtained from the imaginary part of the Green's function is related to the neutron scattering cross section we have here essentially a simple theory of its line shape. From the  $k$ -dependence of  $\Delta$  and  $\tau_c$  we notice that for large values of  $k$  the criterion for slow modulation may be satisfied and the spectral density will be Gaussian. However as one approaches the hydrodynamic limit the condition of fast modulation (36) is fulfilled. The spectral density is narrowed and becomes of the Lorentzian type. In this region the magnetic system is correctly described by perturbation theory. The criterion of validity for a perturbational approach has then been reduced to a narrowing condition ( $\Delta\tau_c \ll 1$ ). For example in the experimental spectrum of Mook, et al [7]

the small  $k$  branch satisfies the narrowing condition while the roton-like dip does not (see below). This large  $k$  part of the spectrum cannot therefore be described by perturbation theory and as we show below satisfies the condition of slow modulation ( $\Delta\tau_c \gg 1$ ).

We haven't considered the case the correlation time  $\tau_c$  falls in physically relevant regions of time such that  $\Delta\tau_c \sim 1$ . This situation however may be described by an useful interpolation formula which reduces to the results obtained above in the correct limits. That is we choose

$\chi(t)$  in the following way [21]

$$\chi(t) = \sigma^2 (\alpha^2 + t^2)^{1/2} / \eta \quad (38)$$

We notice that taking  $\alpha = \tau_c$  and  $\eta = \tau_c^{-1}$  this expression gives (31) and (34) in the limits of short and Long time respectively.

Finally it is interesting to notice that with the narrowing of the line an irreversible behaviour appears in the magnetic system.

Suppose we excite our spin wave at a different time  $t = t_0$  instead of  $t = 0$ . The average Green's function in the slow modulation case will be

$$\langle G_k^{+-}(t) \rangle_{AV} = 2i\sigma\theta(t-t_0) e^{-i\omega(k)(t-t_0) - \frac{1}{2}\sigma^2 \Delta^2(k)(t-t_0)^2} \quad (39)$$

and in the case of fast modulation

$$\langle G_k^{+-}(t) \rangle_{AV} = 2i\sigma\theta(t-t_0) e^{-i\omega(k)(t-t_0) - \sigma^2 \Gamma(k)(t-t_0)} \quad (40)$$

In this last expression the exponential factor which depends on  $t_0$  is just an arbitrary factor multiplying the Green's function and can be neglected. This is not the case in equation (39), where the system actually keeps memory of the time the excitation was created. This memory is lost and irreversible behaviour sets in when the narrowing condition is fulfilled.

#### SPIN WAVE ENERGY AND DAMPING

In this section we calculate explicitly the quantities we have introduced before.

Firstly the average energy of the spin excitations

$$\begin{aligned} \langle \Omega_{\underline{k}\underline{k}'} \rangle_{AV} &= \langle (1/N) \sum_i e^{-i(\underline{k}-\underline{k}') \cdot \underline{R}_i} \rangle_{AV} \langle \sum_j J_{ij} (1 - e^{-i\underline{k}' \cdot (\underline{R}_i - \underline{R}_j)}) \rangle_{AV} \\ &= \delta_{\underline{k}\underline{k}'} \omega(k) \end{aligned} \quad (41)$$

Introducing the pair distribution function  $g(r)$  of the magnetic atoms in the amorphous magnet, one has [13]

$$\omega(k) = 4\pi\rho_0 \int dR R^2 g(R) J(R) \left(1 - \frac{\sin kR}{kR}\right) \quad (42)$$

The quantity  $\rho_0$  is the average density of magnetic atoms ( $\rho_0 = N/V$ ).

We have made use of the spherical symmetry of the problem to obtain (42).

The energy  $\sigma\omega(k)$  of the magnetic excitations increases initially with  $k$  as  $\sigma Dk^2$  ( $D = (2\pi \rho_0/3) \int dR R^4 g(R) J(R)$ ) to reach with oscillations the mean field limit  $\sigma\omega(k = \infty) = \sigma 4\pi \rho_0 \int dR R^2 g(R) J(R)$ . The later energy is directly related to the Curie temperature ( $T_c$ ) of the ferromagnetic glass calculated in a mean field approximation [2]

$$\omega(k = \infty) = \frac{3k_B T_c}{S(S+1)} \quad (43)$$

Equation (42) represents a relation between magnetic properties namely the spin wave spectrum and structural information contained in  $g(R)$ . Since  $\sin x/x = j_1(x)$  is a spherical Bessel function, equation (42) is a Bessel transform which can be inverted to yield [13]

$$g(R) J(R) = [2\pi^2 \rho_0]^{-1} \int dk k^2 (\sigma\omega(k = \infty) - \sigma\omega(k)) \frac{\sin kR}{kR} \quad (44)$$

This formula may be used to obtain the exchange interaction in amorphous ferromagnets. Expression (42) for the spin wave energy is the magnetic equivalent to the spectrum calculated by Hubbard and Beeby [22] for phonons in liquids. We should emphasize that although it can be obtained by perturbation theory, it is a more general expression valid outside the narrowing regime ( $\omega(k)$  is the same in (39) and (40)). It has also some appealing features, like the correct quadratic dispersion relation in the hydrodynamic region and the possible occurrence of a dip in the spectrum at large wave-vectors. From another point of view which is probably more interesting, one may use equation (44) to extract the product

$g(R) J(R)$  from the experimental spin wave spectrum. For the amorphous ferromagnet Fe PC using formula (43) and the Curie temperature measured by Tsuei and Lilienthal [23] one finds  $\omega(k=\infty) = 75 \text{ me V}$  ( $S = 1$  for Fe).

Proceeding as for (41) we may also calculate the damping of the spin excitations. In the case of slow modulation we have the following expression for the damping [24]

$$\Delta^2(k) = (4\pi \rho_0)^2 \int dr_1 r_1^2 \int dr_2 r_2^2 J(r_1) J(r_2) \{ [1 - j_0(kr_1)] [1 - j_0(kr_2)] \\ [Q_0(r_1, r_2) - g(r_1)g(r_2)] + \sum_{\ell=1}^{\infty} (-1)^{2\ell} j_{\ell}(kr_1) j_{\ell}(kr_2) Q_{\ell}(r_1, r_2) \} \quad (45)$$

where we expanded the triplet correlation function  $g(r_1, r_2, \cos \theta)$  as [25]

$$g_3(r_1, r_2, \cos \theta) = \sum_{\ell=0}^{\infty} Q_{\ell}(r_1, r_2) P_{\ell}(\cos \theta) \quad (46)$$

The  $P_{\ell}(x)$  are Legendre polynomials and the  $j_{\ell}$  in (45) are spherical Bessel functions. The triplet correlation function is as high in the hierarchy of correlation functions as one needs to go to describe completely the magnetic properties of our amorphous ferromagnet since a Gaussian distribution correlates its elements pair wise only. In the hydrodynamic limit the damping  $\Delta^2(k)$  is dominated by the  $\ell = 1$  term

$$\Delta^2(k) = \left[ \frac{(4\pi \rho_0)^2}{9} \int dr_1 r_1^3 \int dr_2 r_2^3 J(r_1) J(r_2) Q_1(r_1, r_2) \right] k^2 \quad (47)$$

Since  $\Delta \propto k$  the magnons are overdamped in this approximation.

In the same way we can calculate the damping term in the fast modulation regime, the correlation time of the random modulation and the second order energy corrections. We are going to do that however in a different approximation that essentially splits the many body correlation functions in products of pair correlation

functions. Since it introduces a disorder parameter we obtain expressions which are very useful to substantiate the results of the last section. We follow Montgomery, Krugler and Stubbs (MKS) [3] and decompose the nearest neighbour exchange interaction in a mean interaction and a term representing fluctuations around this mean

$$J(R_i, R_j) = J_0(R_i - R_j) + j(R_i, R_j) \quad (48)$$

The configurational averages are then calculated using the correlation function

$$\langle j(R_f, R_g) j(R_h, R_k) \rangle_{AV} = j^2 [\delta_{fh} \times \delta_{gk} + \delta_{fk} \times \delta_{gh}] \quad (49)$$

Following this procedure we get for the quantities calculated in the last section. Firstly the damping term in the slow modulation case (in the limit  $k \rightarrow 0$ )

$$\Delta^2(k) = j^2 \rho_0 \langle R^2 \rangle k^2 \quad (50)$$

where

$$\langle R^n \rangle = \int_0^{R_0} dR g(R) (\hat{k} \cdot \underline{R})^n \quad (51)$$

The integral is over the nearest neighbour peak in the pair correlation function and  $\hat{k}$  is a unit vector in the direction of  $\underline{k}$ .

In order to obtain the damping in the fast modulation regime we make the standard approximation

$$\begin{aligned} \Gamma(k) &\cong 2\pi \int_{k_1} \delta(\omega(k) - \omega(k_1)) [\langle \Omega_{kk}^2 \rangle - \langle \Omega_{kk} \rangle^2] \\ &\cong 2\pi N [\langle \Omega_{kk}^2 \rangle - \langle \Omega_{kk} \rangle^2] n(\omega(k)) \end{aligned} \quad (52)$$

where  $n(\omega)$  is the density of states. In the hydrodynamic limit one finds

$$\Gamma(k) = \frac{1}{6\pi} \frac{j^2 \langle R^4 \rangle}{J_0 \langle R^2 \rangle} k^5 \quad (53)$$

which is much smaller than the spin wave energy in the same limit and confirms the validity of perturbation theory in this region.

If we describe the excitations around the wave vector  $\underline{k}_0$ , where the spin wave frequency has a dip, by a parabola

[26] the density of these excitations is given by ( $D_1$  is a stiffness constant)

$$n(\omega_{\text{dip}}(k)) = \frac{1}{(2\pi)^2} \frac{k^2}{D_1 (k - k_0)} \quad (54)$$

and may become very large when  $k \rightarrow k_0$ . As a consequence the excitations at the dip turn out to be overdamped [27],

within the fast modulation regime (perturbation theory), in contradiction with the experimental results [7].

It is interesting to notice that the narrowing condition (36) is not satisfied in this case, since  $\tau_c$  becomes very large (due to the divergence of  $n(\omega)$  at the dip while  $\Delta$  remains finite), implying that perturbation theory is not adequate to describe this region of the spectrum of magnetic excitations.

The roton-like excitations satisfy the condition of slow modulation and are correctly described by the Gaussian spectral density with a finite width  $\Delta$ . Within the MKS [3] approximation the damping of these modes is given, for all  $k$ , by

$$\Delta^2(k) = j^2 16\pi\rho_0 \int dR R^2 g(R) \left(1 - \frac{\sin kR}{kR}\right) \quad (55)$$

The ratio between the energy and width of the excitations at the dip provides then a direct estimate of the relative magnitude of the fluctuations of the exchange interactions, that is  $\omega(k_{dip})/\Delta^2(k_{dip}) = J_0/4j^2$ , a constant independent of  $k$ .

Finally one gets for the  $k$ -dependent correlation time of the random modulation in the long wavelength limit

$$\tau_c(k) = \frac{1}{12\pi\rho_0} \frac{1}{J_0} \frac{\langle R^4 \rangle}{\langle R^2 \rangle^2} k^3 \quad (56)$$

The narrowing condition  $\Delta\tau_c \ll 1$  is given by

$$\frac{1}{12\pi} \sqrt{\frac{\langle R^2 \rangle}{\rho_0}} \frac{\langle R^4 \rangle}{\langle R^2 \rangle^2} \frac{j}{J_0} k^4 \ll 1 \quad (57)$$

which is the criterion of validity of perturbation theory.

#### MAGNETIZATION

In a crystalline ferromagnet at low temperatures the existence of well defined spin waves causes the magnetization to decrease with a  $T^{3/2}$  dependence. In the amorphous ferromagnet how does the damping of the spin waves affect the temperature dependence of the magnetization? In order to answer this question let us consider for simplicity the case of spin 1/2. In this case one has the following identity

$$\sigma = S - \langle\langle S^- S^+ \rangle\rangle_{AV} \quad (58)$$

The quantity  $\langle\langle S^- S^+ \rangle\rangle_{AV}$  is related to the imaginary part of the spin wave Green's function  $\langle\langle G_k^-(\omega) \rangle\rangle_{AV}$  through the Fluctuation-Dissipation theorem. We then have

$$\sigma = S - \frac{1}{N} \sum_k \int_{-\infty}^{+\infty} \frac{\text{Im} \langle G_k^{+-}(\omega) \rangle_{AV}}{e^{\beta\omega} - 1} d\omega \quad (59)$$

where  $\text{Im} \langle G_k^{+-}(\omega) \rangle_{AV}$  is given by

$$\begin{aligned} \text{Im} \langle G_k^{+-}(\omega) \rangle_{AV} &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} 2\sigma e^{-i\sigma\omega(k)t - \sigma^2\Gamma(k)|t|} e^{i\omega t} dt \\ &= 2\sigma \frac{1}{\pi} \frac{\sigma^2\Gamma(k)}{(\omega - \sigma\omega(k))^2 + \sigma^4\Gamma^2(k)} \end{aligned} \quad (60)$$

in the fast modulation case and

$$\text{Im} \langle G_k^{+-}(\omega) \rangle_{AV} = 2\sigma \frac{1}{\sqrt{2\pi} \sigma\Delta(k)} \exp\left[-\frac{(\omega - \sigma\omega(k))^2}{2\sigma^2 \Delta^2(k)}\right] \quad (61)$$

in the slow modulation regime.

Consider first the fast modulation case which describes correctly the dynamics of the amorphous magnetic system in the hydrodynamic limit. Using (60) we rewrite (59) as

$$\frac{\sigma}{S} = \frac{1}{1 + 2P} \quad (62)$$

where

$$P = \frac{V}{N} \frac{1}{(2\pi)^3} \int dk \int_{-\infty}^{+\infty} \frac{1}{\pi} \frac{\sigma\Gamma(k)}{(\omega - \omega(k))^2 + (\sigma\Gamma(k))^2} \frac{1}{e^{\beta\omega} - 1} d\omega \quad (63)$$

If we take  $\omega(k) = Dk^2$  and assume that  $\Gamma(k) = Bk^n$  then whenever  $n$  is larger than 2 ( $n > 2$ ) one has

$$P = \frac{V}{N} \left( \frac{k_B T}{4\pi\sigma D} \right)^{3/2} \xi(3/2) \quad (64)$$

in the limit  $T \rightarrow 0$ . The function  $\xi(x)$  is the Zeta function. From (64) the low temperature magnetization can be obtained immediately

$$\frac{M(T)}{M(0)} \cong 1 - 2P = 1 - \frac{2V}{N} \left( \frac{k_B}{4\pi\sigma D} \right)^{3/2} \xi(3/2) T^{3/2} \quad (65)$$

which is known as Bloch's Law here for the particular case of spin 1/2. Since, as showed in the last section  $\Gamma(k) \propto k^5$  we expect Bloch's Law to be valid for the amorphous ferromagnet as indeed is found experimentally. The result (65) may be generalized for higher spin if we rewrite (62) as  $\langle S^2 \rangle = S - (\langle S^- S^+ \rangle_{AV} / 2S)$ . This is equivalent to a transformation of the spin operators to the Holstein-Primakoff representation. We then have

$$\frac{M(T)}{M(0)} = 1 - CT^{3/2} \quad (66)$$

where

$$C = 0.0587 [g\mu_B / M(0)] (k_B / D_{SW})^{3/2} \quad (67)$$

The magnetization  $M(0)$  is in  $\text{emu}/\text{A}^3$  and  $g$  is the  $g$  factor,  $\mu_B$  the Bohr magneton in  $\text{emu}$ ,  $k_B$  Boltzmann constant and  $D_{SW}$  the spin wave stiffness.

Before commenting on the validity of (67) for the amorphous ferromagnet we should remark that if one takes the Gaussian spectral density (64) in expression (59) for the magnetization, a  $T^{3/2}$  law with the same  $C$  coefficient is obtained, assuming  $\Delta \propto k^n$ , as long as

$n > 2$ . As we saw in the last section  $\Delta \propto k$  in the amorphous and the experimental observation of the  $T^{3/2}$  law confirms our conclusion that the Gaussian spectral density does not describe correctly the behaviour of the magnetic glass in the long wavelength limit. We notice however that as long as the damping factor has the correct  $k$ -dependence it does not affect in both cases of (60) and (61) the coefficient of the  $T^{3/2}$  term. This fact seems to rule out the possibility of overdamped spin waves making a contribution to the low temperature  $T^{3/2}$  term as suggested by Coey [28].

Formula (67) provides a relationship between the coefficient of the  $T^{3/2}$  term in the magnetization and the spin wave stiffness. Since the latter can be measured directly by inelastic neutron scattering we may compare the value of  $D_{SW}$  obtained by these different methods. In general for crystalline ferromagnets the agreement is rather good [12]. In amorphous ferromagnets however there is a considerable discrepancy between these values. A typical example is the case of CoP where Mook, et al. [7] using inelastic neutron scattering measured a spin wave stiffness  $D_{SW} = 185 \text{ meV } \text{\AA}^2$  while the low temperature magnetization measurements by Cochrane and Cargill [9] yield  $D_{SW} = 120 \text{ meV } \text{\AA}^2$ . Generally in magnetic glasses the values of  $D_{SW}$  obtained through the magnetization fall consistently below those deduced from the former experiments. This seems to indicate the existence of other low lying magnetic excitations in these glasses

Recently [24] [29] we have shown that longitudinal excitations provide an additional  $T^{3/2}$  term to the decrease of the magnetization of amorphous ferromagnets at low temperatures. This offers an explanation for the discrepancy of the values of the spin wave stiffness as measured by different experiments.

#### Acknowledgements

We would like to thank Prof. R. J. Elliott for useful comments and suggestions. One of us (MAC) acknowledges the CNPq of Brazil for financial support.

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