Rapid Magnon Relaxation

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Abstract. The standard procedure for calculating life-times of magnons is to construct equations of motion for magnon occupancy numbers and to solve them for a state of a weak departure from equilibrium. In the last decade a number of experiments used pulse techniques to study dynamics of magnetization after very short pulses bringing magnetic systems out of equilibrium. Therefore a relevant question is how magnons relax after a short strong pulse of magnetic field. The three- and four-magnon relaxation processes are discussed. It is shown that the inverse relaxation time in a short interval after a strong perturbation is enhanced by a temperature independent contribution, depending on the strength of the perturbation.

1. Introduction

Magnon relaxation in ferromagnets was studied intensively some 50 years ago in connection with investigations of the ferromagnetic resonance and related phenomena of absorption of microwaves. The reason of returning to the problem of magnon relaxation is the recent development of new experimental techniques which allow studying magnon relaxation more directly than in the old resonance experiments. The new methods [1-10] are based upon so called pulse-probe techniques in which a short laser pulse disturbs the system studied. After a well defined and very short delay time another short laser pulse ("probe") examines the momentary state of the system. Thus by changing the delay time it is possible to study the time evolution of the system from a disturbance to equilibrium. The technique is very powerful, and can be applied to various phenomena, notably magnetic relaxation. The technique is steadily improved giving the possibilities of shortening the durations of the pulses and the delay times thus enabling to study faster and faster processes. As an example of the progress an experiment [11] can be mentioned in which direct visualization of electron movement in an atom (of krypton, in this case) was possible operating in the time scale of the order of 100 attoseconds (1 attosec = 10^{-18} sec).

The magnon relaxation theory started by the paper [12] was excellently summarized in the article [13] and the book [14]. The theory is based upon the Boltzmann type equations describing time evolution of magnon numbers of a particular wave vector. Assuming small deviations from equilibrium the Boltzmann equations are linearized with respect to these deviations, easily solved giving exponential decay in time of the deviations, governed by the relaxation time. This simple theoretical scheme is complicated by the fact that there are numerous scattering mechanisms, e.g. involving three, four or even more magnons, interaction of magnons with conduction electrons, lattice phonons and structural or magnetic defects of different kinds contributing to a total relaxation rate (i.e. the sum of the inverse relaxation times of processes involved).

Only a few out of many experiments over the past years were successful in reliably confirming predictions of the theory for some processes (see e.g. [14]). These best experiments were done on a single material, the yttrium-iron garnet (YIG), which is a localized spin, non-conducting ferrimagnet
and can crystallize with negligible amount of defects. By clever experiments particular relaxation processes were found dominant making possible verification of the theory (see [14] for more information).

2. Magnon interaction

For the range of magnon wave vectors accessible to experiments based on excitations of magnon modes by high frequency magnetic fields the important ingredient is the energy of magnetic dipolar interactions of electrons responsible for ferromagnetism. Although the magnetic forces are by far weaker than the exchange interactions of an electrostatic origin, because of their long range they strongly affect energy and interactions of magnons in the experimentally important range of magnon wave vectors small as compared with the inverse lattice constant.

For the description of magnons we shall use the simple localized spin models although the results are in fact model independent, since also for itinerant electron magnetism the spin wave theory can be cast into the traditional formalism [15]. The system considered here is defined by the spin hamiltonian

\[
H = -J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j + \sum_{i,j} \frac{(2\mu_B)^2}{|\vec{R}_i - \vec{R}_j|^3} \left[ \vec{S}_i \cdot (\vec{R}_j - \vec{R}_j) \right] \left[ \vec{S}_j \cdot (\vec{R}_i - \vec{R}_j) \right] - 2\mu_B H \sum_j S_j^z (1)
\]

in the standard notation (e.g. [13]). As usually the spin operators are approximately expressed in terms of the harmonic oscillator operators [16, 17] describing flips of spin moments and those are expanded into their Fourier components. Formally \( S_j^x + iS_j^y = N^{-1/2} \sum_k e^{ik \cdot \vec{R}_j} a_k^+ + \cdots \), where \( k \) are the magnon wave vectors and \( n_k = a_k^+ a_k \) is the (operator of) magnon occupations, attaining 0, 1, 2, etc. as its eigenvalues.

In the representation of boson operators the hamiltonian (1) takes the familiar form

\[
H = \sum_k \epsilon_k n_k^a + \sum_{kq} (W_{kq} \ a_k^+ a_q^+ a_{k+q} + h.c.) + \sum_{kq} (\Omega_{k,q,q'} a_k^+ a_q^+ a_{k+q+q'} + h.c.) + \sum_{kq} (T_{kqq'} a_k^+ a_q^+ a_{k+q}^+ a_{k+q+q'} + h.c.)
\]

Due to the presence of the dipolar interactions in order to diagonalising the harmonic part of the hamiltonian (1) expressed in terms of the Fourier components of the spin flip operators the standard canonical transformation [16] was used to get (2). The magnon energy \( \epsilon_k \) in (2) for long wavelength magnons (e.g. for wave vector \( k \) much smaller than the inverse lattice constant but also much larger than the inverse dimension of the ferromagnetic crystal considered) is

\[
\epsilon_k = \left\{ (2\mu_B H + Dk^2)(2\mu_B H + 8\pi\mu_B M \sin^2 \vartheta_k + Dk^2) \right\}^{1/2}
\]

With small numerical errors, not important in the present problem, the magnon energy \( \epsilon_k \) can be approximated by a simpler expression

\[
\epsilon_k \approx 2\mu_B H + 4\pi\mu_B M \sin^2 \vartheta_k + Dk^2
\]

where \( H = H_z - 4\pi N_a M \), \( \sin^2 \vartheta_k = (k_x^2 + k_y^2)/k^2 \). Here z-axis is taken along the applied magnetic field, \( N_a \) is the demagnetizing factor for that direction.

The procedure leading to eq. (2) is well known (an excellent reference is [13]), so the description here is very brief. But now a comment is needed: the transformation diagonalising the dipolar terms leads to the interaction terms (of higher than the second in \( a \)-s) in eq. (2) containing coefficients \( W \ldots, T \ldots \) renormalized by the so-called ellipticity factors. If \( H \gg M \) the resulting corrections are small...
and usually are neglected in discussing magnon relaxation (for the leading relaxation processes they were shown [18] to produce errors of a few percent).

3. Time evolution of average magnon occupation numbers

The basic variable for describing magnon relaxation is the time dependent average magnon occupation number \( n_k(t) = \langle a_k^\dagger a_k \rangle \) averaged over a non equilibrium statistical ensemble. The time evolution of \( n_k(t) \) is calculated from a balance of scattering processes. The probability \( P_{i\rightarrow f} \) of transition in a unit time from an initial state \( |\Psi_i\rangle \) to a final one \( |\Psi_f\rangle \) is calculated from the well-known Fermi golden rule, \( P_{i\rightarrow f} = \frac{2\pi}{\hbar} |\langle \Psi_i | H_f | \Psi_f \rangle|^2 \delta(E_i - E_f) \) where \( H_f \) is the interaction energy causing the transition, and \( E_i, E_f \) are the energies of the initial and final states, respectively.

For clarity we shall discuss separately relaxation due to various scattering processes allowed by the interaction part of the hamiltonian (2), the 3-magnon and 4-magnon ones. In the standard linearized theory the inverse relaxation times for these various processes are additive (see e.g. [13]), the same holds true for the initial rapid relaxation discussed in the present lecture.

The lowest order are the 3-magnon processes, which can be of two types: the so called magnon confluence processes and magnon splitting ones [13]. In the confluence processes a given magnon, say \( k \) is annihilated together with another magnon \( q \) to creating a magnon of wave vector \( (k + q) \). This is the most effective process for long-wavelengths magnons \( k \) since it is easy to conserve energy in the scattering process for experimentally accessible conditions. For the splitting processes in which a given magnon \( k \) splits into two other, say \( q \) and \( -(k + q) \) the energy conservation can be fulfilled only for large enough \( |k| \).

The number of transitions \( (k) + (q) \rightarrow (k + q) \) in a unit time, determined by the second term in (2), is (cf. [13])

\[
P_{k,q} = W_{kq} n_k n_q (n_{k+q} + 1) \tag{4}
\]

where

\[
W_{kq} = \frac{1}{\tau} N^{-1} |f_k + f_q|^2 \delta(\epsilon_k + \epsilon_q - \epsilon_{k+q}) \tag{5}
\]

and

\[
f_k = k_z (k + ik_y) / k^2 . \tag{6}
\]

For convenience the magnon energy in (5) in dimensionless, measured in units of \( k_B (\times 1^\circ K) \), and \( \tau \equiv (\mu_B M)^2 / (\hbar k_B) \) \( \tau \approx 1.7 \times 10^{-12} \) sec for \( M = 140 \) gauss, as for the yttrium-iron garnet at room temperature), and wave vectors are is measured in units of the inverse lattice constant.

The transition rate for reverse process is \( W_{kq} (n_k + 1)(n_q + 1)n_{k+q} \). The resulting total change of \( n_k(t) \) is thus

\[
\frac{dn_k}{dt} = - \sum_q W_{kq} n_k n_q (n_{k+q} + 1) + \sum_q W_{kq} (n_k + 1)(n_q + 1)n_{k+q} . \tag{7}
\]

In the standard theory this Boltzmann type equation is solved under the assumption of small deviations \( \zeta_k(t) \) of \( n_k(t) \) from their thermal averages \( n_k^0 = \left(e^{\epsilon_k/k_BT} - 1 \right)^{-1} \),

\[
n_k(t) = n_k^0 + \zeta_k(t) ; |\zeta_k(t)| \ll n_k^0 .
\]
The equation (7) linearized with respect to $\zeta_k(t)$ takes the form
\[
\frac{d\zeta_k(t)}{dt} \equiv - \frac{1}{\tau_k}\zeta_k(t) + \cdots
\] (8)
which has the obvious solution $\zeta_k(t) = \zeta_k(0)e^{-t/\tau_k}$. Here
\[
1/\tau_k = \sum_q W_{kq}(n^0_q - n^0_{k+q})
\] (9)
defines the relaxation time $\tau_k$ for 3-magnon confluence processes.

The system of equations (7) will be now solved for a strong pulse-like disturbance from equilibrium. The equations (7) can be cast into the form
\[
\frac{d\zeta_k(t)}{dt} = -\frac{1}{\tau_k} + \Psi_k(t) + \Psi_{1k}(t)
\] (10)
where
\[
\Psi_k(t) = \sum_q (W_{kq} - W_{k,q-k})\zeta_q
\] (11)
and
\[
\Psi_{1k}(t) = -n^0_k\Psi_k(t) + \sum_q W_{kq}\left[(n^0_q + 1)\zeta_{q+k} + n^0_{q+k}\zeta_q\right] + \sum_q W_{kq}\zeta_{q+k}\zeta_{q+k}.
\] (12)
The system of equations (10) is formally equivalent to the following
\[
\zeta_k(t) = \{\zeta_k(0) + \alpha_k(t)\}e^{-t/\tau_k-\eta_k(t)}
\] (13)
where
\[
\eta_k(t) = \int_0^t du\Psi_k(u) = \sum_q (W_{kq} - W_{k,q-k})\int_0^t du\zeta_q(u)
\] (14)
and
\[
\alpha_k(t) = \int_0^t du\Psi_{1k}(u)e^{u/\tau_k+\eta_k(u)}.
\] (15)

Now suppose one can generate a pulse-like excitation of magnons peaked at a particular wave vector, say $Q$. Thus we assume $\zeta_k(0) = f\lambda_{kQ}$ where the distribution $\lambda_{kQ}$ is sharp, e.g. $\lambda_{kQ} = e^{-(kQ)^2/L^2}$. An asymptotically exact result can be obtained assuming $\zeta_k(0) = f\delta_{kQ}$ and $f$ very large. Then, for $k = Q$ from (13), neglecting $\alpha_Q(t)$ as compared with $f \gg 1$ it follows
\[
\zeta_Q(t) \equiv f e^{-t/\tau_Q-\eta_Q(t)}.
\] (16)
From (14) we get the self-consistency condition, retaining in (14) only one leading term
\[
\eta_Q(t) = W_{Q0}f\int_0^t du e^{-u/\tau_Q-\eta_Q(u)}
\] (17)
(since, as can be seen from (5), $W_{Q,q} = 0$ for any $Q$).

The equation (17) can be solved to give (for $f \gg 1$):
\[
\eta_Q(t) = \ln]\left[1 + \tau_Q/W_{Q0}(1 - e^{-t/\tau_Q})\right]\n\] (18)
At the initial evolution of the system, in the limit $t \ll \tau_Q$, $\eta_Q(t) \approx fW_{Q0}t$ whereas finally, for $t \gg \tau_Q$,
\[
\eta_Q(t) \approx \ln(1 + fW_{Q0})\text{ i.e. tends to the constant. Therefore, from (13) we get}
\]
\[
\zeta_Q(t) \approx f e^{-(1/\tau_Q+fW_{Q0})t} \text{ for } t \ll \tau_Q
\] (19a)
The result (19a) tells us that in the initial stage of relaxation, for $t \ll \tau_Q$, where $\tau_Q$ is the standard relaxation time obtained from linearized Boltzmann equation, the effective relaxation rate $1/\tau'_{Q}$ is

$$1/\tau'_{Q} = 1/\tau_Q + fW_{QQ} \tag{20}$$

This is an universal result holding also for higher order processes, as we shall show later on. The important observation is that the initial (i.e. for $t \ll \tau_Q$) relaxation rate is enhanced by a contribution $fW_{QQ}$ which is directly proportional to the strength of the disturbance and is essentially temperature independent (apart from an indirect temperature dependence due to the magnetization $M$ entering into the expression for $W_{QQ}$).

For $t \geq \tau_Q$ the standard relaxation time $\tau_Q$ determines the decay, eq. (19b).

Even as we consider very large initially excited number of magnons $f$, $f \gg 1$, the value of the correction $fW_{QQ}$ in eq. (20) can be detectable under narrowly defined experimental condition. The explicit expression for $W_{QQ}$ is

$$W_{QQ} = \frac{1}{\tau} N^{-1} 4 \cos^2 \theta_Q \sin^2 \theta_Q \cdot \delta(2 \varepsilon_Q - \varepsilon_{2Q}) \tag{21}$$

where $\cos \theta_Q = Q_z / Q$, $\omega_0 = 2\mu_B(\mathcal{H} - 4\pi M)/k_B$, $a = 4\pi\mu_B M / k_B$ and

$$2\varepsilon_Q - \varepsilon_{2Q} = \left[ \omega_0(1 - a \omega_0 \cos^2 \theta_Q) \right] - 2DQ^2.$$

The crucial quantity in (21) is $fN^{-1}$ which is equal to the relative change in magnetization $\Delta M / M$ due to initially exciting $f$ magnons of a given wave vectors $Q = Q(\sin \theta_Q, 0, \cos \theta_Q)$, $fN^{-1} = \Delta M / M$. It is not an easy task to make such an experiment. Although it is experimentally feasible to excite well defined magnons in geometrically constrained ferromagnetic samples (e.g. in thin films), attaining large value of $\Delta M / M$ is difficult. In experiments rather a range of magnon wave vectors from an interval $(Q - \Delta, Q + \Delta)$ can be excited instead of a single well defined value $Q$. Thus a relevant quantity in place of $W_{QQ}$ is the average $\bar{W}_{QQ}$

$$\bar{W}_{QQ} = \frac{1}{2\Delta} \int_{Q-\Delta}^{Q+\Delta} dQ' W_{Q'Q'};$$

explicitly

$$f\bar{W}_{QQ} = \frac{1}{2\Delta D} \left[ \frac{\omega_0}{2}(1 - a \omega_0 \cos^2 \theta_Q) \right] \cdot \frac{1}{\tau fN^{-1}}. \tag{22}$$

An alternative procedure is to average $W_{QQ}$ over directions $Q$ with the result:

$$f\langle W_{QQ} \rangle = fN^{-1} \frac{1}{\tau} \frac{1}{2} \frac{D(aQ^2 - \omega_0)}{a(1 + \omega_0/a - D\frac{a}{Q^2})^{1/2}} \tag{23}$$

for $Q$ satisfying the condition $\omega_0/a < \frac{D}{a} Q^2 < 1 + \frac{\omega_0}{a}$.
To estimate numerical values of the averaged $fW_{QQ}$ as given by eq. (22) and (23) we take the material parameters for YIG and assume a reasonable value for magnetic field, giving $\alpha_0 \approx 2a$, and $Q = 3.5 \times 10^{-2}$ (within the limits imposed by eq. (23)), and $\Delta/Q \approx 10^{-3}$. For such choice of parameters the average of $fW_{QQ}$ from the both approaches, (22) and (23) is of the order of magnitude $\langle fW \rangle \approx 3 \times 10^{11} (f/N)/\text{sec}$. The estimate of the standard, i.e. calculated from linearized Boltzmann equation [14], relaxation rate $1/\tau_Q$ in eq. (20) for the same set of parameters for YIG is roughly equal $1/\tau_Q \approx 4 \times 10^6 (T/300^\circ K) /\text{sec}$. Therefore, even for not too large relative number $(f/N)$ of excited magnons $Q$, say already for $(f/N) \geq 10^{-5}$, both contributions, $1/\tau_Q$ and $\langle fW_{QQ} \rangle$ to the initial relaxation rate $1/\tau^f_Q$ are of importance. The linear dependence of $1/\tau^f_Q$ on temperature and very weak temperature dependence of $fW_{QQ}$ can also be helpful in a possible verification of the present predictions by an experiment.

4. A two-mode model
The analysis so far is incomplete since first, it still lacks an estimate of the function $\alpha_Q(t)$ in eq. (13) for $k = Q$ to confirm that in fact $\alpha_Q(t)$ is much smaller that $f$ and, second, the time evolution of modes $k \neq Q$ is not yet determined. Although the equations (10), (11) and (12) provide a complete set to determine $\zeta_k(t)$, $\alpha_k(t)$ and $\eta_k(t)$ a direct solution of the set is obviously out of question. For a qualitative analysis we shall consider a two-mode model with only two sets of magnons, those externally excited of the vectors $Q$ and magnons $k$, arbitrarily picked from the magnon manifold. We shall consider only short time intervals $t \ll \tau_k$ after the excitation. To simplify the otherwise lengthy formulae we shall assume low temperature so the thermal magnon occupations $n_k^0 = (e^{\epsilon_k/k_BT} - 1)^{-1} \approx e^{-\epsilon_k/k_BT} \ll 1$ are very low and $n_k^0$ can be dropped out from calculations.

It can be shown that for $t \ll \tau_k$

$$\eta_k(t) \approx f(W_{k,Q} - W_{k,k})t + O(t^2)$$

(24)

whereas $\eta_k(t)$ for large $t$ approaches a constant, non-zero value. So properties of $\eta_k(t)$ are similar to those for $k = Q$. Also it follows that $\alpha_Q(t) \approx O(t^2)$ for small $t$ and for $k \neq Q$

$$\alpha_k(t) \approx fW_{k,Q-k}t + O(t^2)$$

(25)

Since $\alpha_Q(t) - O(t^2)$ so indeed $\alpha_Q(t)$ is negligible in comparison with $f$ during the initial stage, $t \ll \tau_k$, of relaxation and the validity of eq. (16) is thus confirmed. For $k \neq Q$ the initial relaxation is given by

$$\zeta_k(t) \approx (fW_{k,Q-k})e^{-t/\tau_k^f}$$

(26)

where the enhanced effective relaxation rate is given by

$$1/\tau_k^f = 1/\tau_k + f(W_{k,Q} - W_{k,k}).$$

(27)

5. Other mechanisms of rapid relaxation
The rapid relaxation by 3-magnon confluence processes of magnetic dipolar origin was discussed in details since they are the dominant ones in prevailing experimental conditions. For the 3-magnon splitting processes the magnon energy conservation conditions restrict the possible enhancement of the
relaxation rates to values of the magnetic field and magnon wave vectors and a result similar to eq. 
(19) is difficult to prove with any rigor.

Rapid relaxation can proceed through two types of 4-magnon processes two in-two out (“2→2”) 
scattering processes of both exchange and magnetic dipolar origin and by three in-one out (“3→1”) 4-
magnon confluence processes of magnetic dipolar origin.

For the higher order processes qualitatively the result is analogous to eq. (20), namely the effective 
relaxation rate consists of the sum of inverses of the standard, Boltzmann type, relaxation times for all 
processes possible (2→1; 2→2; 3→1) and a term which is temperature independent and increases with 
the strengths f of the excitation. The only difference is that for some values of the excited magnons 
wave vector Q besides the linear term in f there appears also a term of the order f^2 due to 3→1 
confluence processes [19].

6. Discussion
The main result of the theory presented here is the following: if we excite a large number f of magnons 
of a particular wave vector Q at a given time, say t=0, then the initial relaxation (i.e. for the time 
interval t ≪ τ_k ) is determined by an effective relaxation time τ_Q ef , eq. (20), which is shorter than the 
relaxation time τ_Q for small departures from equilibrium, and the effective relaxation rate 1/τ_Q ef is 
enhanced by the contribution which is (essentially) temperature independent and proportional to 
excitation strength f. This result was discussed in details for the usually dominant scattering 
mechanisms which are the 3-magnon confluence processes of dipolar origin. Qualitatively the same 
behaviour can be shown for higher order magnon processes. The physical interpretation of eq. (20) is 
clear. At low temperatures the equilibrium magnons occupations n_Q 0 are small, n_k 0 ≪ 1, so only a small 
number of magnons is present in the system. If at t = 0 we excite a large number n_Q (0) ≅ ζ_Q (0) = f 
of magnons Q the dominant scattering processes will consist in combining two magnons Q into a third 
one having the wave vector (2Q). These processes have the relaxation rate f W_Q QQ , essentially 
independent on the presence of a small number of thermal magnons so it is temperature independent 
(apart for a slight temperature dependence of M entering into W_QQQ ). The processes are allowed, i.e. 
W_oo ≠ 0, for a narrow but experimentally accessible interval of Q values.

The decaying of the primary magnons Q is also going through their interaction with the thermal 
magnons at the rate 1/τ_Q in eq. (20). After a short time interval t ≪ τ_Q , τ_k the relaxation of the 
primary magnons Q as well as the secondary ones k is continued by interactions with thermal 
magnons and the decay rates are determined by the standard relaxation times τ_Q , τ_k calculated for 
small deviations from equilibrium, known in literature (see e.g. [13], [14]).

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