Phonon Bandwidth and Rate Equations in Avalanche Relaxation

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The bandwidth of phonons emitted in avalanche spin-phonon relaxation processes is studied when the phonon interruption time is shorter than spin-spin relaxation time. The problem has been solved by the use of second-order Heisenberg equations of motion for phonon-number operators of each lattice mode. It is shown that the usual rate equations, based on the assumption of time-independent bandwidth, are not adequate in these circumstances and that the bandwidth of phonons emitted in the relaxation process decreases during the avalanche. Rough numerical estimates in a typical case suggest that the bandwidth of the emitted phonons, at times at which phonon generation may be considered as practically concluded, may still be larger than the spin-resonance linewidth. Finally, it is emphasized that the results obtained are confirmed by considering the relaxation process in the light of the energy-time uncertainty principle.

In this paper we shall be concerned with paramagnetic relaxation via direct processes in systems which have been artfully displaced from thermal equilibrium. Intuitively speaking it is rather obvious that the spin system in a direct relaxation process should prefer to give more of its energy to lattice modes whose frequencies are near to the Larmor frequency than to others. [1] What is not obvious is how to predict the bandwidth of the emitted phonons under different relaxation conditions, and, indeed, whether the shape of their power spectrum may be considered constant as a function of time. Both problems become relevant in the phonon-bottleneck regime, i.e., when more phonons per unit time are created by the decaying spins than could be dissipated, and particularly in crystals where the phonon interruption time (namely, the average time an emitted phonon lives before being absorbed by some other spin) is shorter than the spin-spin relaxation time.

The first of these problems has been considered by Giordmaine *et al.*, [2] in a very careful analysis of the relaxation of a spin system whose temperature is positive and higher than the rest of the crystal under the conditions mentioned above. Such a situation could hardly be described by models where each spin would relax independently of the others. The above-mentioned authors suggest that in this case the bandwidth of the emitted phonons should be entirely governed by repeated spin-phonon interactions; in a subsequent paper Giordmaine and Nash[3] point out that under these conditions spins and phonons should be considered as a single entity, giving rise to spin-phonon modes extending throughout the crystal. By means of arguments similar to those used by Jacobsen and Stevens[4] to describe propagation properties, they show that the spectral width of the lattice-mode excitations may exceed the spin-resonance linewidth.

The aim of this paper is to investigate the second problem in an avalanche process: We shall study, for interruption times shorter than the spin-spin relaxation time, the frequency distribution of phonons emitted when the initial spin population is inverted. This case is very different from that considered above, since in the presence of a self-regenerative relaxation process it is reasonable to expect a variation of the bandwidth of the emitted phonons during the development of the avalanche.

Separate equations for each lattice-mode population have been found in a previous paper[5] on the basis of a simplified model which should preserve the essential features of the physical system. The model consists of spins and phonons interacting linearly. The spin-spin interaction is neglected and the phonon relaxation time to the external bath is assumed to be much longer than any characteristic time involved. The Hamiltonian is assumed to be of the form[6]

$$\mathcal{H}=\sum_k \omega_k\, \alpha_k^\dagger \alpha_k+\frac{\omega_0}{2}\sum_r \sigma_z^r+\sum_{k,r}\epsilon\,(\alpha_k\sigma_+^r\, e^{ikr}+\text{H. c.}), \quad (1)$$

where the k dependence of the spin-phonon coupling constant has been dropped for simplicity. We have shown that, with a reasonable assumption, the spin-phonon population in the kth mode obeys the equation

$$\frac{d^2 n_k}{dt^2}=(\eta^2\sigma_z-\Delta\omega_k^2)\,n_k+\eta^2\,\frac{1+\sigma_z}{2}\ , \quad (2)$$

where σ_z is the difference of the spin-level population normalized to unity, $\eta^2=2N\epsilon^2$, N is the number of spins in the crystal, and

$$\Delta\omega_k^2=(\omega_k-\omega_0)^2\ .$$

It may be worthwhile to emphasize at this point that the rate equations like those introduced for the first time by Faughnan and Strandberg[7] to describe relaxation from positive temperatures in bottleneck conditions, and subsequently used by Brya and Wagner[8] to describe an avalanche in Ce-La Magn nitrate are not suitable for our purpose. Those equations in fact can be derived from (2) if a *time-independent* phonon bandwidth is *a priori* introduced. Such an assumption is not allowed here, as we shall now show. Straightforward integration of (2), assuming as initial conditions

$$n_k(t=0)=\frac{dn_k}{dt}\bigg|_{t=0}=0\ ,$$

gives

$$n_k(t)=\frac{\eta^2}{2}\int_0^t [1+\sigma_z(t')]\,\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}dt' +\eta^2\int_0^t \sigma_z(t')n_k(t')\,\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}dt'\ . \quad (3)$$

Moreover, since the total number of excitations commutes with the Hamiltonian, we should have

$$\sigma_z+\frac{2}{N}\sum_k n_k(t)=\text{const.} \quad (4)$$

From (3) and (4) we get

$$\begin{aligned}\sigma_z(t)&=\text{const}-\frac{2}{N}\sum_k n_k(t)\\ &\simeq \text{const}-\frac{2}{N}\int_0^{\omega_D} g(\omega_k)n_k(t)\,d\omega_k\\ &=\text{const}-\frac{\eta^2}{N}\int_0^t dt'\,[1+\sigma_z(t')]\int_0^{\omega_D} g(\omega_k)\\ &\quad\times\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}\,d\omega_k-\frac{2\eta^2}{N}\int_0^t dt'\sigma_z(t')\\ &\quad\times\int_0^{\omega_D} g(\omega_k)\,n_k(t')\,\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}d\omega_k\ , \end{aligned} \quad (5)$$

where $g(\omega_k)$ is the density of phonon states at ω_k and ω_D is the Debye temperature of the crystal. In order to obtain the rate equation for the difference of spin-level population, it is essential to take $n_k(t')$ in (5) out of the integration in ω_k; for this we must assume that $n_k(t')$ is constant as a function of ω_k in a band large enough to make

$$g(\omega_k)\,\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}$$

small at the two ends of the band. This condition is easily satisfied in most actual cases, and with this assumption we have

$$\begin{aligned}\int_0^{\omega_D} n_k(t')g(\omega_k)\,\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}\,d\omega_k &= n(\omega_0,t')\int_0^{\omega_D} g(\omega_k)\,\frac{\sin\Delta\omega_k(t-t')}{\Delta\omega_k}\,d\omega_k\\ &\simeq n(\omega_0,t')g(\omega_0)\,\pi\ .\end{aligned} \quad (6)$$

Substituting in (5) we have

$$\sigma_z=\text{const}-\frac{\eta^2}{N}g(\omega_0)\pi\int_0^t \{[1+\sigma_z(t')] +2n(\omega_0,t')\,\sigma_z(t')\}dt'\ , \quad (7)$$

and, differentiating with respect to t and multiplying through by N,

$$\frac{d(n_2-n_1)}{dt}=-\,2A\,\{[n(\omega_0,t)+1] \times n_2-n\,(\omega_0,t)n_1\}\ , \quad (8)$$

where n_1 and n_2 are the populations of the two spin levels and

$$A=2\pi\epsilon^2 g(\omega_0)\ .$$

Equation (8) is the first of the rate equations we are looking for, provided that $n(\omega_0,t)$ is identified with the average population of the modes in speaking terms. Moreover, differentiating (4) we get

$$\frac{d\sigma_z}{dt}=-\frac{2}{N}\int_0^{\omega_D} g(\omega_k)\frac{dn_k}{dt}d\omega_k\ , \quad (9)$$

and, if we wish to obtain from (9) the rate equation for the phonon population, it becomes essential to assume that dn_k/dt is nearly constant within a band $\langle\Delta\omega\rangle$ of modes and negligible outside. With this assumption we find

$$\frac{d\sigma_z}{dt}=-\frac{2}{N}\,\frac{dn(\omega_0,t)}{dt}\int_{\langle\Delta\omega\rangle} g(\omega_k)\,d\omega_k\ , \quad (10)$$

so that

$$\frac{dn(\omega_0,t)}{dt}=-\,\frac{1}{2\langle\Delta\omega\rangle g(\omega_0)}\,\frac{d(n_2-n_1)}{dt}\ . \quad (11)$$

Equation (11) is equivalent to Eq. (2) of Ref. 7, provided that $\langle\Delta\omega\rangle$ does not vary with time, and it is the second of the rate equations which we have derived under the assumption of a time-independent phonon bandwidth. This assumption is, however, in contrast with (2) in avalanche conditions. In fact, although exact solutions of (2) will not be given here, it may be guessed that (as long as $\sigma_z>0$ and for not too large times) each mode distant in frequency from resonance by a quantity $\Delta\omega_k<\eta\,\sigma_z^{1/2}$ should increase its population approximately proportional to $\exp\{[\eta^2\sigma_z-\Delta\omega_k^2]^{1/2}t\}$, leading to a relative decrease of the population of this mode with respect to that of the resonant $\Delta\omega_k=0$ mode, which is far from being constant as a function of t. Moreover, due to the decrease of σ_z during the avalanche, phonon modes should progressively go out of the frequency region of quasiexponential increase when their $\Delta\omega_k$ become larger than $\eta\sigma_z^{1/2}$, thereby magnifying the effect of relative decrease of population. We are therefore led to the conclusion that at least the rate equation (11) with constant $\langle\Delta\omega\rangle$ cannot constitute an adequate description of the avalanche in our model.

We may now go back to the main purpose of this paper: to estimate approximately the distribution of emitted phonons as a function of the frequency. According to our previous considerations, we assume as approximate solutions of (2), for modes with $\Delta\omega_k < \eta\sigma_z^{0\,1/2}$ and for not too long times, the expressions [see also Ref. 5, Eq. (6)]

$$n(x,t) = \frac{1+\sigma_z^0}{2\sigma_z^0}\,\frac{1}{1-x^2}\,[\cosh(1-x^2)^{1/2}\tau - 1], \quad (12)$$

where

$$\sigma_z^0 \equiv \sigma_z(t=0), \quad x^2 = \Delta\omega_k^2/\eta^2\sigma_z^0, \quad \tau = \eta\sigma_z^{0\,1/2}t\ .$$

Solution (12) would be exact if $\sigma_z(\tau) = \sigma_z^0$; the most important effects on the solutions of (2) due to the decrease of σ_z with time will be considered later. We define the width of the band of emitted phonons by the relation

$$n(\bar{x},\tau) = (1/e)[n(x=0,\tau)]\ , \quad (13)$$

which becomes, using (12),

$$(1-\bar{x}^2)\,\frac{\cosh\tau - 1}{\cosh(1-\bar{x}^2)^{1/2}\tau - 1} = e\,. \quad (14)$$

In the region of interest ($\tau > 1$, $\bar{x} < 1$) this equation has a root in the neighborhood of $\bar{x}^2 \sim 2/\tau$, from which we find the bandwidth to be

$$\langle\Delta\omega\rangle^2 \simeq 2\eta^2\sigma_z^0/\tau\ . \quad (15)$$

This is time dependent, as expected, and decreases with increasing time. One should remember, however, that in obtaining (15) we have neglected the effect of the time variation of σ_z, which might become important at large times. Although an accurate evaluation of these effects is far from simple, a rough estimate can be made—observing from (2) that for each mode with a given x a time τ_{out} should exist at which the quasiexponential increase of the population of that mode ceases. This time is defined by the relation

$$\frac{\sigma_z(\tau_{out}(x))}{\sigma_z^0} = x^2\ , \quad (16)$$

and it is now evident that solutions (12) for each x are approximately valid only for times $\tau \le \tau_{out}(x)$. For times τ not much longer than $\tau_{out}(x)$ we shall assume

$$n(x,\tau) \simeq n(x,\tau_{out}(x))\quad . \quad (17)$$

Solutions (12) for $\tau < \tau_{out}$ and (17) for $\tau > \tau_{out}$ now permit an evaluation of the time evolution of the bandwidth of emitted phonons $\langle\Delta\omega\rangle$ up to times when the avalanche is practically concluded, or when σ_z has decreased by a non-negligible amount so that (15) is no longer valid. We proceed in the following way. From the results of our previous work[5] we know that σ_z evolves in time quite rapidly during the avalanche from the initial value σ_z^0 to about zero, where it lingers for times which are orders of magnitude larger than those necessary for the first part of the decay. We may then arbitrarily define a time τ_f such that

$$\frac{\sigma_z(\tau_f)}{\sigma_z^0} = 10^{-1}\ ,$$

at which the avalanche may be considered concluded as far as the generation of further phonons is concerned, but σ_z is still quite rapidly decaying towards zero. Using the results of our previous work we find

$$\tau_f \simeq \ln\frac{N}{4g(\omega_0)\eta}\quad . \quad (18)$$

We observe that the band of modes effectively interacting with the spins can be divided into three regions. In the first we put the modes for which $\tau_{out}(x) < \tau_f$. For these modes we can immediately use (12) and (17), and we get

$$\begin{aligned} n(x,\tau_f) &\simeq \frac{1+\sigma_z^0}{2\sigma_z^0}\,e^{(1-x^2/2)\tau_{out}(x)} \\ &= \frac{1+\sigma_z^0}{2\sigma_z^0}\,e^{\tau_f}\,e^{-[\tau_f-\tau_{out}(x)]} \\ &\quad\times e^{-x^2\tau_f/2}\,e^{x^2[\tau_f-\tau_{out}(x)]/2}\ , \end{aligned} \quad (19)$$

where we have neglected $(1-x^2)$ in the denominator, since the exponential x dependence should cut any effect coming from this factor. The second consists of a very narrow band around the Larmor frequency, which at $\tau=\tau_f$ has not yet entered the region of validity of (17). If the fall of σ_z towards zero immediately before is rapid enough, a good approximation for the population of these modes should be

$$n(x\simeq 0,\tau_f) \simeq \frac{1+\sigma_z^0}{2\sigma_z^0}\,e^{\tau_f}\quad . \quad (20)$$

This amounts to putting $\tau_{out}(x\sim 0) = \tau_f$, which is physically reasonable, since the exponential growth of population of these modes after τ_f certainly becomes very feeble, due to the smallness of $\eta^2\sigma_z(\tau_f) - \Delta\omega_k^2$. The third region consists of the modes in between the first two; since we expect a smooth curve for the distribution of emitted phonons, we shall join (19) and (20), and describe approximately the whole curve at $\tau=\tau_f$ by the expression

$$n(x,\tau_f) \simeq n(x=0,\tau_f)\,e^{-x^2/\bar{x}_f^2}\,e^{-[\tau_f-\tau_{out}(x)]}\ , \quad (21)$$

where we have put $\bar{x}_f^2 = 2/\tau_f$. We wish now to have an explicit expression for $\tau_f - \tau_{out}(x)$ in (21) as a function of x. Since $x<1$, we expand

$$\tau_{out}(x) \simeq \tau_{out}(0) + \frac{d\tau_{out}}{dx}\bigg|_{x=0} x + \frac{1}{2}\frac{d^2\tau_{out}(x)}{dx^2}\bigg|_{x=0} x^2 . \qquad (22)$$

According to our previous discussion, we set $\tau_{out}(0) \sim \tau_f$. Moreover, from (16) we have

$$\frac{d\tau_{out}}{dx}\bigg|_{x=0} = \frac{d\tau_{out}}{d\sigma_z}\frac{d\sigma_z}{dx}\bigg|_{x=0} = \frac{d\tau_{out}}{d\sigma_z} 2\sigma_0 x\bigg|_{x=0} = 0 ;$$

$$\frac{d^2\tau_{out}}{dx^2}\bigg|_{x=0} = 2\sigma_0 \frac{d\tau_{out}}{d\sigma_z}\bigg|_{x=0} + 2\sigma_0 x \frac{d}{dx}\left(\frac{d\tau_{out}}{d\sigma_z}\right)\bigg|_{x=0}$$

$$= 2\sigma_0 \frac{d\tau_{out}}{d\sigma_z}\bigg|_{x=0} .$$

We approximately evaluate the last derivative as

$$\frac{d\tau_{out}}{d\sigma_z}\bigg|_{x=0} \simeq \left(\frac{d\sigma_z}{d\tau}\right)^{-1}\bigg|_{\tau=\tau_f} \equiv -\left|\frac{d\sigma_z}{d\tau}\right|^{-1}_{\tau_f} , \qquad (23)$$

since $d\sigma_z/d\tau$ is always negative, and substitute in (21), which becomes

$$n(x, \tau_f) \simeq n(x=0, \tau_f)\exp(-x^2/\overline{x}_f^2) \times \exp\left(-x^2\bigg/\frac{1}{2\sigma_z^0}\left|\frac{d\sigma_z}{d\tau}\right|_{\tau_f}\right) . \qquad (24)$$

This is the sought-for approximate expression for the distribution of emitted phonons at time τ_f. We see that the band is narrower than it would be if we had extrapolated expression (15), which is valid for short times. This extra narrowing is to be attributed to the mechanism that progressively cuts the modes in the wings of the band out of the region of exponential growth. From (24) the extra narrowing is inversely proportional to $|(d\sigma_z/d\tau)|_{\tau_f}$ and tends to be less effective the steeper the decay of σ_z, as in fact could have been anticipated.

A rough numerical estimate of the bandwidth at τ_f in a typical case is possible using numbers given in Ref. 2. For 1% paramagnetic concentration of the Cu salt considered there, and by identifying the quantity η in this paper with the quantity $2\pi\Delta\nu_L$ calculated there [see Ref. 2, Eq. (8)], we obtain

$$\frac{N}{4g(\omega_0)\eta} \simeq 10^4 ,$$

and through (18) we have $\tau_f \simeq 10$. Moreover, a conservative estimate based on our previous work[5] gives $(1/2\sigma_z^0)|(d\sigma_z/d\tau)|_{\tau_f} \simeq 10^{-1}$. Substituting these values in expression (24) we get that the bandwidth of the emitted phonons is

$$\langle\Delta\omega\rangle \simeq 10^{-1}\eta\sigma_z^{0\,1/2} ; \qquad (25)$$

that is, one-tenth of the bandwidth at the beginning of the decay. This is considerable narrowing, but the band may be still larger than the spin-resonance linewidth.

Finally, we would like to comment on our results in light of the indetermination principle $t\Delta\omega \geq 1$, which Eq. (25) satisfies. We note, under the conditions considered here, the avalanche practically ends before that rigorous conservation of energy might be required. In a following paper we shall report results based on exact solutions of (2) which provide strict energy conservation at $\tau \gg \tau_f$, and which support the results of the present paper.

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