Possibility of spin-phonon superradiance in paramagnetic systems*

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The possibility of superradiant spin-phonon relaxation is theoretically investigated in the framework of the Jacobsen-Stevens model for spins coupled to the lattice. The equations derived are similar to those of the optical case, so that under appropriate conditions superradiance should occur as in the optical case.

As suggested by Dicke[1] and recently observed by Skribanowitz *et al.*,[2,3] a large assembly of molecules, initially all in the excited state, can emit all of its energy in a burst of highly directional optical radiation many orders of magnitude shorter than the single-molecule decay time T_1. This effect, called superradiance, occurs because the molecules do not relax independently, but instead radiate in a collective mode of decay. The characteristic radiation rate in this process is enhanced from T_1^{-1} by a factor $n_0 \lambda^2 L/8\pi$ ($\approx 10^9$ in the experiments[2]), where n_0 is the excitation density of molecules, λ the optical wavelength, and L the length of the sample ($L \gg \lambda$). The superradiant burst is preceded by a delay typically an order of magnitude longer than the duration of the burst and, under appropriate conditions, is accompanied by ringing. The basic condition for optical superradiance to occur is that all relaxation times must be long compared to the characteristic superradiant radiation time.

The purpose of this paper is to explore the possibility that under appropriate conditions, superradiant behavior can also occur in the spin-phonon interaction process in paramagnetic crystals. The system would be prepared with all the spins inverted and the lattice unexcited. Then, under appropriate conditions, kT fluctuations of the lattice would trigger the collective de-excitation of the spins, leaving all the spins in the ground state and all the energy in lattice vibrations. As shall be shown, the characteristic rate T_R^{-1} of this process would be enhanced from the spin-lattice relaxation rate T_1^{-1} by the factor $n_0 \lambda^2 L/4\pi$ (which can be $\gg 1$), where now n_0 is the spin density and λ the acoustic wavelength. The condition for spin-phonon superradiance to occur is that all relaxation times for the spin and lattice processes must be long compared to the characteristic superradiant time T_R. Under this condition, such a system should exhibit all the features of optical superradiance.

Reported results of an experiment by Brya and Wagner[4] on paramagnetic relaxation show a behavior which on the surface appears to be similar to that described in the previous paragraph. The time dependence of the spin-level population difference was observed after excitation of the paramagnetic transition to a negative spin temperature. It was found that after a delay much shorter than the normal spin-lattice relaxation time T_1, the spin system quickly released its energy to the resonant lattice modes ("phonon avalanche"). In contrast to the anticipated behavior, however, no ringing was observed, and the rapid decay stopped as soon as the population of the two spin levels equalized.

Since the emission rate in a superradiant process reaches its maximum when the level populations have become equal,[1] the lack of further rapid decay indicates the absence of a coherent transverse component of the paramagnetic spins. Therefore, this experiment does not exhibit superradiance in the sense of the optical superradiance experiments in which, according to the theory,[3] a sizeable coherent dipole moment is created. This statement is supported by the fact that the Brya-Wagner experimental results can be fit[4] to a rate-equation analysis, which omits phase coherence.[5] The lack of coherence in the radiation process occurs because the spin-spin relaxation time T_2 is sufficiently short, compared to the characteristic time T_R, to destroy phase correlations in the motion of the transverse component of the paramagnetic spins. This has been verified by shortening T_2 sufficiently in the theoretical model[3] describing optical superradiance, in which case the computer solution becomes similar to the behavior observed by Brya and Wagner.

We now present some preliminary theoretical results. We use the Jacobsen-Stevens model for spins coupled to the lattice.[9] Although this model assumes a concentrated lattice with adjacent spins, it should be equally suitable for more dilute systems provided that there are many spins per acoustic wavelength. This Jacobsen-Stevens model contains the essential physical features needed to produce the behavior under discussion. The treatment is similar to the one used to explain the experimental results in the optical case.[3]

The Jacobsen-Stevens Hamiltonian for the system is[9]

$$H_{JS} = \sum_j [\tfrac{1}{2}\mathcal{P}_j^2/m + \tfrac{1}{2}K(U_j - U_{j+1})^2 + g\mu_B H S_z^{(j)} + \hbar\epsilon(U_{j+1} - U_{j-1}) S_x^{(j)}], \quad (1)$$

where one atom of mass m and spin $\frac{1}{2}$ is assumed per unit cell. U_j is the displacement operator and $\mathcal{P}_j$ the momentum operator of atom j in the x direction. K is the restoring force between nearest neighbors and H is the dc magnetic field in the z direction. $S_x^{(j)}$ and $S_z^{(j)}$ are the x and z components of the spin operator for the unpaired electron on atom j, normalized such that $[S_x^{(j)}, S_y^{(j)}] = iS_z^{(j)}$, and $\mu_B = e\hbar/2mc$. The coupling constant between the strain at position j and the spin components $S_x^{(j)}$ is ϵ. This coupling is chosen so that a component in $U_{j+1} - U_{j-1}$ at the resonance frequency of the spin is able to induce spin transitions.

In the spirit of the work of Jacobsen and Stevens, we adopt the semiclassical approximation in which the lattice vibrations are treated classically [i.e., the operator U_j becomes a classical quantity $U(j)$] and the spins are treated quantum mechanically. Consider first the quantum-mechanical Hamiltonian, omitting classical terms which contribute only to the lattice motion:

$$H' = \sum_j \{g\mu_B H S_z^{(j)} + \hbar\epsilon[U(j+1) - U(j-1)] S_x^{(j)}\}. \quad (2)$$

Since the quantity in curly brackets depends only on the operators $S_x^{(j)}$ and $S_z^{(j)}$, the total spin-wave function Ψ of the system is separable into a product of single spin-wave functions ψ_j. We then have a single-particle problem with

$$H_0 = g\mu_B H S_z + D\hbar S_x, \quad (3)$$

where $D = \epsilon[U(j+1) - U(j-1)]$. S_x and S_z are quantum-mechanical operators on the spin-wave function $\psi_j(t)$ of the spin-$\frac{1}{2}$ system, which we expand in the form $\psi_j = a(j, t)|\uparrow\rangle + b(j, t)|\downarrow\rangle$. Then Schrödinger's equation, $H_0\psi_j = i\hbar\partial\psi_j/\partial t$, gives[10]

$$\left(\frac{\partial^2}{\partial t^2} + \omega_0^2\right) P = \omega_0 D n, \quad (4)$$

$$\frac{\partial n}{\partial t} = -\left(\frac{D}{\omega_0}\right)\frac{\partial P}{\partial T}, \quad (5)$$

where $\hbar\omega_0 = g\mu_B H$, $n(j, t)$ is the spin-population difference per unit volume,

$$n(j, t) \equiv 2n_0\langle\Psi|S_z^{(j)}|\Psi\rangle = n_0(|a|^2 - |b|^2), \quad (6)$$

and

$$P(j, t) \equiv 2n_0\langle\Psi|S_x^{(j)}|\Psi\rangle = n_0(a^* b + ab^*), \quad (7)$$

where n_0 is the number of spins per unit volume.

In order to obtain a wave equation for $U(j)$, we treat the original Hamiltonian [Eq. (1)] classically, with operators replaced by their expectation values ($S_x^{(j)} \to \langle\Psi|S_x^{(j)}|\Psi\rangle$). Then Eq. (1) becomes

$$H_{JS} = \sum_j \{\tfrac{1}{2}[P(j)]^2/m + \tfrac{1}{2}K[U(j) - U(j+1)]^2 + \tfrac{1}{2}g\mu_B \times Hn(j)/n_0 - \tfrac{1}{2}\hbar\epsilon[P(j+1) - P(j-1)] U_j/n_0\}, \quad (8)$$

and Hamilton's equations give

$$\frac{\partial^2 U(j)}{\partial t^2} = (K/m)[U(j+1) + U(j-1) - 2U(j)] + (\tfrac{1}{2}\hbar\epsilon/mn_0)[P(j+1) - P(j-1)]. \quad (9)$$

Equations (4), (5), and (9) are the coupled semiclassical equations describing the spin-phonon interaction. Equations (4) and (9) are similar to Eqs. (2) and (3) of Jacobsen and Stevens[9] with the operators replaced by average values. Equation (5) has no counterpart in Ref. 9 because there $\langle S_z\rangle$ is assumed to be time-independent, whereas we allow n to vary in both space and time.

We now let $P = \mathrm{Re}(P' e^{i\omega t - ikja})$ and $U = \mathrm{Re}(U' e^{i\omega t - ikja})$ in Eqs. (4), (5), and (9), where a is the spacing between paramagnetic ions. Assuming that the amplitudes P' and U' are slowly varying in space and time, such that $\partial P'/\partial t \ll \omega P'$, $P'(j+1) - P'(j) \ll kaP'(j)$, etc., these equations become

$$\left(\frac{1}{2ika^2}\right)\{[1 - e^{-ika} - \tfrac{1}{2}(ka)^2] U'(j+1) + [1 - e^{ika} - \tfrac{1}{2}(ka)^2] U'(j-1)\} + \left(\frac{1}{v}\right)\frac{\partial U'(j)}{\partial t} = -\left(\frac{\hbar\epsilon k}{2mn_0\omega^2}\right)\sin(ka) P'(j), \quad (10a)$$

$$\left(\frac{\partial}{\partial t} + T_2^{-1} + i(\omega - \omega_0)\right) P'(j) = -\left(\frac{\omega_0\epsilon a U'(j)}{v}\right) n(j), \quad (10b)$$

$$\frac{\partial n(j)}{\partial t} = -\frac{n(j) + n_0}{T_1} + \epsilon\sin(ka)\,\mathrm{Re}[P'(j)^* U'(j)], \quad (10c)$$

where phenomenological damping terms have been added. Here, T_1 is the spin-lattice relaxation time, T_2 is the spin-spin relaxation time, and $v^2 = \omega^2/k^2 = Ka^2/m$. Equations (10) are similar to Eqs. (14)–(16) of Ref. 3,[11] which describe optical superradiance and coherent pulse propagation, and

therefore all of the coherent optical effects should have analogs in paramagnetic systems. In fact, self-induced transparency in a paramagnetic system has recently been observed by Shiren.[12]

The similarity with the coherent optical equations is most easily seen in the case $ka < \frac{1}{2}$ (which is almost always the case). Then U', P', and n can be treated as continuous functions of x, and Eqs. (10) can be rewritten using retarded time coordinates ($t' = t - x/v$, $x' = x$) so that they are identical in form with the optical equations.

In the limit where relaxation effects can be ignored (i.e., the T's are sufficiently[3] long), it is found that the important superradiant features can be obtained in the simple case of exact resonance ($\omega = \omega_0$), assuming that ka is small and that the initial conditions for P' and $U'(j)$ are real. Then Eqs. (10) have the solution

$$n(j, t) = n_0 \cos\phi(j), \tag{11}$$

$$P'(j, t) = -n_0 \sin\phi(j), \tag{12}$$

$$\frac{\partial\phi(j)}{\partial t} = k\epsilon a U'(j), \tag{13}$$

$$\frac{1}{2}\left(\frac{\partial}{\partial t}\right)[\phi(j+1) - \phi(j-1)] + \left(\frac{1}{v}\right)\frac{\partial\phi(j)}{\partial t} = \left(\frac{\hbar k \epsilon^2 a^2}{2mv^2}\right)\sin\phi(j). \tag{14}$$

Since ka is small, $U'(j)$ and $\phi(j)$ become continuous variables, and using retarded time coordinates, Eq. (14) is identical in form to Eq. (43) of Ref. 3,[13]

$$\frac{\partial^2\phi(x', t')}{\partial x' \partial t'} = \frac{\sin\phi}{T_R L}, \tag{15}$$

with

$$T_R = 2mv^2/\hbar k \epsilon^2 a^2 L \tag{16}$$

for the paramagnetic system. Computer solutions of Eq. (15) subject to the initial condition $\phi(x', t' = 0) = \delta$, with $\delta \ll 1$ (which corresponds to the small kT fluctuations[3]), exhibit the superradiant features described previously including delay, rapid radiation, and ringing. The close connection between Eqs. (10) and (15) leads us to expect similar behavior for paramagnetic systems whenever $T_1 \gg T_R$ and $T_2 \gg T_R$.[14]

The striking similarity between the paramagnetic and the optical equations is further exemplified by deriving a relation between T_R and T_1 in the paramagnetic case. Time-dependent perturbation theory gives

$$T_{1l}^{-1} = (2\pi/\hbar)\left|\epsilon_k e^{ikj'a}\right|^2 \rho_l(\hbar\omega), \tag{17}$$

where T_{1l} is the relaxation time in our linear chain at zero temperature. The quantity $\epsilon_k e^{ikj'a}$ is the matrix element between the state in which only atom j' is excited and the lattice is in the ground state, and the state in which atom j' is de-excited and one phonon has been emitted into the kth mode. Then[15]

$$\epsilon_k = -\epsilon(\hbar^3/2mN\omega)^{1/2}\sin(ka), \tag{18}$$

where N is the total number of paramagnetic spins. The number of modes per unit energy interval is

$$\rho_l(\hbar\omega) = L/\hbar\pi v. \tag{19}$$

Then in the limit $ka \ll 1$,

$$T_{1l} = \frac{mv\omega}{\epsilon^2 a^2 \hbar k^2}\frac{N}{L}. \tag{20}$$

The relaxation time in three dimensions at zero temperature should be

$$T_1 = T_{1l}\,\rho_l(\hbar\omega)/\rho(\hbar\omega), \tag{21}$$

where $\rho(\hbar\omega) = \omega^2 V/2\pi^2\hbar v^3$ is the number of modes per unit energy interval in a sample of volume V, and for the sake of simplicity it is assumed that the magnitude of the matrix element for emission is the same in all directions. Using Eq. (20), one gets

$$T_1 = \frac{\lambda}{a}\frac{1}{\epsilon a}\frac{mv^2}{\hbar\epsilon k^2}\frac{N}{V} \tag{22}$$

and, using Eq. (16),

$$T_R = T_1(n_0\lambda^2 L/4\pi)^{-1}, \tag{23}$$

where $n_0 = N/V$. This relationship is almost identical[16] to that between T_1 and T_R in the optical superradiance case,[3] except that there T_1 is the single atom radiation time ("T_{sp}" of Ref. 3), n_0 is the molecular excitation density, and λ is the optical wavelength.

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[14]For short T's, Eqs. (10) must be solved by computer, and different behavior can occur. Indeed, for $T_2 > T_R$ and $T_1 \gg T_R$, results similar to those of the Brya-Wagner experiment have been obtained.

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[16]In the case of optical superradiance, the corresponding formula is $T_R = T_1 (n_0 \lambda^2 L/8\pi)^{-1}$. The difference of a factor of 2 between the spin-phonon case and the electromagnetic case is due to the fact that in the optical case there are two transverse polarizations for each radiation mode, whereas in the spin-phonon case treated above, the modes are longitudinal with a single polarization. The extra factor of 2 in the optical case arises because T_1^{-1} is the spontaneous emission rate into *both* polarizations.