

Radiation Damping in Magnetic Resonance Experiments

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Magnetic resonance experiments can be described by analogy to a coupled pair of circuits, one of which is the ordinary electrical resonant circuit. The other circuit is formed by the rotating magnetization. For transient phenomena, such as occur, e.g., in the pulse techniques of free nuclear induction, the coupling gives rise to a damping of the magnetic resonance by the electric circuit. Such damping can also be considered as spontaneous radiation damping. It is shown that in certain cases of nuclear induction this radiation damping is more important than the damping from the spin-spin and the spin-lattice relaxation mechanisms usually considered. For ferromagnetic materials at microwave frequencies the radiation damping can become very large.

I. FREE MAGNETIC INDUCTION

BY way of introduction, consider a system of volume V with a uniform macroscopic magnetization per unit volume \mathbf{M}_0 , which precesses with an angular frequency

$$\omega_0 = \gamma H_0 = g\beta\hbar^{-1}H_0 \quad (1)$$

around a constant magnetic field H_0 , parallel to the z axis. Here g is the gyromagnetic ratio, β the Bohr magneton. The angle between \mathbf{M}_0 and \mathbf{H}_0 is θ_0 . Assume that the magnetization can precess freely, $M_x = M_0 \cos\theta \sin\omega t$, $M_y = M_0 \cos\theta \cos\omega t$, and $M_z = M_0 \sin\theta$ and neglect for the time being any internal damping of the magnetic system. When we introduce a pickup coil, tuned by a condenser to the frequency ω_0 , in the x - y plane, a periodic voltage is induced in this coil by the precessing magnetization. If the coil has n turns of cross section A , this induction signal is given by

$$V_s = -nA\eta \frac{4\pi}{c} \frac{dM_x}{dt} = nA \frac{4\pi\omega}{c} \eta M_0 \sin\theta \cos\omega_0 t, \quad (2)$$

where η is a filling factor, equal to unity if the coil is completely immersed in the magnetic material, or otherwise

$$\eta = \frac{\int \mathbf{M} \cdot (\mathbf{H}_c/i_c) dV}{\mathbf{M}_0 \cdot \int (\mathbf{H}_c/i_c) dV}, \quad (3)$$

where \mathbf{M} is the magnetic moment per unit volume and \mathbf{M}_0 its average value, allowing the extension to a sample of nonuniform magnetization. Each point in the integration over the space coordinates is weighted by the field \mathbf{H}_c/i_c which a unit current through the coil would produce at that point.

A current in phase with the induced voltage will flow in the tuned circuit, and the Joule heat dissipated per unit time is $V_s^2/2R$. The energy source which generates this power is the magnetic energy of the magnetized sample in the field H_0 . The energy of the permanent

magnet $\mathbf{M}_0 V$ is

$$W = M_0 V H_0 (1 - \cos\theta). \quad (4)$$

As the current flows the angle θ will gradually decrease to zero. The equation of motion can be written as the requirement of conservation of energy,

$$dW/dt = M_0 V H_0 \sin\theta (d\theta/dt) = V_s^2/2R. \quad (5)$$

The motion can also be found by considering the torque exerted on the magnetization by the field connected with the induced current. In a long coil of n turns over a length l , the induced field is

$$H_z = 4\pi V_s n / clR = 4\pi\eta Q M_0 \sin\theta \cos\omega_0 t.$$

The quality factor Q of the circuit is given by

$$Q = \omega L/R = \omega 4\pi n^2 A / Rlc^2.$$

The torque produced by this field gives rise to the equation of motion:

$$dM_z/dt = -\gamma M_y H_z,$$

or

$$d\theta/dt = -2\pi\eta M_0 Q \gamma \sin\theta. \quad (6)$$

Equation (5) can easily be reduced to this same form. The solution of Eq. (6) for the special case that $\theta = \pi/2$ at $t=0$ is

$$\tan(\frac{1}{2}\theta) = \exp(-2\pi M_0 Q \eta \gamma t), \quad (7)$$

and the amplitude of the induction signal Eq. (2) decreases proportional to $\text{sech}(2\pi M_0 Q \eta \gamma t)$. A damping time constant due to the reaction of the induced field on the magnetization can be defined as

$$\tau_R = (2\pi\eta M_0 Q \gamma)^{-1}. \quad (8)$$

Suryan¹ has first called attention to the importance of this type of damping without giving a detailed quantitative discussion. We shall give the pertinent circuit equations, from which the above result can be derived as a special case, in the next section. The relation of this damping to magnetic relaxation processes will be discussed in a final section, where, also, a comparison will be made with the natural line width of optical spectral lines. This radiation damping has an

¹ G. Suryan, *Current Sci. (India)* 18, 203 (1949).

appreciable magnitude because of the coherence between the individual spins producing the magnetization. These coherence effects have been discussed from a fundamental quantum mechanical point of view by Dicke.² The fact that it is possible by radiospectroscopy techniques to produce states with definite coherent phase relations between the individual elements, allows for a classical discussion of the problem in terms of one macroscopic magnet as first introduced by Bloch³ in his classical theory of nuclear induction. We shall illustrate now with a few examples that the damping described by Eq. (8) is frequently not negligible.

a. The Proton Resonance in Water in a Field of 7000 gauss

In thermal equilibrium the protons acquire a magnetization per unit volume in this field given by

$$M_0 = \chi_0 H_0 = [N_0 \gamma^2 \hbar^2 I(I+1)/3kT] H_0,$$

where χ_0 is the nuclear paramagnetic volume susceptibility. At room temperature the static magnetization is about 2×10^{-6} oersted in a field of 7000 oersteds. A short radiofrequency pulse^{4,5} at the resonance frequency and of amplitude H_{rf} and duration t such that $\gamma H_{rf} t = \frac{1}{2}\pi$, will turn this magnetization into the plane perpendicular to H_0 . A coherent "superradiant" state is thus produced. At the end of the pulse free nuclear induction will occur which is damped in a time τ_R given by Eq. (8). Assuming that the resonant circuit has a $Q=100$ and $\eta=1$, we find $\tau_R=0.03$ sec. This is much shorter than the damping from spin-spin and spin-lattice relaxation mechanisms in pure water. The fact that spin-echo pulses can be obtained after times much longer than τ_R is explained by the absence of this radiation damping when the nuclear spins are out of phase in the inhomogeneous field. To make the radiation per echo negligible the inhomogeneity ΔH should satisfy the inequality $\gamma \Delta H \tau_R \gg 1$. This allows detection of fine structures with separations much smaller than the inverse damping time given by Eq. (8).

When the initial magnetization is reversed by a 180° pulse, an unstable radiationless state is established. The magnetization will change only due to the spin-lattice relaxation mechanism, the angle θ remaining 180° .

b. The Proton Resonance in Water in the Earth's Magnetic Field

Recently Packard and Varian have observed the proton resonance in the earth's field at 2185 cps.⁶ The radiant state was produced by magnetizing the sample in a field H_0' of about 100 gauss perpendicular to the

earth's field. The field H_0' is reduced to a field H_0'' of a few gauss in a time short compared to the spin lattice relaxation time. During this adiabatic demagnetization the magnetization $M_0 = \chi_0 H_0'$ remains unchanged. Then the field H_0'' is reduced to zero in a time t' such that $\gamma H_0'' t' \ll 1$. During this process, which is nonadiabatic in the Ehrenfest sense, the magnetization vector has no time to reorient. Thus, a radiant state is constructed with a magnetization $M_0 \approx 3 \times 10^{-8}$ oersteds precessing with a frequency of 2185 cps in the plane perpendicular to the earth's field. The radiation damping time τ_R , assuming $\eta=1$, is about $200Q^{-1}$ sec. For sufficiently high Q , this could explain the observation that the duration of the signal was shorter than the known relaxation times in pure water. It is interesting to note that in this sense a spectral line in the audiofrequency range of the electromagnetic spectrum has a "natural width."

c. A Small Ferrite Sphere in a Microwave Cavity

Assume that a ferrite with a volume magnetization of 300 gauss cm^{-3} has a g value equal to that of a free electron and is placed in a cavity with $Q=2000$. For a sphere of 0.2 mm in diameter in a cavity of 10 cc, the filling factor η is of the order of 10^{-6} . Then we find that $\tau_R 10^{-8}$ sec. This time is inversely proportional to the filling factor or roughly to the volume of the ferrite sample.

It seems as if the radiation damping would be able to broaden the line so much as to wipe out the resonance completely for larger spheres. When the time τ_R becomes shorter than the characteristic time of the electrical circuit Q/ω , the analysis of this introductory paragraph is not valid. We shall therefore proceed with a more accurate description of the coupling.

II. THE COUPLING BETWEEN THE RESONANT CIRCUIT AND THE MAGNETIZATION

The equations of motion describing the complete system consisting of the magnetic material in a constant field H_0 in the z direction and two crossed coils parallel to the x and y direction, respectively, are

$$\begin{aligned} \frac{dM_{x,y}}{dt} &= \gamma(\mathbf{M} \times \mathbf{H})_{x,y} - \frac{M_{x,y}}{T_2}, \\ \frac{dM_z}{dt} &= \gamma(\mathbf{M} \times \mathbf{H})_z - \frac{M_z - M_0}{T_1}, \\ -K_x \frac{dM_x}{dt} + L_x \frac{di_x}{dt} + R_x i_x + \frac{1}{C_x} \int i_x dt &= V_{x, \text{app}}, \\ -K_y \frac{dM_y}{dt} + L_y \frac{di_y}{dt} + R_y i_y + \frac{1}{C_y} \int i_y dt &= V_{y, \text{app}}, \\ H_x &= K_x' i_x, \quad H_y = K_y' i_y, \quad H_z = H_0. \end{aligned}$$

² R. H. Dicke, Phys. Rev. **93**, 99 (1954).

³ F. Bloch, Phys. Rev. **70**, 460 (1946).

⁴ E. L. Hahn, Phys. Rev. **80**, 580 (1950); **88**, 1070 (1952).

⁵ H. Y. Carr, and E. M. Purcell, Phys. Rev. **94**, 630 (1954).

⁶ M. Packard and R. Varian, Phys. Rev. **93**, 941 (1954).

In general there are five simultaneous nonlinear differential equations connecting the three components of magnetization and the two currents. The first three equations are the familiar Bloch equations.³ The transverse field components H_x and H_y are now, however, related to the magnetization by the last two equations and must not be considered as impressed constants. The phenomenological damping terms for the spin-spin and spin-lattice relaxation are responsible for the fact that the magnetization has now in general not a constant magnitude M_0 . Frequently there will be only one coil, and the number of equations is consequently reduced by one. In writing the relations between the transverse components of the field and the currents we have assumed that demagnetizing and anisotropy fields are zero. The K and K' are geometrical factors. They are related to each other by $KK' = 4\pi L\eta$. The symmetrical arrangement of two crossed coils with identical circuit constants has the advantage that rotating fields can be produced. If the x and y components of the driving force are the real and imaginary parts of one complex function $V_{\text{app}}^+ = V_{x,\text{app}} + jV_{y,\text{app}}$ the solutions for $M^+ = M_x + jM_y$, $i^+ = i_x + ji_y$ can be found from a set of only three differential equations:

$$-K \frac{dM^+}{dt} + L \frac{di^+}{dt} + Ri^+ + \frac{1}{C} \int i^+ dt = V_{\text{app}}^+, \quad (9)$$

$$\frac{dM^+}{dt} = -j\gamma M^+ H_0 - \frac{M^+}{T_2} - j\gamma M_z K' i^+, \quad (10)$$

$$\frac{dM_z}{dt} = \text{Im}(\gamma K' M^+ i^+) - \frac{M_z - M_0}{T_1}. \quad (11)$$

We first give the steady-state solution, when the circuit is driven by a harmonic rotating potential $V_{x,\text{app}} + jV_{y,\text{app}} = V_{\text{app}} \exp(-j\omega t)$. A solution with $dM_z/dt = 0$ and the other components varying with the frequency ω is obtained, $M^+ = M_1 \exp(-j\omega t)$, $i^+ = i \exp(-j\omega t)$.

$$M_z = \frac{1 + (\Delta\omega T_2)^2}{1 + (\Delta\omega T_2)^2 + \gamma^2 H_0^2 T_1 T_2} M_0, \quad (12)$$

$$M_1 = \chi H_1 = \chi K' i = -\frac{j|\gamma| M_z T_2}{1 + j(\omega_0 - \omega) T_2} K' i, \quad (13)$$

$$i = [j\omega L(1 + 4\pi\eta\chi) + R + (j\omega C)^{-1}]^{-1} V_{\text{app}}. \quad (14)$$

This solution is identical with those usually given for the stationary state.⁷ The effect of the rotating magnetization can be described by a complex susceptibility χ . The over-all impedance of each circuit is given by the expression between square brackets in Eq. (14). There is no additional radiation damping of the magnetic resonance in this case. The z component of the magnet-

ization is constant. The generator supplies a current which exactly balances the effect of the radiation damping. The steady-state response of a circuit should be analyzed in terms of the impedance given by Eq. (14). Under suitable steady-state conditions it is possible to resolve fine structure lines of nuclear magnetic resonance in liquids which are only a few cycles apart, although these lines under conditions of free nuclear induction would be damped in a time short compared to their inverse spacing. The same situation prevails in ferromagnetic resonance experiments. Ordinary resonance lines are observed under steady-state conditions, although the freely precessing magnetization would radiate with a time short compared to the inverse line width.

The expression for the impedance for small H_1 , allowing M_z to be taken equal to M_0 , can be seen to be identical to that of a double tuned coupled circuit. It is therefore apparent that a condition of critical or more than critical coupling can be established and that a resonance detector that uses the electrical circuit as the frequency determining element of oscillation⁸ is subject to frequency pulling in a discontinuous manner, sometimes called a "drag link" effect.

For transient phenomena the Eqs. (10) and (11) for the magnetization cannot be considered independently from the circuit Eq. (9). The solution in the introductory section is obtained as an approximation by dropping the last two terms in Eq. (10) and the last term in Eq. (11). The damping time τ_R is found to be half as long as given by Eq. (8) because of the presence of two crossed coils creating a rotating reaction field.

A general solution cannot be obtained since nonlinear terms containing products $M_z i^+$ and $M^+ i^+$ occur. For small values of θ , we can, however, replace M_z in Eq. (10) by M_0 . Equations (9) and (10) then represent two coupled linear circuits. In the absence of a driving force we obtain a third degree equation for the proper frequencies of the system. If we assume that the electric circuit is tuned to the precession frequency of the magnetization $\omega_0^2 LC = \gamma^2 H_0^2 LC = 1$ and neglect terms of the order $\Delta\omega/\omega_0$, we obtain a quadratic equation for the complex proper frequencies $\omega = \omega_0 + \Delta\omega$ of the system, with the solution

$$\Delta\omega = \frac{j}{2} \left(\frac{\omega_0}{2Q} + \frac{1}{T_2} \right) \pm \left\{ -\frac{1}{4} \left(\frac{\omega_0}{2Q} + \frac{1}{T_2} \right)^2 + \frac{\omega_0}{Q} \left(\frac{1}{\tau_R} + \frac{1}{T_2} \right) \right\}^{\frac{1}{2}},$$

where τ_R is given by Eq. (8).

For negligible spin-spin damping, we can consider the following two limiting cases:

Case I, $1/\tau_R \ll \omega_0/Q$. The solutions are $\Delta\omega = j\omega_0/2Q$ or $\Delta\omega = 2j/\tau_R$. The first solution corresponds to the damping of the mode with the electric circuit excited,

⁸ R. V. Pound and W. D. Knight, Rev. Sci. Instr. **21**, 219 (1950).

⁷ N. Bloembergen, thesis, Leiden, 1948.

the second to the damping of the magnetization. This last solution corresponds to the examples of nuclear induction of Sec. I. In example **b** there is no current flowing at $t=0$. We must take a linear combination of the two solutions, and initially no damping. Only after a time Q/ω_0 when the electric circuit has been excited, is the damping given by the characteristic time τ_R .

Case II, $1/\tau_R \gg \omega_0/Q$. The solutions are now

$$\Delta\omega \approx j\omega_0/4Q \pm (\omega_0/Q\tau_R)^{1/2}.$$

The energy swings back and forth between the electric circuit and the magnet system with a frequency $(\omega_0/Q\tau_R)^{1/2}$ until the energy is eventually dissipated in the resistance of the electric circuit. This situation can be realized by ferrites in a microwave cavity. The energy is never dissipated faster than is permitted by the Q of the cavity,⁹ so long as $1/T_2$ is negligible. It is quite possible, however, that the magnetization of the ferrite at the end of a microwave pulse will return to its position parallel to H_0 in a time short compared to both relaxation times T_1 and T_2 . This observation does not alter the interpretation of the data in reference 9 since the relaxation times were evaluated from the power absorbed in the steady-state existing for the duration of the microwave pulse. The relaxation times thus obtained are independent of the size of the sample or the filling factor.

In the third mode of the system $\omega \approx -\omega_0$, the electrical circuits are excited at the same frequency but produce a field rotating in the opposite direction. This mode is only very slightly perturbed by the presence of the magnetization.

III. SPONTANEOUS EMISSION AND THERMAL RELAXATION MECHANISMS

The transition probability for spontaneous emission of a quantum $h\nu = \gamma\hbar H_0$ by a single spin $I = \frac{1}{2}$ is given by the well-known formula of radiation theory,¹⁰

$$W = 16\pi^2\gamma^2 h\nu^3 c^{-3}. \quad (15)$$

Substituting numerical values, we find for the lifetime of a single free proton in a field of 10^4 oersted the astronomical value $T_1 = 10^{25}$ sec. In the earth's magnetic field the lifetime against spontaneous radiation would be $T_1 \approx 10^{38}$ sec. How can this result be consistent with the radiation damping of the order of one second calculated in Sec. I? This discrepancy is resolved by considering two factors. One is the coherence which exists between the individual proton spins, the other is the increase in the density of the radiation field in the tuned circuit over that in free space.¹¹

In the situation of interest, the spins of protons have

⁹ N. Bloembergen and S. Wang, Phys. Rev. **93**, 72 (1954).

¹⁰ W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1943).

¹¹ E. M. Purcell, Phys. Rev. **69**, 681 (1946).

such phase relations that they create a macroscopic magnetic dipole which is N times as large as that of a single one. Since the emitted radiation is proportional to the square of the dipole moment, the transition probability is increased by a factor N^2 . Dicke² has given a quantum mechanical analysis of the coherent state and also arrives at this result. The damping is, however, not increased simply by a factor N^2 , but by a factor N . The quantum levels of our macroscopic system are all equally spaced. We must not consider a single transition but the damping of an N -fold excitation with coherence between all possible transitions. The same problem arises in calculating the lifetime of an excited harmonic oscillator.¹² Classically, the factor N is immediately obvious, as the radiation rate is proportional to N^2 and the stored energy proportional to N . The factor N is related to the magnetization introduced earlier by

$$N = M_0 V_m / \gamma \hbar. \quad (16)$$

Here V_m is the volume of sample.

The magnetic radiation density in the coil of volume V_c of a resonant circuit is increased over the density in free space by a factor

$$Q\lambda^3/8\pi^2 V_c. \quad (17)$$

Note that this factor increases with increasing wavelength, cancelling the decrease predicted by Eq. (15). Multiplying Eq. (15) by Eqs. (16) and (17) we find a radiation damping time τ_R which is identical with Eq. (8), provided we multiply by a factor $\frac{1}{2}$ to take account of the fact that only one of the two circularly polarized modes in the coil is effective.

The interaction of a single spin with a thermal radiation field leads to induced emission and absorption. The transition probability for these processes is obtained by multiplying Eq. (15) by the Bose-Einstein factor,

$$[\exp(h\nu/kT) - 1]^{-1} \approx kT/h\nu \gg 1, \quad (18)$$

for the frequency range of interest in magnetic resonance. The damping for the radiation from a single spin is consequently increased by this rather large factor.¹³

This factor should, however, not be added to Eq. (8). The influence of the thermal radiation field in the case of coherent spins in a resonant circuit can best be discussed in a classical manner. The classical analog of the magnetic thermal radiation field is the field produced in the coil by the thermal noise current $i(t)$. This current produces a fluctuating torque on the magnetization, leading to the equation of motion:

$$d\theta/dt = \gamma H_{th}(t). \quad (19)$$

Since $\langle H_{th}(t) \rangle_{av} = 0$, there is on the average no change in θ , due to the noise current. There are only fluctuations

¹² V. Weisskopf and E. Wigner, Z. Physik **65**, 18 (1931).

¹³ V. Weisskopf, Z. Physik **34**, 1 (1933).

in θ . The mean square deviation of θ after a time t' is

$$\begin{aligned} \langle \theta_{t'}^2 \rangle_{Av} &= \gamma^2 t' \left\langle \int_0^\infty H_{th}(t) H_{th}(t+t') dt' \right\rangle_{Av} \\ &= \gamma^2 \langle H_{th}^2 \rangle_{Av} \frac{L}{R} t' = \gamma^2 \frac{L}{R} \frac{4\pi kT}{V_c} t'. \end{aligned}$$

The average rate of change in the stored energy due to the interaction with the thermal noise field is

$$\begin{aligned} \langle (dW/dt)_{\text{noise}} \rangle_{Av} &= \frac{1}{2} M_0 V H_0 \cos\theta \langle d(\theta^2)_{Av}/dt \rangle \\ &= 2\pi kT \gamma Q \eta M_0 \cos\theta, \end{aligned} \quad (20)$$

and the average thermal damping is consequently

$$\left\langle \frac{1}{\tau_{th}} \right\rangle_{Av} = 2\pi \gamma Q M_0 \frac{kT}{M_0 H_0 V_c} \frac{\cos\theta}{(1-\cos\theta)}. \quad (21)$$

Apart from the angular dependence, this is smaller than Eq. (8) by a factor equal to the thermal energy kT over the total magnetic energy of the system. It is very small for a large coherent magnetization, but reduces properly to the result of increased damping for an individual spin. One might put the result Eq. (21) in words by saying that there is no coherence between the rotating magnetization and the thermal field, while there is between the rotating magnetization and its own reaction field.

The relaxation or damping discussed here refers only to the approach to equilibrium energy of orientation of the classical precessing magnet that corresponds to the given coherent state. Changes in the over-all magnetization of the system that lead toward a true equilibrium state of magnetization can only occur through the mechanisms leading to T_2 and thus cannot occur in times shorter than T_2 . Thus, a spin temperature is defined only after the time T_2 , the spin-spin relaxation time, has elapsed. It is not correct to say that after a radiofrequency pulse which has turned the magnetization through an angle between 90° and 180° , the spin system has assumed a negative temperature. Immediately after the pulse there are certain phase

relations between the spins, which change to a situation of internal thermal equilibrium¹⁴ only after a time T_2 . The radiation processes which do not change the coherence relations among the spins, do not contribute to the spin-spin relaxation mechanism.

A significant remark may be made about the conditions under which τ_R may be expected to be as short as T_2 . In a rigid lattice, T_2 is of the order of magnitude of $r^3/\gamma^2 \hbar$ where r is the interspin distance. Since r^3 is approximately $1/N_0$, for magnetization described by Curie's law, $(\tau_R/T_2) \sim kT/\omega \hbar \eta Q$. Thus, unless ηQ is quite large, only under the condition $\omega \hbar \gg kT$, leading to saturation magnetization, would $\tau_R \ll T_2$. The liquid narrowing, in the example of proton resonance, and the exchange effect, in the ferromagnetic case, are essential to the presence of a significant radiative effect.

Finally, we wish to stress the fact that a coherently radiating state can only be produced after a certain macroscopic magnetization has been established. It is still necessary that a relaxation mechanism produces a thermal distribution over the various energy levels of the individual spins, which are initially incoherent. A homogeneous magnetic field, such as exists in the coil and in general in a system which has dimensions small compared to the wavelength of the radiation, will never produce the initial thermal relaxation. The Hamiltonian for the interaction energy with a homogeneous magnetic field commutes with the total angular momentum or magnetization of the sample, which is therefore a constant of the motion. Radiation damping discussed in this paper does therefore not represent a spin-lattice relaxation mechanism in the usual sense. To establish a magnetization in thermal equilibrium with the surroundings, the inhomogeneous internal or local fields⁷ are essential. The inhomogeneity of the rf field in the coil was explicitly taken into account in the definition of the filling factor η . This results in a complication that the total magnetization is not rigorously a constant of the motion. The gist of the statement, however, that the radiation field in the coil does not provide a microscopic thermal relaxation mechanism is still valid.

¹⁴ E. M. Purcell and R. V. Pound, Phys. Rev. **81**, 279 (1951).