T_2 relaxation due to two-level field fluctuations

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The NMR relaxation rates T_1 and T_2 are usually assumed to be proportional to the spectral density of the field fluctuations at some frequency ω_p. While this is true for T_1 (with ω_p the Larmor frequency), this study of T_2 relaxation induced by two-level fluctuations of the z component of the magnetic field shows that T_2 cannot be described by taking the spectral density function at any one frequency. [S0163-1829(97)04613-4]

The NMR spin-phase memory time T_2 is often used as a probe for low-frequency magnetic field oscillations. For example, it allows the study of the mobility of vortices in superconductors.[1–5] However, relating the spin-phase memory relaxation time to physical parameters is not trivial: We show that even in the simplest case, the two-level fluctuation of the z component of the field, room for improvement of current understanding is available. It is generally assumed that T_2^{-1} probes the fluctuating field spectral density $f(\omega)$ at some frequency ω_p, so that $T_2^{-1} \propto \gamma_n^2 f(\omega_p)$. This probing frequency ω_p is often approximated by 0, although it has been argued[1] that $\omega_p = T_{2\mathrm{res}}^{-1}$ gives better results, with $T_{2\mathrm{res}}$ the relaxation time in the absence of these fluctuating fields. While this spectral density approach works for T_1 (with ω_p equal to the Larmor frequency), this article shows that for T_2, no such simplification can be made.

For the analysis, we take an infinite number of spins, oriented in the xy plane (the situation after a $\pi/2$ pulse and during a $\pi/2$-π pulse T_2 measurement). All spins ''feel'' an individual magnetic field b_z jumping between $\pm b_z$ with a correlation time τ $[\langle b_z(0) b_z(t)\rangle = b_z^2(0)\exp(-t/\tau)]$. Spin-lattice and spin-spin interactions are neglected. The time derivative of the phase in the presence of the field is $\Omega = \pm \gamma_{\mathrm{n}} b_z$. The number of spins per phase unit with a given phase ϕ at time t is written as $\overline{\Phi}(\phi,t)$. At $t=0$, we prepare the sample with $\overline{\Phi}(\phi,0) = \delta(\phi)$ (the situation after the $\pi/2$ pulse). Obviously, $\overline{\Phi} = \overline{\Phi}_+ + \overline{\Phi}_-$, with $\overline{\Phi}_\pm$ the density of spins that ''feel'' a positive resp. negative field. The differential equations that govern all are

$$\frac{d\overline{\Phi}_\pm(\phi,t)}{dt} = \mp \Omega \frac{d}{d\phi}\overline{\Phi}_\pm(\phi,t) + \tau^{-1}[\overline{\Phi}_\mp(\phi,t) - \overline{\Phi}_\pm(\phi,t)]. \tag{1}$$

Using $\overline{\Phi}_+(\phi,t) = \overline{\Phi}_-(-\phi,t)$, and substituting $\Phi_\pm(\phi,t) = e^{t/\tau}\overline{\Phi}_\pm(\phi,t)$, this simplifies to the uncoupled differential equations

$$\frac{d\Phi_\pm(\phi,t)}{dt} = \mp \Omega \frac{d}{d\phi}\Phi_\pm(\phi,t) + \tau^{-1}\Phi_\pm(-\phi,t). \tag{2}$$

With the definitions $x = (t\Omega - \phi)/\tau\Omega$ and $y = \sqrt{x(t\Omega+\phi)/\tau\Omega}$ it can easily be seen that for all[6] $|\phi| \leqslant \Omega t$ (or all real y)

$$4\tau\Omega\Phi_\pm(\pm\phi,t) = 2\tau\Omega\,\delta(\phi - \Omega t) + I_0(y) + xy^{-1}I_1(y), \tag{3}$$

with

$$I_0(y) = \sum_{n=0}^{\infty} \frac{y^{2n}}{(n!2^n)^2}$$

and

$$I_1(y) = \frac{y}{2}\sum_{n=0}^{\infty} \frac{y^{2n}}{(n!2^n)^2(n+1)}$$

the zeroth- and first-order modified Bessel functions, is a solution to Eq. (2); see also Fig. 1. It may be noted that, had the initial distribution been $\Phi'(\phi,0) = \delta(\phi - \phi_0)$, it evolves just like Eq. (3), only with the $2\delta + I_1$ term shifted by ϕ_0 to the left and the I_0 term shifted by the same amount to the right. This is useful in calculating the time evolution of a given $\Phi_0(\phi,t)$ (as was done in the numerical analysis later in this article).

Next the echo intensity decay after a $\pi/2$ - $t/2$ - π - $t/2$ pulse sequence will be evaluated, in the limits $\tau\Omega \ll \sqrt{2}$ and $\tau\Omega \gg \sqrt{2}$. In the first limit $\tau\Omega \ll \sqrt{2}$, all real decay takes place when $t/\tau \gg 1$: The signal amplitude decreases from the initial $M(0)$ to $[1 - \alpha(\tau\Omega)^2]M(0)$ at $t = \tau$ (with α of order 1), and

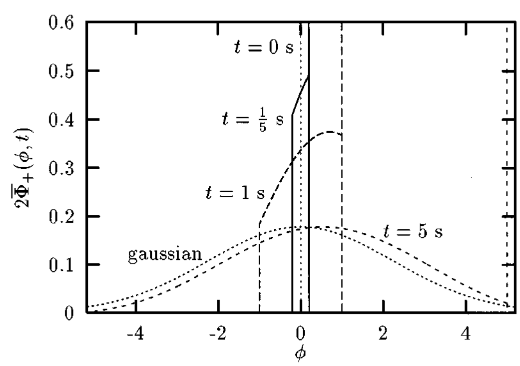

FIG. 1. Evolution of the phase density $\overline{\Phi}_+(\phi,t)$ of spins that ''feel'' a positive field at the time of measurement. The initial δ function at $t=0$ can be seen to move with angular frequency Ω to the right ($\Omega = \gamma_n b_z = 1\ \mathrm{s}^{-1}$ for all curves). At long times, the distribution becomes Gaussian [the line indicated with ''Gaussian'' is Eq. (5) with $t = 5$ s]. For all curves, $\tau = 1$ s.

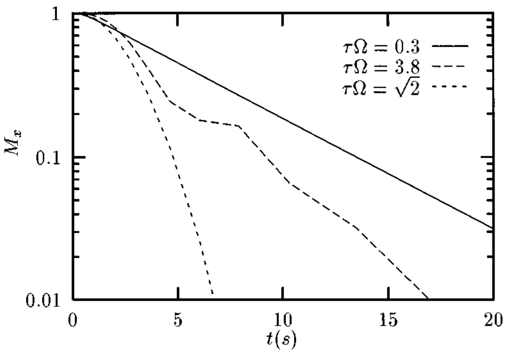

FIG. 2. Relaxation curves for various values of $\tau\Omega$ ($\Omega = 1.0\mathrm{s}^{-1}$ for all curves). The oscillation of the $\tau\Omega = 3.8$ line is not due to noise.

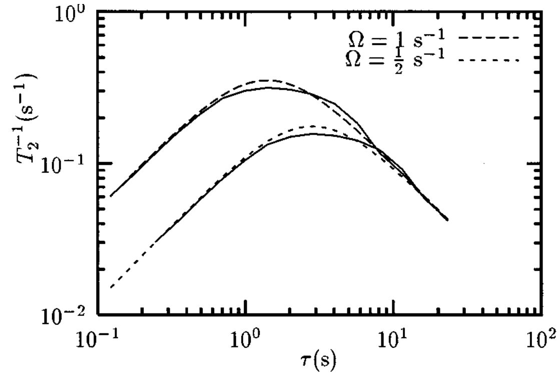

FIG. 3. T_2^{-1} vs τ. The simulation is for $\Omega = 1$ (upper curve) and $\Omega = 0.5$ (lower curve). The lines marked $\Omega = \frac{1}{2}$ and $\Omega = 1$ are the function $\tau\Omega^2/[2 + (\tau\Omega)^2]$ with the above-mentioned Ω's.

all measurable decay takes place when $t/\tau \gg 1$. On the other hand, in the limit of $\tau\Omega \gg 1$, one can simply observe the intensity of the δ peak that contains the spins whose field has not changed, and thus falls off in time as $e^{-t/\tau}$.

The total magnetization at time t can be evaluated using

$$M_x(t) = e^{-t/\tau} \int_{\phi=-\infty}^{\phi=+\infty} d\phi \cos\phi [\Phi_+(\phi,t) + \Phi_-(\phi,t)]. \qquad (4)$$

When $t/\tau \gg 1$ (the limit where all decay happens when $\tau\Omega \ll \sqrt{2}$), the Bessel functions can be approximated by $I_n(y) = (2\pi y)^{-1/2} e^y$, and, as $\Phi(\phi,t)$ is multiplied by $e^{-t/\tau}$, only values of $y \approx t/\tau$ or $|\phi| \ll t\Omega$ contribute in Eq. (4). Thus, we can approximate $y = t/\tau - \frac{1}{2}\phi^2/\tau t\Omega^2$, and $xy^{-1} = 1$, and the resulting phase distribution becomes

$$\overline{\Phi}_\pm(\phi,t) = \frac{1}{2\Omega\sqrt{2\pi t\tau}} e^{(-\phi^2/\tau t\Omega^2)/2}. \qquad (5)$$

As this phase distribution is symmetric in ϕ, the π pulse at time $t/2$ can be neglected, giving an echo (and free induction decay, in the case of no additional static line broadening) magnetization intensity of

$$M_x(t) = e^{-\tau t\Omega^2/2} = e^{-\tau(\gamma b_z)^2 t/2}. \qquad (6)$$

Thus, $T_2^{-1} = \frac{1}{2}\tau(\gamma b_z)^2$ for $\tau\Omega \ll 1$.

In the other limit $\tau\Omega \gg \sqrt{2}$, we only need to consider the evolution of the δ peak [as soon as a field hops, the phase of the corresponding spin moves so fast away from 0 that $\cos\phi$ averages to 0 in Eq. (4)], and Eqs. (3) and (4) simplify to

$$\overline{\Phi}_\pm(\phi,t) = \tfrac{1}{2}\delta(\phi) e^{-t/\tau}, \qquad (7)$$

$$M_x(t) = e^{-t/\tau}. \qquad (8)$$

The δ peak is at zero as t is the time of the echo. Equation (8) gives $T_2 = \tau$ when $\tau\Omega \gg \sqrt{2}$.

To address the region where $\tau\Omega \approx \sqrt{2}$, Eq. (3) was evaluated numerically. A π pulse was simulated at time $t/2$ by inverting the ϕ axis, and to evaluate the total magnetization at echo time t the resulting distribution was allowed to evolve again in time [also Eq. (2) was simulated to confirm the correctness of Eq. (3)]. This was done for various values of Ω, τ, and t, and from these experiments, T_2 was obtained by taking the time at which the intensity dropped to $1/e$ of the maximum. Three of the obtained M vs t curves are shown in Fig. 2. It can clearly be seen that the relaxation for $\tau\Omega = \sqrt{2}$ is Gaussian-like.

The T_2^{-1} versus τ curves are shown in Fig. 3. The above predicted limiting cases can be seen to be very accurate, and the (empirical) formula

$$T_2^{-1} = \tau\Omega^2/[2 + (\tau\Omega)^2] \qquad (9)$$

describes the data very well, with some deviation in the middle region. If we measure T_2 as a function of temperature (and thus vary τ), the condition for the fastest relaxation rate becomes $\tau\gamma_n b_z = \sqrt{2}$, with the optimum rate $T_2^{-1} = \frac{1}{4}\sqrt{2}\gamma_n b_z$.

In conclusion, we have shown that in the limit of $\tau\gamma_n b_z$ very large or very small, the spin-spin relaxation caused by a fluctuation of the z component of the magnetic field is equal to $T_2^{-1} = \tau(\gamma_n b_z)^2/[2 + (\tau\gamma_n b_z)^2]$. This contrasts with the spectral density approach $[T_2^{-1} \propto f(\omega_p)]$, with ω_p some given probing frequency, usually approximated by 0, sometimes taken to be just a low frequency. For example, in the two-level fluctuation case studied here, the spectral density is given by $f(\omega_p) \propto b_z^2\tau/[1 + (\tau\omega_p)^2]$, suggesting a relaxation rate $T_2^{-1} = (\gamma_n b_z)^2\tau/[1 + (\tau\omega_p)^2]$. This gives, whatever value of ω_p we take, incorrect T_2 values (a) at ''optimum'' τ, as we show it to be proportional to b_z, whereas the spectral density approach predicts $T_2 \propto b_z^2$, and (b) in the long-τ limit, as T_2 is shown to be independent of b_z in this limit, but the spectral density again predicts a proportionality to b_z^2 (this is because the ω_p in the spectral density approach is constant, and does not depend on b_z). Note that the exact rate in the small-τ limit differs by a factor of 2 from that derived by Slichter,[7] as the τ he uses is twice the average time between hops.

These results allow an estimation of b_z and τ from experimental T_2 curves.

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