Dynamics of a nuclear spin system at low temperatures

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We examine the saturation of a nuclear spin system by a radio-frequency magnetic field with allowance for the nonlinear effects due to the polarization dependence of the dynamic frequency shift and the linewidth. We study the dynamics of the spin system in the nonstationary mode and calculate the stationary values of polarization. Finally, we note that in the nonstationary case, as well as in the stationary, nonlinear effects weaken the influence of the radio-frequency field on the saturation of the nuclear spin system. © *1996 American Institute of Physics.* [S1063-7761(96)02609-1]

It is known that if the interaction between magnetic moments is long-range ($r_0 > a$, where r_0 is the interaction range, and a is the lattice constant), the so-called dynamic frequency shift[1] exceeds the linewidth even for small polarizations ($p \ll 1$), as observed experimentally. But if the interaction range r_0 is of the order of a, the dynamic frequency shift, i.e., the first moment M_1 (see Ref. 2), becomes comparable to the linewidth or exceeds it at high polarizations ($1-p \ll 1$), and hence at extremely low spin temperatures.

An example of an interaction of the first type is the one between nuclear spins in magnetically ordered specimens, where $r_0 \sim 100a$; the dynamic frequency shift shows up in spin echo experiments[1,3] and should be observed in stationary experiments.[4] In describing the saturation of the resonance line, the dependence on polarization is taken into account only in the shift of the resonance frequency, while the linewidth is assumed independent of polarization due to the smallness of the latter. In spite of this the problem acquires a nonlinearity, and the given phenomenon becomes more complicated to study but, as a result, more interesting.

Kurkin[4] examined the effect of the dynamic frequency shift on the formation of a stationary state of the nuclear spin system in ferromagnets when a low-amplitude radio-frequency magnetic field pumped the system. He found that due to nonlinear effects resulting from a sizable dynamic shift (larger than the linewidth), there can be different stationary states, including stable states. Note that nonlinear effects also show up when the saturation times are short, $t < T_2$ (with T_2^{-1} the linewidth), and they are appreciable in spin echo experiments.[1,3]

An example of an interaction of the second type is the nuclear spin system in diamagnetic materials, where there is magnetic dipole–dipole interaction between the magnetic moments.[2] In the given case, where at high polarizations ($1-p \ll 1$) a substantial dynamic frequency shift appears, we must take into account the polarization dependence of the linewidth. This complicates the problem, but here we consider the special cases in which nonlinearity emerges but the problem can still be solved completely.

When saturation times are short, or $t < T_2$, we must allow for polarization dependence only in the dynamic frequency shift, since such a dependence does not show up in the linewidth. Hence the dynamics of the nuclear spin system in diamagnetic materials does not differ significantly from the dynamics of the nuclear spin system in magnetically ordered specimens.

But if $T_2 < t < T_1$, with T_1 the longitudinal relaxation time, the variation of polarization can be described by

$$\frac{dp}{dt} = -2W(\Delta)p, \tag{1}$$

where $W(\Delta)$ is the transition probability between Zeeman levels stimulated by the radio-frequency field, $\Delta = \omega_0 - \omega$ is the offset from resonance, ω_0 is the Larmor frequency, and ω is the frequency of the radio-frequency field. Here

$$W(\Delta) = \tfrac{1}{2}\pi\omega_1^2 g(\Delta), \tag{2}$$

ω_1 is the amplitude of the radio-frequency field in frequency units, and $g(\Delta)$ is the line profile, which can be found by employing the method of moments.[2]

At high polarizations ($1-p \ll 1$) the line profile is Lorentzian:

$$g(\Delta) = \frac{1}{\pi}\frac{\tau_0^{-1}}{\tau_0^{-2} + (\Delta + M_1)^2}. \tag{3}$$

Here $M_1 = \alpha p$ is the first moment, and $\tau_0^{-1} = \sqrt{\pi/2}\sqrt{M_2/\mu}$, with $M_2 = M_2^{(0)}(1-p^2)$ the second moment, $\mu = M_4/M_2^2$, and $M_4 \simeq (M_2^{(0)})^2(1-p^2)$ the fourth moment (we assume that $p \simeq 1$). If the polarization of the nuclear spin system differs much from unity, the profile is not Lorentzian and the given approach becomes invalid, so that at high polarizations we restrict our discussion to small deviations of the initial value.

On the other hand, at small deviations we can ignore the effect of the variation of the average energy of the dipole–

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dipole reservoir on the dynamics of polarization variations,[5,6] so that Eq. (1) can be considered a good approximation when describing the saturation of the nuclear spin system in diamagnetic materials.

Summing up and defining T_2^{-1} as $\sqrt{2\pi M_2^{(0)}}$, we can write Eq. (1) in the following form[1)] (because $1-p\ll 1$, we must put $1-p^2\simeq 2(1-p)$):

$$\frac{dp}{dt}\simeq -\omega_1^2\frac{T_2(1-p)}{(1-p)^2+(\Delta+\alpha p)^2T_2^2}p. \tag{4}$$

When $p\ll 1$ but the dynamic frequency shift is still large ($\alpha pT_2>1$), as is the case with the nuclear spin system in magnetically ordered specimens, Eq. (4) assumes the form

$$\frac{dp}{dt}\simeq -\omega_1^2\frac{T_2}{1+(\Delta+\alpha p)^2T_2^2}p. \tag{5}$$

Equation (5) can easily be integrated, which yields an equation law governing the polarization for $p\ll 1$:

$$(1+\Delta^2T_2^2)\ln\frac{p}{p_0}+2\Delta\alpha T_2^2(p-p_0)$$
$$+\frac{1}{2}\alpha^2T_2^2(p^2-p_0^2)=-\omega_1^2T_2t, \tag{6}$$

where p_0 is the polarization at $t=0$ (the instant the radio-frequency field is turned on). Clearly, polarization gradually decreases to zero as $t\to\infty$ (spin–lattice relaxation is ignored). For small deviations from the initial state ($p_0-p\ll p_0$) Eq. (6) yields

$$\frac{p_0-p}{p_0}\simeq\frac{1}{1+(\Delta+\omega_p)^2T_2^2}\omega_1^2T_2t,$$

where $\omega_p\equiv\alpha p_0$. Hence the polarization shows the same behavior ($p_0-p\propto t$) as for zero dynamic shift ($\omega_p=0$). However, the rate at which the polarization varies depends on ω_p.

In the final stage of relaxation, when $p\ll p_0$, as Eq. (6) clearly shows, the dynamics of polarization is of an exponential nature and in no way differs from such dynamics when $\omega_p=0$.

Next, as Eq. (5) implies, when resonant pumping is initially present (i.e., $\Delta+\alpha p_0=0$), the rate at which polarization varies decreases as the deviation of polarization from the initial value grows. But when $\Delta=0$ at $t=0$, the rate at which polarization varies increases with time.

By integrating Eq. (4) we find that for high polarizations

$$T_2^2(\Delta+\alpha)^2\ln\frac{1-p}{1-p_0}+(p_0-p)\left[(\alpha^2T_2^2+1)\left(1-\frac{p+p_0}{2}\right)\right.$$
$$\left.-2\alpha(\Delta+\alpha)T_2^2\right]\simeq\omega_1^2T_2t. \tag{7}$$

When there is initially resonant pumping, the equation gets much simpler:

$$T_2^2\alpha^2(1-p_0)^2\ln\frac{1-p}{1-p_0}+(p_0-p)\left[(\alpha^2T_2^2+1)\right.$$
$$\left.\times\left(1-\frac{p+p_0}{2}\right)-2\alpha^2(1-p_0)T_2^2\right]\simeq\omega_1^2T_2t.$$

If the deviation of polarization from the initial value is small relative to $1-p_0$, i.e., $(p_0-p)/(1-p_0)\ll 1$, the polarization is proportional to time, as it is in the case of low polarizations:

$$p_0-p=\omega_1^2\frac{T_2t}{1-p_0}.$$

But if the relative deviation is large, i.e., $(p_0-p)/(1-p_0)\gg 1$, the polarization varies according to the following square-root law[2)]:

$$p_0-p=\sqrt{\frac{2\omega_1^2}{1+T_2^2\alpha^2}T_2t}, \tag{8}$$

which means that this case differs considerably from the one in which nonlinear effects are ignored and the deviation is proportional to time.

When $p_0\to 1$, the dynamics of saturation of the spin system obeys a square-root law from the very beginning (with respect to time).

Note that if $p_0=1$, saturation occurs only when $\Delta+\alpha p_0=0$. Indeed, when $\Delta+\alpha p_0\neq 0$, Eq. (7) has the solution[3)] $p=\text{const}=1$.

To examine the saturation of the NMR line with allowance for spin–lattice relaxation ($t>T_1$), we must add the relaxation term $(p-p_0)/T_1$ to the right-hand sides of Eqs. (4) and (5). For low polarizations Kurkin[4] obtained an equation for a stationary polarization value. At low pumping levels ($\omega_1^2T_1T_2\ll 1$) this equation has three real positive roots in the tuning range

$$\frac{3}{T_2}\left(\frac{\omega_pT_2s}{4}\right)^{1/3}\leqslant\tilde{\Delta}\leqslant\frac{1}{T_2}\omega_pT_2s, \tag{9}$$

where $s\equiv\omega_1^2T_1T_2$, and $\tilde{\Delta}=\Delta+\alpha p_0$. Then the expressions for the stable values of $z=(p_0-p_{st})/p_0$ have the form[4]

$$z_1\simeq\frac{s}{(\tilde{\Delta}T_2)^2},\quad z_3\simeq\frac{\tilde{\Delta}}{\omega_p}.$$

Next, if we compare the necessary condition for the existence of three roots, $(\tilde{\Delta})^2>3/T_2^2$, with Eq. (9), we arrive at

$$\omega_pT_2s>\sqrt{3},$$

i.e., the dynamic frequency shift at the start of saturation must be much larger than the line width (the reader will recall that $s\ll 1$). But if this condition is not met, there is only one unsaturated stable value,

$$z\simeq\frac{s}{1+\tilde{\Delta}^2T_2^2}.$$

For large initial polarizations ($1-p_0\ll 1$), by adding the relaxation term $(p_0-p)/T_1$ to the right-hand side of Eq. (4)

we can find the stationary polarization value (for small deviations from the initial value) in the limit $p_0=1$ of interest here:

$$z=\frac{s}{z^2+(\tilde{\Delta}-\alpha z)^2T_2^2}z. \quad (10)$$

The roots of this equation are

$$z_1=0,$$

$$z_{2,3}=\frac{\tilde{\Delta}\alpha T_2^2\pm\sqrt{\tilde{\Delta}^2\alpha^2T_2^4-(\tilde{\Delta}^2T_2^2-s)(1+\alpha^2T_2^2)}}{1+\alpha^2T_2^2}.$$

At

$$\sqrt{s}\leqslant\tilde{\Delta}T_2\leqslant\sqrt{s(1+\alpha^2T_2^2)} \quad (11)$$

Eq. (10) has three positive roots, of which z_1 and z_3 are stable and stationary. But at

$$|\tilde{\Delta}T_2|\leqslant\sqrt{s} \quad (12)$$

the state with z_1 is not stable and the saturated state with z_2 is realized. Since only small deviations are considered, the condition $\tilde{\Delta}\alpha T_2^2/(1+\alpha^2T_2^2)\ll 1$ must also be met, which is automatically the case in the given tuning range for a low pump amplitude $s\ll 1$.

Outside the ranges (11) and (12) we have only one solution, with $z=0$.

For $p_0\neq 1$ instead of (10) we have

$$z\simeq\frac{s(1-p_0+z)}{(1-p_0+z)^2+(\tilde{\Delta}-\alpha z)^2T_2^2}. \quad (13)$$

Here we examine the special (but realistic) case with $\alpha T_2\sim 1$, $s\ll 1$, and $(p_0-1)<s$. Analyzing the cubic equation (13) and allowing for the results obtained in the limit $p_0=1$, we derive approximate expressions here for the stationary stable values of polarization in various resonance-tuning ranges.

Only for $\sqrt{s(1+\kappa)}\leqslant\tilde{\Delta}T_2\leqslant\sqrt{s(1+\alpha^2T_2^2)}$, where ($\kappa\simeq\sqrt{1-p_0}/s^{1/4}\ll 1$), does Eq. (13) have three real positive roots, of which two correspond to stationary stable values of polarization:

$$z_1\approx\frac{(1-p_0)s}{\tilde{\Delta}^2T_2^2-s},$$

$$z_3=\frac{\tilde{\Delta}\alpha T_2^2+\sqrt{\tilde{\Delta}^2\alpha^2T_2^4-(\tilde{\Delta}^2T_2^2-s)(1+\alpha^2T_2^2)}}{1+\alpha^2T_2^2}.$$

In the remaining range of $\tilde{\Delta}$ there is only one solution. In particular, for

$$s(1+\alpha^2T_2^2)\leqslant\tilde{\Delta}^2T_2^2<\infty$$

and

$$-\sqrt{s(1+\alpha^2T_2^2)}\leqslant\tilde{\Delta}T_2<-\sqrt{s(1+\kappa)}$$

the stationary state with

$$z\approx\frac{(1-p_0)s}{\tilde{\Delta}^2T_2^2-s},$$

is realized, while for $-\sqrt{s(1+\kappa)}\leqslant\tilde{\Delta}T_2\leqslant-\sqrt{s}$ we have the state with

$$z\approx\sqrt{1-p_0}s^{1/4}.$$

In the range $\sqrt{s}\leqslant\tilde{\Delta}T_2\leqslant\sqrt{s(1+\kappa)}$ we already have the state with $z\approx\sqrt{s}$, while for $\tilde{\Delta}^2T_2^2<s$ we have the state with

$$z\approx\frac{\tilde{\Delta}\alpha T_2^2+\sqrt{\tilde{\Delta}^2\alpha^2T_2^4-(\tilde{\Delta}^2T_2^2-s)(1+\alpha^2T_2^2)}}{1+\alpha^2T_2^2}.$$

Note that the nontrivial solutions (with three roots) at high polarizations and low pump amplitudes ($s\ll 1$) exist even when the dynamic frequency shift is small compared to the line width ($\omega_pT_2\sim 1$). This is different from the situation for low polarizations, where the existence of nontrivial solutions for $s\ll 1$ requires that the dynamic frequency shifts at the start of saturation be large compared to the line width ($\omega_pT_2\gg 1$).

Note that in both cases, the nonstationary and the stationary, the effect of the radio-frequency field on the dynamics of saturation of the nuclear spin system due to nonlinear effects weakens. For instance, in the nonstationary mode Eq. (8) implies that the dynamics of polarization of the nuclear spin system follows a square-root law, while without nonlinear effects the variation would be proportional to time.

In the stationary case, comparison of the expressions for the stationary values of polarizations for $p\ll 1$ and $1-p\ll 1$ shows that the s-dependence of z is much stronger for $p\ll 1$ than it is for $1-p\ll 1$. The reason is that at high polarizations, the line width is polarization-dependent, and with increasing deviation of the polarization from its initial value (i.e., for decreasing polarization), the transition probability $W(\Delta)$ decreases (Eq. (4)), which weakens the s-dependence of z. At low polarizations the polarization dependence of the line width (due to the smallness of the former) can be ignored.

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[1)]Here for the sake of simplicity we restrict our discussion to positive polarization of the nuclear spin system. Our conclusions can easily be generalized to negative polarizations.

[2)]Here $1-p$ must always be much smaller than unity.

[3)]Here spin–lattice relaxation is ignored. If we take it into account, the derivation remains valid only if the state with $p=1$ is stable (later in our discussion we touch upon these matters in greater detail).

*Deceased

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