Thermodynamic theory of multipulse NQR experiments

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A thermodynamic theory of multipulse NQR is developed with the simplest multipulse spin locking sequence as the example. The influence of spin-lattice relaxation is taken into account. It is shown that the theory proposed agrees well with the known experimental data.

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In recently reported experiments[1,2] nuclear quadrupole resonance was observed by using multipulse sequences similar to those used in NMR. The observed echo signals attenuated in times much longer than the characteristic dipole-dipole interaction time T_2. This weakening of the influence of the dipole interaction provides new possibilities of increasing the sensitivity of the NQR method,[1-3] as well as of increasing the efficiency of the methods of studying molecular motions in solids.[3,4]

We note that multipulse NMR spectra were obtained way back[5] in 1966, and by now mutlipulse NMR spectroscopy is a rather advanced field. The advantages of multipulse spectroscopy over the ordinary one lies in the ability of suppressing the dipole interactions completely or in part by rapidly modulating them with sequences of specially chosen radiofrequency pulses. A rapidly oscillating dipole interaction, just as any rapidly oscillating force in classical mechanics,[6] is weakened by a factor $(\omega_m/\omega_{\text{loc}})^n$ times (ω_m is the dipole-interaction modulation frequency and ω_{loc} is the strength of the local magnetic fields), where n depends on the type of sequence employed. Multipulse spectroscopy is in fact spectroscopy with adjustable line width, in which a strong radiofrequency field is used not only to produce observable spectra, but also to regulate the widths of the individual lines in the spectrum.

In substances with quadrupole nuclei, the dipole interactions make also a substantial contribution to the form of the observable spectra, therefore multipulse methods offer great promise also in quadrupole resonance.

The experiments in Refs. 1 and 2 were performed by the multipulse spin-locking method,[5] in which the simplest sequence φ_0-$(\tau$-φ-$\tau)^n$ is used, where $\phi=\gamma H_1 t_w$ (H_1 is the field in the pulse and t_w is the pulse duration and 2τ is the distance between the pulses. The echo-production mechanism was explained in Ref. 7, and the behavior of the signal over long times was obtained in Ref. 8, where the principles underlying the theory of multipulse locking are briefly presented and some features of the experimental spectra of Refs. 1 and 2 are explained without allowance for spin lattice relaxation.

Up to now the multipulse spectra excited by the indicated sequence were investigated in greatest detail by Osokin,[9] and the purpose of the present paper is a consistent theoretical analysis of such spectra and a comparison of the theory with experiment. The experiments were performed on N^{14} nuclei. We confine ourselves therefore to the case of unity spin. Although we present here a solution for multipulse spin locking, the method can be applied to the analysis of any sequence. We consider a crystal sample containing nuclei with unity spin. We assume that the electric field gradients at different nuclei practically coincide (the scatter of the quadrupole frequencies does not exceed the value of the dipole-dipole interaction). The quadrupole interaction Hamiltonian

$$\mathcal{H}_Q=\tfrac{1}{3}\omega_Q[3I_z^2-I^2+\eta(I_x^2-I_y^2)] \tag{1}$$

can be expressed with the aid of the operators[10]

$$I_{p,1}=\tfrac{1}{2}I_p,\quad I_{p,2}=\tfrac{1}{2}(I_qI_r+I_rI_q),\quad I_{p,3}=\tfrac{1}{2}(I_r^2-I_q^2),$$
$$[I_{p,i}\,I_{p,j}]=iI_{p,k}, \tag{2}$$

where $p, q, r = x, y, z$; i, j, k are equal to 1, 2, 3 or their cyclic permutation, in the form[7]

$$\begin{aligned}\mathcal{H}_Q&=-\tfrac{2}{3}\omega_Q\eta I_{z,3}+\tfrac{2}{3}\omega_Q(I_{x,3}-I_{y,3})\\&=\omega_Q(1+\eta/3)I_{x,3}-\tfrac{1}{3}\omega_Q(I_{y,3}-I_{z,3})\\&=-\omega_Q(1-\eta/3)I_{y,3}-\tfrac{1}{3}\omega_Q(1+\eta)(I_{z,3}-I_{x,3}).\end{aligned} \tag{3}$$

Each expression consists here of two commuting terms

$$[I_{p,i},\,I_{q,3}-I_{r,3}]=0.$$

Let the carrier frequency of the RF pulses be

$$\omega=\omega_y-\Delta=-\omega_Q(1-\eta/3)-\Delta,$$

where ω_y is one of the three resonant frequencies of the quadrupole resonance. The interaction with the pulse is of the form

$$\begin{aligned}\mathcal{H}_1&=\mathbf{l}If(t)\cos\omega t\quad\text{(first pulse)},\\\mathcal{H}_1&=-\mathbf{l}If(t)\sin\omega t\quad\text{(remaining pulses)},\end{aligned} \tag{4}$$

where $\mathbf{l}$ is a unit vector along the RF-coil axis and $f(t)$ is the pulse function (the envelope of the pulses).

We write down the equation for the density matrix:

$$d\rho/dt=-i[\mathcal{H}_Q+\mathcal{H}_1(t)+\mathcal{H}_d+\mathcal{H}_\delta+G+\mathcal{H}_{IS}+\mathcal{H}_S+\mathcal{F},\ \rho], \tag{5}$$

where $\mathcal{H}_d$ is the dipole-dipole interaction of the considered nuclei; $\mathcal{H}_\delta$ is the Hamiltonian of the inhomogeneities and describes the scatter of the quadrupole frequencies; G is the spin-lattice interaction Hamiltonian; $\mathcal{H}_{IS}$ is the Hamiltonian that describes the motion of the nuclei S; $\mathcal{F}$ is the lattice Hamiltonian. At $|\mathcal{H}_\delta|\lesssim|\mathcal{H}_d|$ the inhomogeneity can be considered just as a dipole interaction. We shall therefore regard $\mathcal{H}_\delta$ as included in $\mathcal{H}_d$.

In the interaction representation, defined by the Hamiltonian

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$$\mathcal{H}_0=\omega I_{y,3}-{}^1/_3\omega_Q(1+\eta)(I_{z,3}-I_{x,3})+\mathcal{H}_S, \quad (6)$$

the equation for the density matrix takes the form

$$d\tilde{\rho}/dt=-i[\Delta I_{y,3}+\mathcal{H}_1^{\rm sec}(t)+\mathcal{H}_d^{\rm sec}+\mathcal{H}_{IS}^{\rm sec}+G^{\rm sec}+G'(t)+\mathcal{F}+V(t),\tilde{\rho}], \quad (7)$$

where the terms secular in $\mathcal{H}_0$ have been separated, $G'(t)$ is the nonsecular part of the spin-lattice interaction, and $V(t)$ are the remaining nonsecular terms. If all the frequencies $\mathcal{H}_Q$ and $\mathcal{H}_S$ are different (this is the only case we consider), $\mathcal{H}_{IS}^{\rm sec}=0$, for in the presence of quadrupole interaction all the local fields will oscillate in this case with frequencies on the order of ω_Q. In addition, we assume that the main contribution to the spin-lattice relaxation is made by the fluctuations of the electric field gradients. This makes it possible to leave out of (7) all the nonsecular terms except G', and assume that $\mathcal{F}$ commutes with all the operators except $G^{\rm sec}$ and G'.

The interaction with the pulses and the dipole interaction are given by

$$\mathcal{H}_1^{\rm sec}=f(t)\cos\theta I_{y,1}\ \text{(first pulse)},$$
$$\mathcal{H}_1^{\rm sec}=f(t)\cos\theta I_{y,2}\ \text{(remaining pulses)}, \quad (8)$$

$$\mathcal{H}_d^{\rm sec}=\sum_{i\neq j}\frac{\gamma^2\hbar^2}{r_{ij}^3}\sum_{p=x,y,z}a_{ij}^p(I_{p,1}^iI_{p,1}^j+I_{p,2}^iI_{p,2}^j)+\sum_i\delta_iI_{y,3}^i, \quad (9)$$

where θ is the angle between $\mathbf{l}$ and the y axis; $a_{ij}^p=1-3\cos^2\gamma_{ij}^p$; γ_{ij}^p is the angle between the internuclear vector $\mathbf{r}_{ij}$ and the p axis. If the duration of the pulses is neglected, the pulse envelope can be represented in the form

$$f(t)=\varphi_0\delta(t)+\varphi\sum_{k=0}^{\infty}\delta(\tau+2k\tau-t). \quad (10)$$

With the aid of (8) we find that after the first pulse the initial density matrix $\rho_0=1-\alpha_0\mathcal{H}_Q$ goes over into

$$\rho_+=\exp(-i\varphi_0\cos\theta I_{y,1})\rho_0\exp(i\varphi_0\cos\theta I_{y,1})=1+\alpha_0\omega_Q(1-\eta/3)\times\{I_{y,3}\cos(\varphi_0\cos\theta)-I_{y,2}\sin(\varphi_0\cos\theta)\}+\alpha_0\frac{\omega_Q}{3}(1+\eta)(I_{z,3}-I_{x,3}). \quad (11)$$

We determine now the effective field[11] ω_e that describes the action of the pulses and of the detuning during the time of the pulse sequence:

$$\exp(-2i\tau\omega_e\mathbf{I}_y)=\exp(-i\Delta\tau I_{y,3})\exp(-i\varphi\cos\theta I_{y,2})\exp(-i\Delta\tau I_{y,3}), \quad (12)$$

where $\mathbf{I}_y$ is the vector in the space $\{I_{y,i}\}$ and the modulus ω_e in the direction $\mathbf{n}$ of the vector ω_e are defined by the expressions

$$\cos(2\omega_e\tau)=\cos(\varphi\cos\theta)\cos^2(\Delta\tau)-\sin^2(\Delta\tau),$$
$$n_1=0,\quad n_2=\frac{\sin(\varphi\cos\theta)\cos(\Delta\tau)}{\sin(2\omega_e\tau)},$$
$$n_3=\frac{\sin(2\Delta\tau)\cos^2((\varphi\cos\theta)/2)}{\sin(2\omega_e\tau)}. \quad (13)$$

With the aid of the transformation

$$L(t)=\exp(i\omega_e\mathbf{I}_yt)T\exp\left(-i\int_{+0}^{t}[f(t')\cos\theta I_{y,2}+\Delta I_{y,3}]dt'\right) \quad (14)$$

we change over to a system of coordinates with an effective field.[11] It is convenient to represent the dipole interaction in the form

$$\mathcal{H}_d^{\rm sec}=\sum_{m=-2}^{2}\mathcal{H}_d^m,\quad [\mathbf{I}_y\mathbf{n},\mathcal{H}_d^m]=m\mathcal{H}_d^m. \quad (15)$$

In the coordinate system defined by the transformations (14), the equation for the density matrix takes the form

$$d\tilde{\rho}/dt=-i[\omega_e\mathbf{I}_y+\mathcal{H}_d^0+\Phi(t)\mathcal{H}_d^1+\Phi^*(t)\mathcal{H}_d^{-1}+\chi(t)\mathcal{H}_d^2+\chi^*(t)\mathcal{H}_d^{-2}+\tilde{G}^{\rm sec}(t)+\tilde{G}'(t)+\mathcal{F},\tilde{\rho}], \quad (16)$$

where $\tilde{G}=LGL-1$. The transformation (14) was chosen such that $L(2k\tau)=1$. $\Phi(t)$ and $\chi(t)$ are periodic functions of the time with period 2τ. The solution of (16) is formally similar to that of the problem of multipulse spin locking in NMR and considered in Refs. 11 and 12. We present here only the main results.

At $\omega_e\gg\omega_{\rm loc}\approx1/T_2$ there is established in the system after a time $t\approx T_2$ the quasiequilibrium

$$\rho_{eq}=1-\alpha_{eq}\omega_e\mathbf{I}_y-\beta_{eq}\mathcal{H}_d^0+{}^1/_3\gamma_{eq}\omega_Q(1+\eta)(I_{z,3}-I_{x,3}), \quad (17)$$

where $\alpha_{\rm eq}$, $\beta_{\rm eq}$) and $\gamma_{\rm eq}$ can be obtained from (11) and from the condition that the corresponding quantities are quasiintegral, i.e., they do not chnage over times $t\approx T_2$ ($\mathrm{Tr}\rho+Q_i=\mathrm{Tr}\,\delta_{\rm eq}Q_i$, $Q_i=\omega_e\cdot\mathbf{I}_y$, $\mathcal{H}_d^0$, $I_{z,3}-I_{x,3}$):

$$\alpha_{eq}=-\alpha_0\frac{\omega_Q}{\omega_e}\left(1-\frac{\eta}{3}\right)\cos(\varphi_0\cos\theta-\arccos n_3),$$
$$\beta_{eq}\approx0,\quad \gamma_{eq}=\alpha_0. \quad (18)$$

It follows therefore that during the time $t\approx T_2$ the signal decreases [the observed signal, as can be seen from (8), is proportional to $\mathrm{Tr}\,\rho\,I_{y,2}$, i.e., to αn_2]. For example, for $\phi_0\cos\theta=\pi/2$ the magnetization decreases like $n_2{}^2$ compared with the initial one. The further evolution of the density matrix consists of slow changes of α, β, and γ.

To find the rates of these changes, we separate with the aid of a canonical transformation[12] the slowly varying part of the density matrix

$$\rho_S=U(t)\tilde{\rho}U^*(t), \quad (19)$$

$$U(t)=\exp\left(i\sum_{n=1}^{\infty}U_n(t)\right), \quad (20)$$

where the operators U_1, U_2, ... depend periodically on the time with period 2τ and are chosen such as to exclude from the equation for ρ_S the rapidly oscillating terms of order $\omega_{\rm loc}$, $\tau\omega^2{}_{\rm loc}$,.... The slow evolution of ρ_S describes thus the damping of the magnetization (i.e., the envelope of the echo signal), and the periodic transformation (20) describes the shape of the echo.

The equation for ρ_S contains only secular (relative to the $\mathbf{n}$ axis) terms and resonant multispin terms[12]:

$$\frac{d\rho_s}{dt}=-i\Big[\omega_e I_y+\mathcal{H}_d^0+\sum_{n,m}(e^{im\pi t/\tau}R_m^n+e^{-im\pi t/\tau}R_m^{-n})$$

$$+UG^{\text{сек}}U^*+UG'U^*+\mathcal{F},\rho_s\Big]. \qquad (21)$$

The term R_m^n describes a process in which n spins absorb m quanta of frequency π/τ. It becomes effective upon satisfaction of the resonance condition

$$n\omega_e=m\pi/\tau. \qquad (22)$$

For example, at $\varphi\cos\theta=\pi/2$ and $\Delta=0$ the four-spin-resonance condition is satisfied and the corresponding resonance term takes the form

$$R_1^4={}^1/_{12}\tau^2[\mathcal{H}_d^2[\mathcal{H}_d^2,\ \mathcal{H}_d^0]]. \qquad (23)$$

We have left out of (21) for simplicity small multispin secular terms which are of no importance in our problem.

The resonant processes are taken into account in analogy with saturation theory.[13] The spin-lattice relaxation can be treeated in standard fashion,[14,15] wherein G' gives the spin-lattice relaxation determined by the spectral densities of the motions at the quadrupole-resonance frequencies, while G^{sec} gives the analogous relaxation at frequencies of the order of ω_e and ω_{loc}. The contribution of G' can be expressed in terms of the probabilities W_{ik} of the relaxation transitions between the levels i and k of the quadrupole nucleus in the lab (the frequency ω_y corresponds to the pair of levels 1 and 3, and $W_{ik}=W_{ki}$). As a result we obtain for α,β, and γ the following system of equations:

$$\frac{d\alpha}{dt}=-\sum_{n,m}F_m^n\left(\alpha-\frac{\omega_m^n}{\omega_e}\beta\right)-\alpha\{n_3^2({}^1/_2W_{12}+2W_{13}+{}^1/_2W_{23})$$

$$+n_2^2(W_{12}+3W_{13}+W_{23})+T_{1\rho}^{-1}\}+\gamma n_3\frac{\omega_y'}{\omega_e}(-{}^3/_2W_{12}+W_{23})$$

$$+\alpha_0 n_3\frac{\omega_y}{\omega_e}\left\{\left({}^1/_2+{}^3/_2\frac{\omega_y'}{\omega_y}\right)W_{12}+2W_{13}+\left({}^1/_2-{}^3/_2\frac{\omega_y'}{\omega_y}\right)W_{23}\right\},$$

$$\frac{d\beta}{dt}=\sum_{n,m}F_m^n\frac{(\omega_m^n)^2}{\omega_{\text{loc}}^2}\left(\frac{\omega_e}{\omega_m^n}\alpha-\beta\right)-T_{1d}^{-1}\beta, \qquad (24)$$

$$\frac{d\gamma}{dt}=\alpha n_3\frac{\omega_e}{\omega_y'}(-{}^1/_2W_{12}+{}^1/_2W_{23})-\gamma({}^3/_2W_{12}+{}^3/_2W_{23})$$

$$+\alpha_0\left\{\left({}^3/_2+{}^1/_2\frac{\omega_y}{\omega_y'}\right)W_{12}+\left({}^3/_2-{}^1/_2\frac{\omega_y}{\omega_y'}\right)W_{23}\right\},$$

where

$$\omega_m^n=\omega_e-m\pi/n\tau, \qquad (25)$$

F_m^n are the transition rates determined by the multispin resonant processes

$$F_m^n=\frac{n^2}{\text{Sp}\,I_{y,1}^2}\int_0^\infty dt\,\{G_m^n(t)\,e^{in\omega_m^n t}+\text{c.c.}\}, \qquad (26)$$

$$G_m^n(t)=\text{Sp}\{R_m^n\exp(i\mathcal{H}_d^0 t)R_m^{-n}\exp(-i\mathcal{H}_d^0 t)\}, \qquad (27)$$

$\omega'_y=(1/3)\omega_Q(1+\eta)$, $T_{1\rho}$ is the spin-lattice relaxation time in the "rotating" coordinate frame and is determined by the spectral density of the motions at frequencies on the order of ω_e, and T_{1d} is the time of the spin-lattice relaxation of the dipole-dipole reservoir.

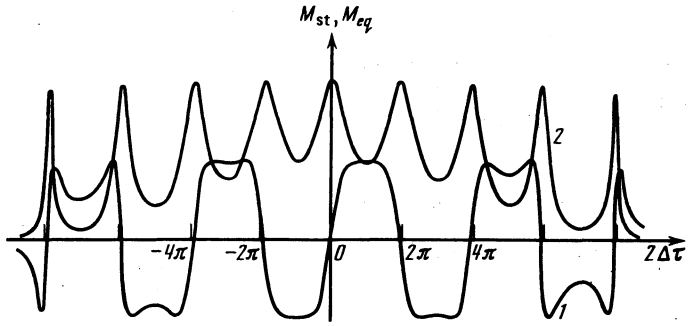

FIG. 1. Dependences of M_{st} (1) and of M_{eq} (2) on $\Delta\tau$ at $t_w/2\pi=0.2$.

The system (24) shows that the observed signal proportional to α is in the general case the sum of three exponentials and contains a dc component.

We consider first the case when spin-lattice relaxation can be neglected. Equations (24) show then that when the resonance condition (22) is satisfied and the quantity α and the magnetization proportional to it attenuate exponentially to zero. On deviation from the resonance condition the relaxation time increases and the magnetization attenuates exponentially to a certain constant value. The signal damping decrement is determined by the amplitude of the corresponding resonant term. It is proportional to τ_2 and τ_4 respectively for three- and four-spin resonant processes.

It was found in Ref. 9 that at $\varphi\cos\theta=\pi/2$ and $\Delta=0$ (the four-spin resonance condition) the damping decrement in single-crystal $NaNO_2$ is proportional to τ^4.

To obtain the signal from a polycrystal it is necessary to average over the orientations, i.e., over the angle θ.

The echo-signal envelope for a polycrystalline sample is not exponential and its characteristic damping time depends strongly on τ. At $\varphi=\pi/2$ and $\Delta=0$, for example, it is proportional to τ^{-4}.

Under these conditions, the signal from $NaNO_2$ powder was described in Ref. 1 as a sum of two exponentials with relaxation times proportional to τ^{-5}.

The spin-lattice relaxation at $n_3\neq0$ ($\Delta\neq0$) leads to the appearance of an undamped stationary signal. The stationary value of α can be obtained from (24) and from the condition $d\alpha/dt=d\beta/dt=d\gamma/dt=0$,

$$\alpha_{\text{st}}=\frac{\alpha_0 n_3}{\omega_e}\Big\{({}^1/_2\omega_y+{}^3/_2\omega_y')W_{12}+2\omega_y W_{13}+({}^1/_2\omega_y-{}^3/_2\omega_y')W_{23}$$

$$+[({}^1/_2\omega_y+{}^3/_2\omega_y')W_{12}+(-{}^1/_2\omega_y+{}^3/_2\omega_y')W_{23}]\frac{-W_{12}+W_{23}}{W_{12}+W_{23}}\Big\}$$

$$\times\Big\{\sum_{m,n}F_m^n+n_3^2({}^1/_2W_{12}+2W_{13}+{}^1/_2W_{23})$$

$$+n_2^2(W_{12}+3W_{13}+W_{23})$$

$$+T_{1\rho}^{-1}-\frac{\left(\sum_{n,m}F_m^n\omega_m^n\right)^2}{\sum_{n,m}F_m^n(\omega_m^n)^2+\omega_{\text{лок}}^2T_{1d}^{-1}}-\frac{n_3^2}{2}\frac{(W_{12}-W_{23})^2}{W_{12}+W_{23}}\Big\}^{-1}. \qquad (28)$$

This expression shows that if the relaxation is determined by a multispin process, i.e., $|F_m^n| \gg |W_{ik}|$, the stationary value of the magnetization is small and can be approximately expressed as

$$M_{st} \approx \alpha_0 \omega_0 n_2 n_3 |W_{ik}| / |F_m^n| \quad (29)$$

($M_{st} = n_2 \omega_e \alpha_{st}$). In our case, when the attenuation of the magnetization is determined by the spin-lattice relaxation, the equilibrium value of the magnetization can be comparable with the initial one, and its dependence on the parameters of the sequence can be expressed in the form

$$M_{st} \sim n_2 n_3 / (1 + b n_2^2), \quad (30)$$

where the coefficient b depends on the probabilities of the transitions that enter in (28). Expressions (29) and (39), with (13) taken into account, show that M_{st} is a periodic odd function of $\Delta\tau$ and reverses sign when the pulse phase is changed by 180°. This behavior was obtained in experiment.[9]

It should be noted that Eqs. (13) are valid at not too large values of Δ. At large values of Δ it is necessary to take into account the finite pulse duration t_w and calculate more accurately the direction of the effective field. The dependence of M_{st} on Δ then takes the form of oscillations that are damped at $\Delta \approx t_w^{-1}$. A qualitative dependence of M_{st} on $\Delta\tau$ at $t_w/2\pi = 0.2$ is shown in Fig. 1. Also shown there is the dependence of the quasiequilibrium value of the magnetization M_{eq}, observed after a time of the order of T_2, on $\Delta\tau$ at $\varphi_0 \cos\theta = \pi/2$. These dependences agree qualitatively with those obtained in Ref. 9. Some discrepancy in the behavior of M_{eq} may be due to deviation of the initial pulse from $\pi/2$.

The proposed thermodynamic theory describes thus satisfactorily the experimental data published to date.

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