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APPLICATIONS OF NEW NMR TECHNIQUES

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SUMMARY

The density matrix formalism is employed to derive the spin response to the radiofrequency field and the magnetic field gradients at MR imaging experiment. In this approach the effects of spin parameters and their application in medicine and physics of condensed matter are visualized clearly.

INTRODUCTION

Nowadays, the nuclear magnetic resonance[1-3] is the tool used for investigations in different fields of physics, chemistry, biology and medicine. With new techniques which allow us to determine the image of spatial distribution of spins and their properties inside the macroscopic sample and which were first introduced in 1952[20] and developed later[21-25], it is used for the visualizing the spatial spin distribution, the distribution of relaxation times, the chemical shifts and for the mapping of the flow velocity distribution in a fluid. It allows us to measure the spin parameters[26-29] and provides us with the important information about the structural and dynamical properties of matter and in medicine with functional assessment and physiological status of body organs.

The aim of this review is to consider systematically the parameters which reflect the effects of spin interactions and its migration on NMR signal and provide a classification of the different imaging methods. In spite of the fact that spin dynamics is usually described by modified phenomenological Bloch equations, the density matrix formalism is employed in this review. The advantage of this approach is that the expressions for response of the spins to external magnetic and radiofrequency (rf) fields are more compact and allow the effect of spin interactions in matter and spin spatial motion to be visualized more clearly.

SPIN RESPONSE

The free energy of a coil containing a sample of magnetic moment M at constant temperature produced by a change in magnetic field δB_1 is

$$\delta F = -M\,\delta B_1 \tag{1}$$

where B_1 is the magnetic field at the location of the magnetic moment. The free energy can be also expressed in terms of an electric current i_c which produces a field B_1, and a magnetic flux Φ induced by magnetic moment of the sample

$$\delta F = -\Phi\,\delta i_c \tag{2}$$

Thus the voltage induced in the coil [30] is

$$U = -\frac{d\Phi}{dt} = \frac{d}{dt}\frac{\delta F}{\delta i_c} = -\frac{d}{dt}\left(M\frac{\delta B_1}{\delta i_c}\right) \tag{3}$$

To distinguish between the magnetic fields in the receiver and transmitter coils, that of the receiver coil will be denoted as B_{1r}, while the magnetic field of the transmitter coil is given a B_{1t}. In a sample with many microscopic magnetic moments (the spins), the induced emf is given by

$$U = -\frac{\hbar\gamma}{i_c}\frac{d}{dt}\sum_j\langle B_{1r}(r_j)\,I_{yj}\rangle \tag{4}$$

where γ is the magnetogyric ratio and r_j is the radius vector of the location of jth spin, I_{yj} is the component of the spin operator along the coil axis. Expression (4) shows that if a coil carrying a unit current produces a field B_{1r} at the point of a spin then the rotating magnetic dipole of the same spin induces in the coil an emf proportional to the sum of the products between the spins and their related magnetic fields. The average can be evaluated using the density matrix operator $\rho(t)$ to give

$$U = \frac{\hbar}{i_c}\frac{d}{dt}\sum_j \mathrm{Tr}\ \rho(t)\,\omega_{1r}(r_j)\,I_{yj} \tag{5}$$

where $\omega_{1r} = \gamma B_{1r}$

By using the time evolution operator $U(t)$ the density matrix can be formally written as

$$\rho(t) = U(t)\,\rho(0)\,U^{-1}(t) \quad (6)$$

where $\rho(0)$ is the initial density matrix at the moment when spin excitation starts. In the high temperature approximation the equilibrium density matrix is

$$\rho(0) = \rho_L(1 - h\omega_o \sum_j \beta_j I_{zj}) \quad (7)$$

where the matrix operator of the lattice is ρ and where β_j describes the degree of spin longitudinal magnetization. The longitudinal magnetization of spins may be non-uniform either due to an inhomogeneous static magnetic field or due to some perturbation prior to the experiment, and thus β_j depends upon the location of spins. An example is flow measurement made by the time-of-flight method, where β_j depends upon time which the spins spend in the magnet.
The time evolution operator in the expression (6) is defined with the hamiltonian by the formal relation

$$U(t) = T\exp(-i\int_0^t H(t')\,dt') \quad (8)$$

The time ordering operator T prevents direct evaluation of eqn. (8) by simple integration of the comulant and it is necessary to employ some approximate techniques. The general objective of the spatialy resolved measuring techniques is to label the spins in some manner. In magnetic resonance this is achieved by applying a non-uniform magnetic field. If the static magnetic field B_o is inhomogeneous along the sample, then the precession angular frequency of the spins

$$\omega_o = \gamma B_o \quad (9)$$

depends upon he spin location as well as the spin magnetization. Thus the spatial displacement is shown as a frequency displacement. A similar effect can be achieved by non-uniform field of the transmitter or receiver coil. If the rf field of the transmitter coil B_{lt} is non-uniform, the degree of spin excitation depends upon the spin location and if the field B is non-uniform the receiver coil detects signals which also depends upon the spin location.
The transmitter coil generates a magnetic field

$$B_{lt} = B_{lto}(r, t)\sin\omega_o t \quad (10)$$

which excites the spins. In a selective excitation experiment where only the magnetization in a thin slice of the sample precesses in a transverse plane the magnetic field gradient is applied during the rf irradiation period. Thus only spins in a certain plane are at exact resonance and interact strongly with the rf pulse. The shape is well-defined if the rf pulse amplitude is modulated with the desired spectral distribution. Thus the magnitude of the rf excitation pulse is a function of time and position in general.

Here it is assumed that the hamiltonian describing the system, and the spin dynamics in particular, consists of five parts

$$H(t) = H_Z + H_g(t) + H_{rf}(t) + H_{int} + H_L \tag{11}$$

The Zeeman part

$$H_Z = -h\omega_0 \sum_j I_{zj} \tag{12}$$

the gradient part

$$H_g = h\nu\, G(t) \sum_j (r_j - r_0)\, I_{zj} \tag{13}$$

the radiofrequency field

$$H_{rf}(t) = -h \sum_j \omega_{1t}(r_j, t) \sin(\omega_0 t + \delta)\, I_{xj} \tag{14}$$

The hamiltonian H_{int} includes all remaining spin interactions such as the spin-spin interaction, the interactions with electrons, the quadrupole interactions and the interactions between spins and the lattice. H_L is the lattice hamiltonian itself.

In the ordinary NMR free precession experiment only the excitation rf pulse is applied. Here we shall consider a more general approach where spin excitation is achieved using a type of spin-echo rf pulse sequence consisting of a soft $\pi/2$ pulse and a sequence of π pulses of various phase. In contrast to the initial excitation pulse whose magnitude depends upon the time and the spin location, the field B_{1t} of the π rf pulses is assumed to be completely uniform and time independent. In order to simplify further consideration the π pulses are assumed to be short enough to neglect their spatial selectivity when applied simultaneously with the magnetic gradient. Here we shall adopt a treatment of the spins which uses a time evolution operator written as a product (31, 25)

$$U(t) = U_L\, U_Z\, U_{rf\pi}\, U_g\, U_{rfe} \tag{15}$$

where U_L includes only the hamiltonian H_L and H_{int}. Uz is the part due to the Zeeman interactions, $U_{rf\pi}$ is from the interactions with the π pulses, U_g is related to the gradient interaction and U_{rfe} represents the spin time evolution due to the excitation by the initial rf pulse. Each separation of the time evolution operator means the transformation in a new interaction representation.

Substitution of the expression (15) into eqn(6) gives the emf voltage as

$$U = \frac{h^2\omega_0}{i_c}\frac{d}{dt}\sum_{ji}\left\langle \mathrm{Tr}\, U_L U_Z U_{rf\pi} U_g U_{rfe} I_{zi} U_{rfe}^{-1} U_g^{-1} U_{rf\pi}^{-1} U_Z^{-1} U_L^{-1} I_{yj}\ \omega(r_j(t)) \right\rangle_L \beta_i \tag{16}$$

Here the bracket $\langle \ldots \rangle_L$ represents the average over the all other degrees of freedom except that of the spin. The purpose of the initial excitation rf pulse is to transform the spins into coherent state either in the whole volume of the sample or in a certain selected part. In order to to achieve a selective excitation, the rf pulse is applied simultaneously with the magnetic field gradient. The effect of such combined excitation on the spin system has been treated using different approaches[28]. In the following it will be expressed in general form as

$$U_{rfe}\, I_{zj}\, U_{rfe}^{-1} = A_j(t) I_{xj} + B_j(t) I_{yj} + C_j(t) I_{zj} \,. \tag{17}$$

The further evaluation with the gradient term gives

$$U_g\, U_{rfe}\, I_{zj}\, U_{rfe}^{-1}\, U_g^{-1} = M(r_j)\left[I_{xj}\cos(\varphi_j(t) + \alpha_j) + \right. \tag{18}$$
$$\left. + I_{yj}\sin(\varphi_j(t) + \alpha_j)\right] + C_j\, I_{zj}$$

with

$$M(r_j) = \sqrt{A_j^2(t) + B_j^2(t)} \tag{19}$$

and

$$\alpha_j = \tan^{-1} \frac{A_j(t)}{B_j(t)} \tag{20}$$

and where

$$\varphi_j(t) = \gamma \int G_{eff}(t')\left[r_j(t') - r_o\right] dt' \,. \tag{21}$$

Transformation into the tilted frame by $U_{rf\pi}$ changes the sign of the operator I_{zj}. It also changes the sign either of I_{xj} or I_{yj} depending upon the phase of the π rf pulses and on the number of pulses before the time of measurements. In general we denote the sign of I_{xj} as $P_x(t)$ and of I_{yj} as $P_y(t)$ at the time t. It is assumed that the system is in a strong magnetic field and that all other spin interactions but Zeeman one are small perturbation. In taking the time derivative in the expression (16) all other modulations, except the free precession with the averaged frequency ω_o have been neglected. Thus by substitution of eqn. (18) into eqn. (16) emf induced in the receiver coil becomes

$$U = \frac{h^2\omega_o^2}{i_c} P_x \sum_j \beta_j \left\langle \mathrm{Tr}\, I_{xj} I_{xj}(t)\, M[r_j(0)]\, \omega_{1r}[r_j(t)] \cos[\varphi_j(t) + \right.$$
$$\left. + \alpha_j + P_x P_y \omega_o t \,]\right\rangle_L \,. \tag{22}$$

The parameter $M(r)$ in eqn. (22) becomes independent of the spin location if there is no magnetic field gradient and when B_{1t} is uniform along the sample

$$M = \sin\left[\gamma \int_0^t B_{1t}(t')\, dt'\right] \,. \tag{23}$$

Under the same condition the phase α is zero. In the ordinary free induction decay experiment in the absence of any π rf pulses, P_x

and P_y are equal to unity, but in the spin-echo experiment which use a sequence of π rf pulses their value may be +1 or -1, leading to the change in sign of the signal and the sign of the phase. They change also the sign of the magnetic field gradient. After each π rf pulse the sign of the gradient is reversed so that effective gradient can be written

$$G_{eff}(t) = P_x(t)\, P_y(t)\, G(t) \tag{24}$$

Equation (22) represents the basic expression for the spin response in the case of the selective excitation and application of magnetic field gradients. The measuring or imaging of the spin density, the distribution of spin relaxation times, the flow, and the effects of the spin migration are determined by the parameters in the expression. Thus the parameter β_j is proportional to the longitudinal magnetization, and depends upon the spin-lattice relaxation time. It also depends upon the velocity at which the inflowing spins have insufficient time to reach the equilibrium value β_o. The parameter $Tr\, I_{xj}\, I_{xj}(t)$ describes spin-spin interactions. It modulates the free precession signal with additional frequency depending upon the internal magnetic fields created by spins neighbours. In some cases it can be described only by the relaxation time T_2 as

$$\sum_j Tr\, I_{xj} I_{xj}(t) = Tr\, I_{xj}^2 \exp\left(-\frac{t}{T_2}\right) \tag{25}$$

SELF-DIFFUSION AND FLOW

The idea of using spins for measuring molecular migration dates back to the beginning of nuclear magnetic resonance (NMR). The potential of using NMR for studying either random microscopic flow (self-diffusion)(1-3) or macroscopic flow(4-10) was quickly realized. The major advantage of using NMR for these measurements is that the sample is unaffected by the measuring process since there is no direct contact with the fluid. Thus, from the very early days of NMR flow measurements have been made on very different systems(11) which even included some physiological applications (12,13). Several methods for measuring the spatial migration of spins have been proposed and most of them have been applied successfully(14-19).

The radius vector of the migrating spins rj changes with time. An averaged spin location can be expressed simply as

$$\langle r_j(t)\rangle = r_{jo} + v_j\, t \tag{26}$$

where v_j is the averaged velocity vector of jth spin during acquisition time of the FID in the NMR experiment. In the spin response given by eqn. (22) the migration effects the receiving ability of the coil through the parameter $\omega_{1r}(r_j(t))$ and also changes the phase

$$\varphi(t) = \gamma \int_0^t G_{eff}(t')[r_j(t') - r_o]\, dt \qquad (27)$$

The detailed treatment of the effect caused by variation in ω_{1r} is given elsewhere(25, 35) but here only the effects of the phase changes caused by motion of the spins through the magnetic field gradient will be considered. Neglecting the rf non-uniformity then the average only of the phase in eqn.(22) needs to be evaluated. This can be done using the comulant expansion theorem[32]. If only the first two terms of the expansion are taken into account it is transformed into

$$\langle \cos[\varphi_j(t) + \alpha_j] \rangle_L = \exp[-\xi_j(t)]\cos[\langle \varphi_j(t)\rangle_L + \alpha_j] \qquad (28)$$

where

$$\xi_j(t) = \frac{\gamma^2}{2}\int_0^t dt_1 \int_0^t dt_2\, G_{eff}(t_1)\langle r_j(t_1)\, r_j(t_2)\rangle_{LC} G_{eff}(t_2) \qquad (29)$$

with

$$\langle r_j r_j \rangle_{LC} = \langle r_j r_j \rangle_L - \langle r_j \rangle_L \langle r_j \rangle_L \qquad (30)$$

and

$$\langle \varphi_j(t)\rangle_L = (r_{jo} - r_o)\, F(t) + v_j\, f(t) \qquad (31)$$

with

$$F(t) = \gamma \int_0^t G_{eff}(t')\, dt', \qquad (32)$$

and

$$f(t) = \gamma \int_0^t t'\, G_{eff}(t')\, dt' \qquad (33)$$

The expression (28) represents an attenuated oscillation, with the attenuation determined by the parameter $\xi(t)$, which depends upon the spin coordinates at different times and is related to the randomization of spins caused by migration since it arises from turbulent flow or microscopic self-diffusion. Applying a gradient along the x-axis, $\xi(t)$ becomes

$$\xi_j(t) = \frac{\gamma^2}{2}\int_0^t dt_1 \int_0^t dt_2\, G_{xeff}(t_1)\, G_{xeff}(t_2)\, \langle x_j(t_1)\, x_j(t_2)\rangle_{LC} \qquad (34)$$

where location correlation can be expressed by the velocity autocorrelation function

$$\langle x_j(t_1)\, x_j(t_2)\rangle_{LC} = \frac{1}{\pi}\int_{-\infty}^{\infty} D_{xxj}(\omega)\, \frac{\exp[i\omega(t_1 - t_2)]}{\omega^2}\, d\omega \qquad (35)$$

where $D_{xxj}(\omega)$ is the spectrum of the x-component of the velocity correlation:

$$D_{xxj}(\omega) = \int_0^{\infty} \langle v_{xj}(0)\, v_{xj}(t) \rangle_L \exp(i\omega t)\, dt. \qquad (36)$$

$D_{xx}(0)$, the value at $\omega = 0$, is identical to the self-diffusion coefficient, D. If the spectrum of the magnetic field gradient is defined as

$$G(\omega, t) = \int_0^{t} G_{xeff}(t') \exp(i\omega t')\, dt' , \qquad (37)$$

then by substitution of eqns.(35)-(37) into eqn.(34) the attenuation parameter becomes

$$\xi_j(t) = \frac{\gamma^2}{2\pi} \int_{-\infty}^{\infty} \frac{D_{xxj}(\omega)}{\omega^2} | G(\omega, t) |^2 d\omega \qquad (38)$$

This is the convolution between the spectrum of the velocity autocorrelations and the spectrum of the effective gradient. Whenever the correlation time of the molecular translation motion is short compared to the time scale of the effective gradient changes only the low frequency part of $D_{xx}(\omega)$ plays a role in eqn.(38). In this case $\xi(t)$ can be approximated to

$$\xi_j(t) = \frac{\gamma^2}{2\pi} D_{xxj}(0) \int_{-\infty}^{\infty} \frac{| G(\omega, t)|^2}{\omega^2} d\omega , \qquad (39)$$

which is equivalent to the well-known result derived by Torrey[3] for spin echo-dampening if the Parceval identity is taken into account.

$$\xi_j(t) = \gamma^2 D_{xxj}(0) \int_0^{t} \left| \int_0^{u} G_{xeff}(t')\, dt' \right|^2 du \qquad (40)$$

This means that eqn. (40) is just an approximation of the more general expression (38) which valid for any correlation times. With a certain reinterpretations it can be applied when the dynamics of turbulent flow is considered. Thus this consideration gives that the measurements of self-diffusion by NMR might yield information not only about the diffusion constant but also about the velocity autocorrelations, i.e. its spectrum.

CONCLUSION

By replacing eqns (25) and (28) in eqn. (22) the emf of the receiver coil is

$$U = \frac{h^2\omega_o^2}{i_c} P_x \sum_j \beta_j M(r_{jo}) \,Tr\, I_{xj}^2 \exp\left[-\frac{t}{T_{2j}} - \xi_j(t)\right] \cdot$$

$$\cdot\, \omega_{1r}(r_{jo} + v_j t) \cos\left[F(t)(r_{jo} - r_O) - f(t)\, v_j + \alpha_j + P_x P_y \omega_o t\right] . \quad (41)$$

which can be written in the continuum limit as

$$U = \frac{h^2\omega_o^2}{i_c} P_x \,Tr\, I_x^2 \int dr^3 \rho(r)\ \beta(r)\ M(r) \exp\left[-\frac{t}{T_2(r)} - \xi(r, t)\right] \cdot$$

$$\cdot\, \omega_{1r}\left[r + v(r)t\right] \cos\left[F(t)(r - r_O) - f(t)\, v(r) + \alpha(r) + P_x P_y \omega_o t\right] . \quad (42)$$

Here the spins density is defined as

$$\rho(r) = \sum_j \delta(r - r_{jo}) . \quad (43)$$

and $Tr\, I_x^2$ means the trace over one spin.
In eqns (41) and (42) B_{1t} of the π rf pulses is assumed to be uniform in the excited part of the sample. This means that the active region of the transmitter coil extend the width of the selected slice, and spin migration should be slow enough to avoid an appreciable outflow of spins from the coil in the time acquisition of the FID. These restrictions are related only to the π rf refocusing pulses, the magnitude of the initial rf pulse may be non-uniform along the sample. A detailed analysis of the case of spin refocusing by non-uniform π rf pulses gives a spin response which is much more complicated than eqn. (42). Such a case has been already treated[25] by considering a spin-echo experiment on a flow in liquid. However, with the restriction discussed above, eqn. (41) is a general description of the spin response of the flowing fluid. There are various parameters dependent upon the flow velocity which can play the dominant role in any particular chosen experimental procedure. In the NMR coherent FID experiment the emf signal given by eqn.(42) is converted by quadrature detection into two signals U_{d1} and U_{d2} which can be written in complex form as

$$U = U_{d1} + U_{d2} = K \int dr^3 \rho_{eff}(r,v,t) \exp\left\{i\left[F(t)(r_O - r) - f(t)\, v(r) + \alpha(r)\right]\right\} \quad (44)$$

with

$$K = \frac{h^2\omega_o^2}{i_c} P_x \,Tr\, I_x^2 \quad (45)$$

and

$$\rho_{eff}(r, v, t) = \omega_{1r}\left[r + v(r)t\right] \rho(r)\ \beta(r)\ M(r) \exp\left[-\frac{t}{T_2(r)} - \xi(r, t)\right] . \quad (46)$$

This expression is applicable to various imaging schemes and gives us, after Fourier transformation, information about the distribution of spin density, the distribution of relaxation times T_1 and T_2 and also information about random flow $\xi(t)$ or velocity distribution of the flow. The velocity distribution can be obtained either from incomplete magnetization recovery and transfer magnetization from β, the effects due to the rf field non-uniformity seen in ω_{1r} and $M(r)$, or from the phase changes $\exp(i\phi)$. These particular cases have been considered in detail elsewhere[35, 25].

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