Dependence of the spin-echo signal on the phases of input pulses

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The statistical tensor method is used to develop a theory of the dependences of the intensity and phase of an echo signal on the phases of the input pulses. General relationships are obtained between the components of a statistical tensor before and after the passage of a pulse. An analysis is made of the formation of an echo signal allowing for the scatter of local fields which alter the statistical tensor. It is shown that the appearance of a signal independent of the phase of the second pulse is associated with the quadrupole inhomogeneous broadening mechanism. The adopted method also predicted a characteristic feature of the quadrupole inhomogeneous broadening mechanism involving a change in the statistical tensor rank. The possibility of detecting this feature is discussed. Some properties of the "forbidden" quadrupole echo signals are discussed.

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INTRODUCTION

It has been thought that the dependences of the intensity and phase of a spin echo signal on the phases of the input pulses are well understood. However, the case when one or both pulses are acoustic has been attracting special attention because it is necessary to allow not only for the initial phase but also for the phase $\mathbf{k}\cdot\mathbf{r}$, where $\mathbf{k}$ is the wave vector of the acoustic vibrations and $\mathbf{r}$ is the radius vector of the nucleus under consideration. Averaging over the volume of a sample reduces the echo signal by a factor of at least λ/l, where λ is the wavelength and l is the length of the sample. Therefore, several pulse combinations with mutually compensating phases have been suggested. [1,2] The first observations of a nuclear spin echo signal, excited by a combination of electromagnetic (M) and acoustic (A) pulses, were reported recently. [3,4] It was found that the intensity and phase of the echo signal were completely independent of the phase of the A pulse. The explanation was based on the fact that the formation of the observed echo signal was due to the quadrupole inhomogeneous broadening mechanism. In all these investigations the calculations were carried out for nuclei with certain specific values of the spin. As shown below, a number of inaccuracies was committed or use was made of additional ideas not related to the dependences of the echo on the input pulse phases.

Our aim is to investigate, for arbitrary spin, the dependences of the A and M pulses on the phases and to identify the role of the inhomogeneous broadening mechanism. We shall develop a theory of the spin echo using the method of statistical tensors. We shall show that this method is particularly convenient in determining the dependence of the echo signal on the input pulse phases. The adoption of a coordinate system rotating with the initial phase makes it possible to obtain general relationships between the statistical tensor components R_q^l before and after the passage of a pulse. These relationships are valid for any spin and any type of a periodically varying classical field on condition that the spin system has an equidistant spectrum. Such relationships depend only on the change in the magnetic quantum number Δm governed by the nature of the input pulse. The statistical tensor method allows us also to consider, from a general point of view, the formation of an echo signal allowing for the scatter of the local fields which alter the components. We shall show that the appearance of a signal independent of the phase of the second pulse is entirely due to the quadrupole inhomogeneous broadening mechanism and is independent of the nature of the external field. The method predicts also another characteristic feature of the quadrupole inhomogeneous broadening mechanism, which involves a change in the rank of the statistical tensor. Such a change gives rise to precessing magnetization components also in those cases in which this has been regarded (so far) as impossible. [1] Moreover, we shall consider the possibility of detection of electromagnetic signals due to a change in the statistical tensor rank.

1. PHASE RELATIONSHIPS IN A SPIN SYSTEM AFTER ONE INPUT PULSE

In the most general case the interaction of a nucleus with an external classical field is described by the Hamiltonian

$$\mathscr{H}=\sum_q (-1)^q T_q^l V_{-q}^l, \tag{1.1}$$

where T^l and V^l are spherical irreducible tensor operators of rank $l\leqslant 2I$, representing respecitvely a nucleus of spin I and an external classical field. Let us assume that the spin system is described by the density matrix $\rho(t)$, related to the statistical operators R^l of rank l and to the components R_q^l by[5]

$$R_q^l=\sum_{m,m'} (-1)^{I-m}(2l+1)^{1/2}\begin{pmatrix} I & l & I \\ -m' & q & m \end{pmatrix}\langle Im'|\rho|Im\rangle, \tag{1.2}$$

where (:::) are the $3j$ symbols.

If we use the expansion (1.2) and the commutation relationships between two irreducible tensor operators T_q^l and $T_{q'}^{l'}$, [6] the equations of motion for the density matrix yield the following system of equations for R_q^l:

$$\frac{dR_q^l}{dt}=\frac{i}{\hbar}\sum_{l',l'',q',q''}(-1)^{q+q'+q''}C^{ll'l''}_{q-q'q''}V^{l'}_{-q'}R^{l''}_{q''},\quad (1.3)$$

where the structure constants are

$$C^{ll'l''}_{qq'q''}=(-1)^{2I-q''}[(-1)^{l+l'+l''}-1][(2l+1)(2l'+1)(2l''+1)]^{1/2}\times\begin{Bmatrix} l & l' & l'' \\ I & I & I\end{Bmatrix}\begin{pmatrix} l & l' & l'' \\ q & q' & -q''\end{pmatrix}.\quad (1.4)$$

Here, $\{:::\}$ are the $6j$ symbols.

We shall first consider the interaction of a nucleus in a static magnetic field H with a rotating rf field of frequency ω, amplitude ω_1, and initial phase φ. The action of such a M pulse is described by the Hamiltonian

$$\mathscr{H}=\hbar\omega_n I_z+\frac{\hbar\omega_1}{2}(I_+\exp[-i(\omega t+\varphi)]+I_-\exp[i(\omega t+\varphi)]),\quad (1.5)$$

where ω_n is the Zeeman frequency. According to Eq. (1.1), the expression (1.5) contains only irreducible tensor operators of the first rank. It then follows from Eqs. (1.3) and (1.4) that

$$\frac{dR_q^l}{dt}=-iq\omega_n-\frac{i\omega_1}{2}[l(l+1)-q(q-1)]^{1/2}R^l_{q-1}\exp[-i(\omega t+\varphi)]-\frac{i\omega_1}{2}[l(l+1)-q(q+1)]^{1/2}R_{q+1}\exp[i(\omega t+\varphi)].\quad (1.6)$$

We shall introduce new statistical tensors P_q^l using the condition

$$P_q^l=\exp\{-iq(\omega t+\varphi)\}R_q^l.\quad (1.7)$$

The transformation (1.7) represents conversion to a coordinate system rotating with the initial phase φ. The system (1.6) reduces to

$$\frac{dP_q^l}{dt}=-iq(\omega_n-\omega)P_q^l-\frac{i\omega_1}{2}[l(l+1)-q(q-1)]^{1/2}P^l_{q-1}-\frac{i\omega_1}{2}[l(l+1)-q(q+1)]^{1/2}P^l_{q+1},\quad (1.8)$$

which is easily solved[7] giving

$$P_{q_1}^l(t)=F^{ll}_{q_1q}(\omega,\omega_1,t)P_q^l(0),\quad (1.9)$$

$$F^l_{q_1q}(\omega,\omega_1,t)=\sum_{q'',q}d^l_{q_1q''}(\alpha)\exp[-iq''\omega_{eff}t]d^{-1l}_{q''q}(\alpha)P_q^l(0).\quad (1.10)$$

Here, $d^l_{qq'}(\alpha)$ are the rotation matrices, $\omega_{eff}=[(\Delta\omega^2)+\omega_1^2]^{1/2}$, $\Delta\omega=\omega_n-\omega$, $\tan\alpha=\omega_1/\omega$.

If we now return to the laboratory coordinate system by the inverse of the transformation (1.7), we obtain the following dependence of the M-pulse phase:

$$R^l_{q'}(t)=\exp\{-i(q-q')\varphi\}F^{ll}_{q'q}(\omega,\omega_1,t)\exp(-iq'\omega t)R_q^l(0).\quad (1.11)$$

If the input pulse has a randomly varying phase φ, we have to average Eq. (1.11). Then, only the components $R^{l'}_{q'}(t)$ with $q'=q$ do not vanish. If the initial state is described by a set of tensors $R^l_0(0)$ associated only with the diagonal elements of the density matrix, the application of a noncoherent pulse does not produce $R^l_q(t)$ with $q\neq 0$. Hence, it follows that the transverse components of the magnetic moment vanish. Similar results are obtained if the spin system is subjected to an A pulse producing transitions which alter the magnetic quantum number by $\Delta m=\pm 1$. However, in contrast to a M pulse, the ranks of the tensors R^l_q before and after the passage of the pulse may be different.

We shall now consider an acoustic pulse causing transitions with $\Delta m=\pm 2$. The interaction Hamiltonian is

$$\mathscr{H}_A=\hbar\omega_n I_z+a[I_+^2\exp\{-i(\omega t+\psi)\}+I_-^2\exp\{i(\omega t+\psi)\}],\quad (1.12)$$

where $\psi=\mathbf{k}\cdot\mathbf{r}_j+\varphi$, $\mathbf{k}$ is the wave vector of sound, $\mathbf{r}$ is the radius vector of a spin j, φ is the initial phase of the A pulse, and $|a|$ is a constant governed by the elastic properties of the investigated crystal and by the direction of the acoustic vibrations relative to the magnetic field.

Formulas (1.3) and (1.4) give the following system of coupled equations

$$\frac{dR_q^l}{dt}=-iq\omega_n R_q^l+AR^{l+1}_{q\pm2}\exp[\mp i(\omega t+\psi)]+A'R^{l-1}_{q\pm2}\exp[\mp i(\omega t+\psi)],\quad (1.13)$$

where the coefficients A and A' are expressed in terms of the structure constants, $6j$ symbols, and $3j$ symbols. The explicit form of these coefficients is unimportant because the phase dependence of interest to us can be obtained in a general form. The substitution

$$P_q^l=\exp\left[-iq\left(\frac{\omega t+\psi}{2}\right)\right]R_q^l\quad (1.14)$$

transforms the system (1.12) to a system of linear differential equations with constant coefficients:

$$\frac{dP_q^l}{dt}=-iq(\omega_n-\omega/2)P_q^l+AP^{l+1}_{q\pm2}+A'P^{l-1}_{q\pm2}.\quad (1.15)$$

The general solution of this system is analogous to (1.9) but with a somewhat different function $\Phi^{l'l}_{q'q}(\omega,\omega_1,t)$:

$$P^{l'}_{q'}(t)=\Phi^{l'l}_{q'q}(\omega,\omega_1,t)P_q^l(0).$$

The application, to this solution, of the transformation which is the inverse of (1.14) gives the following relationship between the statistical tensors before and after the passage of a pulse:

$$R^{l'}_{q'}(t)=\exp\left\{-i(q-q')\frac{\psi}{2}\right\}\Phi^{l'}_{q'q}(\omega,\omega_1,t)\exp(-iq'\omega t)R_q^l(0).\quad (1.16)$$

Formulas (1.11) and (1.16) are easily generalized to the case of any periodic perturbations characterized by irreducible tensors of rank l and causing transitions with $\Delta m=p$. We still have Eq. (1.16), but now $\psi/2$ should be replaced with ψ/p. In this way we obtain the following results: in the case of an equidistant spectrum the phase of an input pulse affects all the components of the statistical tensors with the exception of those for which $q'=q$. The rank l can change in an arbitrary manner. It follows from the relationship (1.2) that the average values of the irreducible tensor operators are $\overline{T}^l_q\propto R^l_q$. Therefore, there is no dependence on the phase not only in the case of sequences such as $\langle I_\pm(0)\rangle\to\langle I_\pm(t)\rangle$, but also such as, for example, $\langle I_\pm(0)\rangle\to\langle(I_\pm I_z+I_zI_\pm)(t)\rangle$. The existence of such a phase dependence is particularly important in the case of A pulses because ψ depends on the radius vector of the nucleus under consideration and we have to average over the whole sample, which reduces greatly the average spin operators. It also follows from the relationship (1.16) that the assumption of many-quantum nature of the interaction between the spin system and a A pulse, which is made by Golenishchev-Kutuzov *et al.*,[4] is not related to the dependence of the echo signal on the input pulse phase.

In solving the system (1.3) we have to use not only Eqs. (1.11) and (1.16) but also allow for the initial conditions. In the high-temperature approximation ($g\beta H \ll kT$, where g denotes the nuclear g factor), we obtain the following initial conditions from Eq. (1.2): $R_0^1(0) \neq 0$, $R_0^l(0) \equiv 0$ for all values of $l>1$. It follows from the system (1.6) that an rf field does not alter the rank l. Then, for the stated initial conditions we obtain the nonzero components $R_0^1(t)$ and $R_{\pm 1}^1(t)$. For the A pulses causing transitions with $\Delta m=\pm 1$, the coupled system of equations relates statistical tensors of different ranks l. In contrast to the case of a M pulse, the same initial conditions give $R_{\pm 1}^2(\pi_1)$, $R_{\pm 2}^3(\pi_1), \ldots, R_{\pm|2I-1|}^{2I}(\pi_1)$, where π_1 is the duration of action of an A pulse. For A pulses with $\Delta m=\pm 2$, we obtain the following set of the statistical tensors: $R_0^2(\pi_1)$, $R_{\pm 2}^2(\pi_1), \ldots, R_0^{2I}(\pi_1)$, $R_{|2I|}^{2I}(\pi_1)$. However, the dependence of the phases is given by Eqs. (1.11) and (1.16). Before investigating the dependences of an echo signal on the input pulse phases, we shall consider the formation of this signal employing the statistical tensor concept.

2. FORMATION OF AN ECHO SIGNAL

Let us assume that after the action of a first pulse of duration π_1 a system of nuclei is described by the statistical tensors $R_{q1}^{l1}(\pi_1)$. An echo signal after the action of two pulses separated by an interval τ_{12} is obtained if allowance is made for the deviation $\Delta\omega$ of the resonance frequency ω_0 from its average value ω. As usual, let us assume that the scatter of $\Delta\omega$ obeys the normal law $\exp[-(\Delta\omega)^2T_2^{*2}/2]$ of width $1/T_2^*$. We shall postulate that these deviations are due to local fields. The influence of a static local field after the action of input pulses can be described by the perturbation factor

$$G_{q_1q_2}^{l_1l_2}(t)=\sum_{m_a,m_b}(-1)^{2I+m_a+m_b}[(2l_2+1)(2l_1+1)]^{1/2} \times\begin{pmatrix} I & I & l_1\\ m_a & -m_a' & q_1\end{pmatrix}\begin{pmatrix} I & I & l_2\\ m_b & -m_b' & q_2\end{pmatrix}\langle m_b|\Lambda(t)|m_a\rangle\langle m_b'|\Lambda(t)|m_a'\rangle^*, \quad (2.1)$$

where $\langle m_b|\Lambda(t)|m_a\rangle$ are the matrix elements of the evolution operator $\Lambda(t)$. Usually, deviations from the resonance frequencies are described in the first approximation[8] by the Hamiltonian $\mathscr{H}_1$, which commutes with the Zeeman interaction Hamiltonian. In this case the perturbation factors simplify to

$$G_{qq}^{l_1l_2}(t)=[(2l_1+1)(2l_2+1)]^{1/2}\sum_m\begin{pmatrix} I & I & l_1\\ m' & -m & q\end{pmatrix} \times\begin{pmatrix} I & I & l_2\\ m' & -m & q\end{pmatrix}\exp\left[-\frac{it}{\hbar}(E_m-E_{m'})\right]. \quad (2.2)$$

Here, E_m are the eigenvalues of the operator $\mathscr{H}_1$. Up to the moment τ_{12} from the application of the first pulse the spin system is described by the following statistical tensors:

$$R_{q_1}^{l_1}(\pi_1,\tau_{12})=G_{q_1q}^{l_1l}(\tau_{12})R_q^l(\pi_1). \quad (2.3)$$

In the case of the dipole inhomogeneous broadening mechanism the deviation from the resonance frequency is governed by the Hamiltonian $\mathscr{H}_1=-\hbar\Delta\omega I_z$ and the perturbation factor assumes, in accordance with Eq. (2.2), the form

$$G_{qq}^{ll}(t)=\exp[-iq\Delta\omega t]. \quad (2.4)$$

so that up to the moment τ_{12} when the second pulse arrives, we have

$$R_{\pm 1}^1(\pi_1,\tau_{12})=R_{\pm 1}^1(\pi_1)\exp[\mp i\Delta\omega\tau_{12}]. \quad (2.5)$$

The action of the second pulse of duration π_2 is described by Eq. (1.10) but $R_0^1(0)$ should be replaced with the expression for $R_{\pm 1}^1(\pi_1,\tau_{12})$. After two pulses separated by an interval τ_{12} between them at a time t from the first pulse the spin system is described by the following statistical tensors:

$$R_{q_1}^{l_1}(\pi_1,\tau_{12},\pi_2,t)=G_{q_1q_1}^{l_1l_1}(t-\tau_{12})R_{q_1}^{l_1}(\pi_1,\tau_{12},\pi_2). \quad (2.6)$$

The observed echo signal is governed by the average value of $\langle I_x\rangle$, which—in accordance with Eq. (1.2)—is $\langle I_x(\pi_1,\tau_{12},\pi_2,t)\rangle\propto(R_1^1+R_{-1}^1)$. Substituting in Eq. (2.6) the values of R_q^l obtained by means of Eqs. (2.4) and (2.3), we find that the final expression for $\langle I_x\rangle$ is a sum of terms multiplied by the following exponential functions:

$$\exp[-i\Delta\omega t],\quad \exp[-i\Delta\omega(t-\tau_{12})],\quad \exp[-i\Delta\omega(t-2\tau_{12})]. \quad (2.7)$$

Averaging over a normal distribution gives, respectively,

$$\exp\left[-\frac{t^2}{2T_2^*}\right],\quad \exp\left[-\frac{(t-\tau_{12})^2}{2T_2^*}\right],\quad \exp\left[-\frac{(t-2\tau_{12})^2}{2T_2^*}\right]. \quad (2.8)$$

The last term represents the echo signal at the moment $t=2\tau_{12}$.

3. DEPENDENCE OF AN ECHO SIGNAL ON THE PHASE OF THE SECOND PULSE

We shall define the phase φ of the second pulse as the difference between the phases of the second and first pulses. This can be done always by redefining the initial phase.

It follows from the results of the preceding section that only a sequence of statistical tensors

$$R_0^1(0)\to R_{\pm 1}^1(\pi_1)\to R_{\mp 1}^1(\pi_1,\pi_2,\tau_{12}) \quad (3.1)$$

gives a spin echo signal at a moment $t=2\tau_{12}$ in the case of the dipole inhomogeneous broadening mechanism. Then, according to Eq. (1.11), a change in the phase of the second M pulse by φ relative to the first alters the echo phase by 2φ in agreement with the well-known result.[9] However, M and A pulses produce also a second sequence of statistical tensors:

$$R_0^1(0)\to R_{\pm 1}^1(\pi_1)\to R_{\pm 1}^1(\pi_1,\tau_{12},\pi_2). \quad (3.2)$$

The components of the tensors $R_{\pm 1}^1(\pi_1)$ and $R_{\pm 1}^1(\pi_1,\tau_{12},\pi_2)$ are identical and, consequently, there is no dependence on the phase of the second pulse. Substituting the expressions (2.4) for the perturbation factors allowing for the dipole inhomogeneous broadening, we find that the sequence (3. 2) does not produce an echo signal. The situation is different in the case of the quadrupole inhomogeneous broadening mechanism, which—in the first approximation—can be described by the Hamiltonian[10]

$$\mathscr{H}_1=\hbar\Delta\omega[I_z^2-{}^1/_3 I(I+1)]. \qquad (3.3)$$

Using Eqs. (2. 2) and (2. 3), we then obtain

$$R_{q_1}^{l_1}(\tau_{12},\pi_1)=[(2l_1+1)(2l_2+1)]^{1/2}\sum_{m=-I}^{I}\begin{pmatrix} I & I & l_1 \\ m & -(m+q) & q\end{pmatrix}$$

$$\times\begin{pmatrix} I & I & l \\ m & -(m+q) & q\end{pmatrix}\exp[-iq\tau_{12}(2m+1)\Delta\omega]R_q^l(\pi_1). \qquad (3.4)$$

A similar expression describing the action of the second pulse readily shows that a sequence of the (3. 1) type again produces an echo signal at a moment $t=2\tau_{12}$ and this signal depends on the phase of the second pulse. However, in contrast to the situation when the dipole broadening mechanism predominates, Eq. (3. 4) for the perturbation factor now describes a number of new properties: 1) the same value of R_q^l corresponds to a set of frequencies $\Delta\omega_m=(2m+1)q\Delta\omega$; 2) for all values of $p \leqslant |I-1|$, we have $\Delta\omega_p=-\Delta\omega_{-p-1}$; 3) the rank of the tensors R_q^l may change. After the second pulse transforming $R_q^l(\pi_1, \tau_{12})$ into $R_q^{l_1}(\pi_1, \tau_{12}, \pi_2)$, the perturbation factor again contains terms of the (3. 4) type, but instead of the time interval τ_{12} we now have $t-\tau_{12}$. The first of these properties produces a series of quadrupole echo signals. From the second property it follows that, in particular, the echo signal appears at $t=2\tau_{12}$ also for the sequence (3. 2) independent of the phase of the second pulse. These results apply to M and A pulses and they demonstrate that the echo signal has two components: one depends on the phase of the second pulse and the other is independent of this phase. In the special case of $I=\frac{5}{2}$ this is in agreement with the conclusions reached by Alekseev *et al.*[2] However, in addition to this conclusion, Alekseev *et al.*[2] assert that for $I=2$ and the dipole inhomogeneous broadening mechanism, we can have an echo signal independent of the second-pulse phase, which is incorrect.

The property 3) may be important in sequences of two or more acoustic pulses. For example, it has been stated[1] that two identical A pulses produce no precessing components of the magnetic moment if the spin is $I=1$. In fact, a general analysis of the solutions of the system (1. 6) made at the end of the first section of the present paper seems to confirm this conclusion for arbitrary spin. However, in view of the quadrupole inhomogeneous broadening mechanism which alters the rank of R_q^l, we can have the following situation for a sequence of two A pulses with $\Delta m=\pm 1$:

$$R_0^1(0)\to R_{\pm(2p-1)}^{2p}(\pi_2)\to R_{\pm(2p-1)}^{2p-1}(\pi_1,\tau_{12})\to R_{\pm 1}^1(\pi_1,\tau_{12},\pi_2). \qquad (3.5)$$

The average values of $\langle I_x\rangle$ do not vanish only because of the presence of the perturbation terms $G_{kk}^{p(p-1)}(\tau_{12})$. However, there are also additional phase factors containing $\mathbf{k}\cdot\mathbf{r}_j$, which weaken the echo signal. If a third acoustic pulse with $\Delta m=\pm 2$ is applied, the phase shift is compensated in accordance with Eqs. (1. 11) and (1. 16) and the signal intensity rises again.

Let us consider the case when two acoustic pulses generate an electromagnetic echo signal. Difficulties associated with the generation of sound at two frequencies by means of the same piezoacoustic transducer can be avoided by applying A pulses with $\Delta m=\pm 1$ to one side of a crystal and A pulses at twice the frequency to the opposite side. According to Eq. (1. 16), in this case the phases of the A pulses become compensated and the generation of an electromagnetic signal is possible because of a change in the rank of R_q^l by analogy to Eq. (3. 5).

We shall conclude by considering certain features of the "forbidden" quadrupole echo signals.[7] So far, we have ignored the quadrupole splitting $\Delta\omega_Q$ during a pulse and allowed for its action only in the interval between the pulses. However, if $\Delta\omega_Q\sim\omega_1$, it is necessary to allow for such splitting also during a pulse. The system (1. 8) now acquires terms $-i\Delta\omega_Q R_q^{l\pm 1}$ on the right and, therefore, we find that even for M pulses—in contrast to the sequences (3. 1) and (3. 2) when there is no phase dependence or the echo signal depends on φ—the "forbidden" echo signals may be governed by different phase dependences on $\varphi, 3\varphi, 4\varphi, \ldots, |2I+1|\varphi$.

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