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Spin Echoes after Arbitrary *N* Pulses

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Based on the Bloch equations, the general response of a nuclear spin-$\frac{1}{2}$ system to multiple RF pulses with arbitrary phases and flip angles is presented. The general solution is expressed in a form including the pathway vectors specifying the magnetization paths generated by a string of pulses. The pathway vectors are useful for predicting the exact positions, magnitudes, and number of echoes. Also, these vectors make it easy to trace the physical origin of an echo formation. It is shown that the maximum number of echoes after *N* pulses is $(3^{N-1} - 1)/2$ using this concept. © 1997 Academic Press

INTRODUCTION

Pulse NMR techniques have been developed with a lot of discussions on echo signals with multiple pulses, and the discussion still continues. Spin echoes with multiple pulses have been analyzed independently in each application depending on the rotation angle of the nuclear moment, the direction of the rotation axis, the number of pulses, and whether the individual pulses are the same or not. In this work, we present the general solution for the response of a nuclear spin-$\frac{1}{2}$ system to arbitrary multiple pulses and show that the information on echoes such as the positions, magnitudes, and number is obtained simply from the general solution.

Generally, it is easy to analyze spin echoes with multiple RF pulses by the simple phenomenological Bloch equations. The response of a spin system obtained by solving the Bloch equations is described by either the magnetization components in the Cartesian coordinates (*1, 2*) or the rotating magnetization components (*3–5*). The former is intuitive while the latter gives simple mathematical expressions. The rotating magnetization components are appropriate for describing the response of a nuclear spin system to multiple pulses because each component evolves time independently in the absence of RF pulses and the nuclear magnetization rotation matrix due to an RF pulse is simple. The recent work studying the response of a nuclear spin system using the rotating magnetization components is the partition formalism of Kaiser *et al.* (*5–7*). They described the response of a spin system to repetitive RF pulses that rotated the nuclear magnetization by an arbitrary angle in a particular axis (*x* axis) by introducing the echo path concept. But there are many cases where the rotation axis is not fixed in a particular direction. In the CPMG method (*8*) or the phase-cycling procedures (*9, 10*) devised to correct the schematic errors generated during data acquisition by the incompleteness of an instrument, RF pulses of various phase angles are required in the *x–y* plane. Besides, the rotation axis is not even in the *x–y* plane for the nuclei which are off resonance. Therefore, the application of the analyses of Kaiser *et al.* is restricted.

In this work, we present the general solution of the Bloch equations for pulse sequences, where each pulse has arbitrary phase and flip angle, using the magnetization pathway vectors. A magnetization path can be expressed by an array of states which the nuclear magnetic moment experiences after each pulse. We defined "pathway vectors" to represent the pathways. This pathway vector is similar to the coherence-transfer pathway vector introduced by Bodenhausen *et al.* in the analysis of multiple-quantum-coherence signals (*11*). While Bodenhausen's vector takes the change of the coherence order due to an RF pulse as a vector element, our pathway vector consists of the coherence order itself which is more useful for describing the formation of echoes.

In the next section, the general response of a nuclear spin system to an arbitrary pulse sequence is obtained by introducing the pathway vectors. The procedure for estimating the magnitudes and positions of multiple echoes for arbitrary multiple pulses from this general solution is given in the following section. The explicit solution of the well-known three-pulse case is analyzed as an example of this procedure. In the last section, the maximum number of echoes formed after *N* pulses is obtained, and it is explained why our result differs from the previous report (*2*).

THE RESPONSE OF A NUCLEAR SPIN SYSTEM TO MULTIPLE PULSES

The motion of nuclear magnetization in a static field B_0 along the *z* axis and a pulsed RF field B_1 in the *x–y* plane is well described by the phenomenological Bloch equations as

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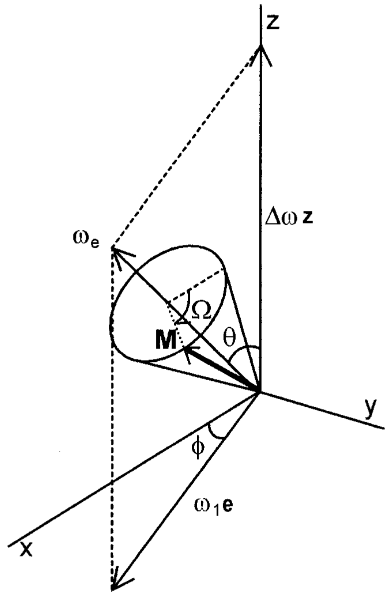

FIG. 1. The orientation of the rotation axis of an RF pulse field in the rotating frame. ω_e represents the effective field, and the other quantities are defined in the text.

$$\frac{d\mathbf{M}}{dt} = \mathbf{M} \times (\Delta\omega\mathbf{z} + \omega_1\mathbf{e}) - \frac{1}{T_2}(M_x\mathbf{x} + M_y\mathbf{y}) + \frac{1}{T_1}(M_{eq} - M_z)\mathbf{z}, \quad [1]$$

where **M** is the magnetization vector; **x**, **y**, and **z** are unit vectors in the frame rotating at an angular frequency ω; and **e** is the unit vector in the direction of the pulsed RF field in the rotating frame (Fig. 1). M_{eq} is the equilibrium magnetization and T_1 and T_2 are the spin–lattice and spin–spin relaxation times, respectively. The offset frequency $\Delta\omega$ is the difference between the resonance frequency, $\omega_0 = \gamma B_0$, and the RF frequency ω, and $\omega_1 = \gamma B_1$, γ being the gyromagnetic ratio.

In the absence of a pulse, $\omega_1 = 0$. If the last pulse ended at time τ, the solution to Eq. [1] may be written as

$$M_{\pm 1}(t + \tau) = \exp\left[-\left(\frac{1}{T_2} \pm i\Delta\omega\right)t\right]M_{\pm 1}(\tau)$$

$$\equiv d_{\pm 1}(t)\exp(\mp i\Delta\omega t)M_{\pm 1}(\tau), \quad [2]$$

$$M_0(t + \tau) = \exp\left(-\frac{t}{T_1}\right)M_0(\tau) + M_{eq}\left[1 - \exp\left(-\frac{t}{T_1}\right)\right]$$

$$\equiv d_0(t)M_0(\tau) + M_{eq}g(t), \quad [3]$$

where $M_{\pm 1}$, M_{-1}, and M_0 are defined as

$$M_{\pm 1} = \frac{1}{\sqrt{2}}(\mp M_x - iM_y), \quad [4]$$

$$M_0 = M_z. \quad [5]$$

The transverse magnetization components M_{+1} and M_{-1} rotate clockwise and counterclockwise, respectively, and M_0 is the longitudinal magnetization component. $d_m(t)$ represents m (=+1, −1, 0)-state magnetization decay due to the T_2 or T_1 relaxation processes, and $g(t)$ represents 0-state magnetization growth due to the T_1 relaxation process during the evolution period. While the time dependence of $M_{\pm 1}$ is described by only one term which decays with a time constant T_2 keeping the past phase memory during the evolution period, that of M_0 is described by two terms. The first term decays with a time constant T_1, also keeping the past phase memory, but the second term grows with the same time constant to the thermal equilibrium without phase coherence. Since these magnetization states are mixed only by an RF pulse and evolve independently in the absence of a pulse, the description of the response of a spin system to multiple pulses is simple.

If a pulsed RF field is applied for a short time Δt negligible compared to the relaxation times, the solution to Eq. [1] at the end of the pulse is expressed as

$$\mathbf{M}(\tau) = \mathbf{M}(\tau - \Delta t)\cdot\mathbf{R}(\Omega; \theta, \phi), \quad [6]$$

where

$$\Omega = \Delta t\sqrt{\omega_1^2 + \Delta\omega^2}, \quad [7]$$

$$\theta = \tan^{-1}\left(\frac{\omega_1}{\Delta\omega}\right), \quad [8]$$

and ϕ is the phase angle of the pulse (Fig. 1). The general rotation of the magnetization **R** is composed of the Euler rotation matrices as (*12*)

$$\mathbf{R}(\Omega; \theta, \phi) = R_z(\phi)R_y(\theta)R_z(\Omega)R_y^{-1}(\theta)R_z^{-1}(\phi). \quad [9]$$

If the magnetization vector **M** in Eq. [6] is expressed by the rotating components in Eqs. [4] and [5], the rotation matrix has the elements (*13*)

$$[\mathbf{R}(\Omega; \theta, \phi)]_{11} = e^{-i\Omega}\cos^4\frac{\theta}{2} + \frac{1}{2}\sin^2\theta + e^{i\Omega}\sin^4\frac{\theta}{2}, \quad [10]$$

$$[\mathbf{R}(\Omega; \theta, \phi)]_{01} = \frac{1}{\sqrt{2}}e^{i\phi}\sin\theta\left(e^{-i\Omega}\cos^2\frac{\theta}{2} - \cos\theta - e^{i\Omega}\sin^2\frac{\theta}{2}\right), \quad [11]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{-11} = \frac{1}{4}e^{i2\phi}\sin^2\theta(e^{-i\Omega} - 2 + e^{i\Omega}), \quad [12]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{10} = [\mathbf{R}(\Omega;\theta,-\phi)]_{01}, \quad [13]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{00} = \frac{1}{2}(e^{-i\Omega}\sin^2\theta + 2\cos^2\theta + e^{i\Omega}\sin^2\theta), \quad [14]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{-10} = [\mathbf{R}(-\Omega;-\theta,\phi)]_{01}, \quad [15]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{1-1} = [\mathbf{R}(\Omega;\theta,\phi)]_{-11}, \quad [16]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{0-1} = [\mathbf{R}(-\Omega;-\theta,-\phi)]_{01}, \quad [17]$$

$$[\mathbf{R}(\Omega;\theta,\phi)]_{-1-1} = [\mathbf{R}(-\Omega;\theta,\phi)]_{11}. \quad [18]$$

The elements of this rotation matrix in Cartesian coordinates are too complicated to be practical. A simple representation of the rotation matrix is another advantage we get by using the rotating magnetization components. As shown in Fig. 1, the angles θ and ϕ specify the direction of the rotation axis, and Ω the clockwise flip angle about the axis. Each element of the rotation matrix represents the transfer fraction of a magnetization state to another state by an RF pulse (*4*).

Successive multiplication of the solutions in the presence and absence of a pulse leads to the response of a spin system to a pulse sequence. Generally, an RF pulse transfers one magnetization state to a sum of three magnetization states. Each magnetization state evolves time independently in the absence of a pulse and is transferred to the linear combination of three states again when another pulse is applied. A pathway experienced by a magnetization component after N pulses can be represented by the array of the states after each pulse as $\{m_0, m_1, \ldots, m_N\}$. Here m_0 is the magnetization state before the first pulse which is 0 if the initial state is in thermal equilibrium, and m_i $(i \geqslant 1)$ is the state after the ith pulse. A pathway may start not only from m_0 but also in the middle of a pulse sequence as $\{0, m_i, m_{i+1}, \ldots, m_N\}$ $(i > 1)$. The subpathway like this indicates the path which starts from a longitudinal component generated in the interval between the $(i-1)$th and ith pulses with no coherence. The main pathway vector defined as $\mathbf{m}^1 = (m_1, m_2, \ldots, m_N)$ and subpathway vectors defined as $\mathbf{m}^i = (m_i, m_{i+1}, \ldots, m_N)$ $(i > 1)$, dropping the first states in the pathways, are useful for describing the general response of a spin system to multiple pulses as shown below.

From Eqs. [2] and [3] and Eq. [6], a magnetization state formed through a main pathway can be expressed as

$$M(\mathbf{m}^1) = M_{m_0} r_{m_0m_1} r_{m_1m_2} \cdots r_{m_{N-1}m_N} d_{m_1}(\tau_1) d_{m_2} \times (\tau_2) \cdots d_{m_N}(\tau_N) \exp[-i\Delta\omega(\mathbf{m}^1 \cdot \boldsymbol{\tau}^1)], \quad [19]$$

where M_{m_0} is the magnetization of the initial m_0 state, $r_{m_im_{i+1}}$ is the (m_i, m_{j+1}) element of the rotation matrix $\mathbf{R}$, and $\boldsymbol{\tau}^1$ is the interval vector defined as

$$\boldsymbol{\tau}^1 = (\tau_1, \tau_2, \ldots, \tau_N). \quad [20]$$

Here τ_i $(i < N)$ is the time interval between the ith and $(i + 1)$th pulses, and τ_N is the time elapsed after the Nth pulse. A magnetization state formed through a subpathway is similarly described except that the initial value is $M_{eq}g(\tau_{i-1})$ instead of M_{m_0}. Defining the general interval vector $\boldsymbol{\tau}^i = (\tau_i, \tau_{i+1}, \ldots, \tau_N)$, a magnetization state formed through a pathway vector $\mathbf{m}^i$ is described as

$$M(t, \Delta\omega; \mathbf{m}^i) = M_{m_{i-1}}\left[\prod_{j=i}^{N} r_{m_{j-1}m_j} d_{m_j}(\tau_j)\right]\exp[-i\Delta\omega(\mathbf{m}^i \cdot \boldsymbol{\tau}^i)], \quad [21]$$

where the initial magnetization $M_{m_{i-1}}$ is M_{m_0} or $M_{eq}g(\tau_{i-1})$ according to whether the pathway vector is a main vector or a subvector. The time after the last pulse τ_N is now replaced by a more familiar expression t.

The pathway vector introduced here is analogous to the coherence-transfer pathway vector $\Delta\mathbf{p}$ of Bodenhausen *et al.* which is known to be useful for analyzing the signals in a multiple-quantum coherence. This vector takes the change of the coherence order by an RF pulse as an element, but our pathway vector takes the coherence order itself. The coherence order, or the index of the state, determines the phase-angle change of a magnetization state during the evolution period as seen in Eq. [21]. Since an echo is formed when the phase angle becomes zero as discussed in the next section, it is the coherence order itself rather than the coherence order difference that is directly related with echo formation.

The total NMR signal from a sample is given by the integral of Eq. [21] over various $\Delta\omega$. The state measured in quadrature detection is $+1$ or -1. If the $+1$ state is measured, only the pathway vectors whose last elements are $+1$ contribute to the signal. The signal observed after the Nth pulse is

$$s_{\mathbf{m}^i}(t) \propto \int_{-\infty}^{\infty} g(\Delta\omega) M(t, \Delta\omega; \mathbf{m}^i) d(\Delta\omega) = M_{m_{i-1}}\left[\prod_{j=i}^{n} r_{m_{j-1}m_j} d_{m_j}(\tau_j)\right]\int_{-\infty}^{\infty} g(\Delta\omega) \times \exp[-i\Delta\omega(\mathbf{m}^i \cdot \boldsymbol{\tau}^i)] d(\Delta\omega), \quad [22]$$

for a pathway vector $\mathbf{m}^i$. Here $g(\Delta\omega)$ is the normalized distribution function of the external magnetic field, and the dependence of the rotation matrix element r_{ij} on $\Delta\omega$ is neglected for simplicity of calculation. One of the phenomena generated by this dependence is the edge echo which appears even with a single RF pulse in a very inhomogeneous field (*3, 14*). After all, the NMR signal $S(t)$ is obtained by summing Eq. [22] over all possible pathway vectors as

$$S(t) = \sum_i \sum_{\mathbf{m}^i} S_{\mathbf{m}^i}(t). \quad [23]$$

This equation shows explicitly that the sum is accomplished over all the main and subpathway vectors.

THE ANALYSIS OF MULTIPLE ECHOES DUE TO MULTIPLE PULSES

The free-induction signal generally depends on the Fourier transform of the static magnetic field distribution $g(\Delta\omega)$ as seen in Eq. [22]. But at the echo time, i.e., when the inner product of two vectors $\mathbf{m}^i$ and $\boldsymbol{\tau}^i$ is zero so that the kernel of the Fourier transform becomes 1, the signal becomes independent of external field inhomogeneity. Consequently, for $\mathbf{m}^i$ satisfying the following condition with positive t

$$\mathbf{m}^i \cdot \boldsymbol{\tau}^i = 0, \quad [24]$$

the echo amplitude is given by

$$s_{\mathbf{m}^i}(t) \propto M_{m_{i-1}} \prod_{j=i}^{N} r_{m_{j-1}m_j} d_{m_j}(\tau_j), \quad [25]$$

because the field distribution function $g(\Delta\omega)$ is normalized. Of course, it is assumed that the angular precession frequency remains constant in each interval between pulses. Otherwise, there will be signal decays owing to the extra phase angle distribution in addition to the intrinsic relaxation decay, such as the signal attenuation by molecular diffusion in an inhomogeneous field.

Among the various pathway vectors, only those which satisfy Eq. [24] for positive t form echoes. With each echo, one main pathway vector is related. If a main pathway forming an echo experiences 0 states initially, there are subpathways forming echoes at the same echo time. Suppose that a main pathway vector $\mathbf{m}^1 = (0, 0, \ldots, 0, m_i, m_{i+1}, \ldots, m_N)$ $(i > 1)$ having $(i-1)$ 0 states initially forms an echo at time t. Then the subvectors $\mathbf{m}^k = (0, 0, \ldots, m_i, m_{i+1}, \ldots, m_N)$ $(i > 1)$ having $(i-k)$ 0 states initially form echoes at the same time t. For example, suppose that a main pathway vector forming an echo is $(0, 0, -1, 1)$ in an experiment with four pulses. Then the subpathway vectors $(0, -1, 1)$ and $(-1, 1)$ also satisfy Eq. [24] with same time t. Therefore, these three pathway vectors make echoes at the same position.

In general, for a main pathway vector having n succesive 0 elements initially, there are n subpathway vectors that make echoes at the same time, whose dimensions are lowered from $N-1$ to $N-n$ by eliminating the initial zeroes of the corresponding main pathway vector one by one. Predicting echoes after a pulse sequence is equivalent to finding the pathway vectors corresponding to the echoes. This process of finding echoes is applicable not only after the last pulse but also after any pulse in the middle of a pulse sequence. But in the middle of a pulse sequence, some of the expected echoes may not appear if the time interval between adjacent pulses is not long enough.

As an example, we apply the general solution to the case of three pulses. If three pulses of flip angles α_1, α_2, and α_3 about the rotation axis $\theta = 90°$ and $\phi = 0°$ are applied to a spin system with pulse intervals τ_1 and τ_2 $(>\tau_1)$, five echoes are observed. One is formed after the second pulse, and the other four are formed after the third pulse. The conventional primary echo after the second pulse is generated by a pathway vector $(-1, 1)$. Among the nine main pathway vectors with three pulses, only $\mathbf{a} = (-1, 0, 1)$, $\mathbf{b} = (0, -1, 1)$, $\mathbf{c} = (-1, -1, 1)$, and $\mathbf{d} = (1, -1, 1)$ generate echoes. Echoes are formed at $t = \tau_1, \tau_2, \tau_1 + \tau_2$, and $\tau_2 - \tau_1$, respectively, from $\mathbf{m}^i \cdot \boldsymbol{\tau}^i = 0$. The subpathway vector generating an echo after the final pulse is only $(-1, 1)$ which forms an echo at $t = \tau_2$ as $\mathbf{b}$. The echo amplitudes are obtained from Eq. [25] as

$$M_{\text{eq}} r^{(1)}_{0-1} r^{(2)}_{10} r^{(3)}_{01} d_{-1}(\tau_1) d_0(\tau_2) d_1(\tau_1), \quad [26]$$

$$M_{\text{eq}}[r^{(1)}_{00} d_0(\tau_1) + g(\tau_1)] r^{(2)}_{0-1} r^{(3)}_{11} d_{-1}(\tau_2) d_1(\tau_2), \quad [27]$$

$$M_{\text{eq}} r^{(1)}_{0-1} r^{(2)}_{-1-1} r^{(3)}_{-11} d_{-1}(\tau_1) d_{-1}(\tau_2) d_1(\tau_1 + \tau_2), \quad [28]$$

$$M_{\text{eq}} r^{(1)}_{01} r^{(2)}_{1-1} r^{(3)}_{-11} d_1(\tau_1) d_{-1}(\tau_2) d_1(\tau_2 - \tau_1), \quad [29]$$

which are proportional to the following explicit expressions:

$$\frac{1}{2} \sin\alpha_1 \sin\alpha_2 \sin\alpha_3 \exp(-\tau_2/T_1 - 2\tau_1/T_2), \quad [30]$$

$$[1 - (1 - \cos\alpha_1)\exp(-\tau_1/T_1)] \times \sin\alpha_2 \sin^2\frac{\alpha_3}{2} \exp(-2\tau_2/T_2), \quad [31]$$

$$\sin\alpha_1 \cos^2\frac{\alpha_2}{2} \sin^2\frac{\alpha_3}{2} \exp[-2(\tau_1 + \tau_2)/T_2], \quad [32]$$

$$-\sin\alpha_1 \sin^2\frac{\alpha_2}{2} \sin^2\frac{\alpha_3}{2} \exp(-2\tau_2/T_2). \quad [33]$$

This is the same as Woessner's result (*4*). By applying the general solution using the magnetization pathway to any multiple-pulse sequence, we can easily obtain the number, time, and magnitude of echoes with much less chance of calculation mistakes, and also easily find the physical origin of an echo. The concept of the magnetization pathway vector

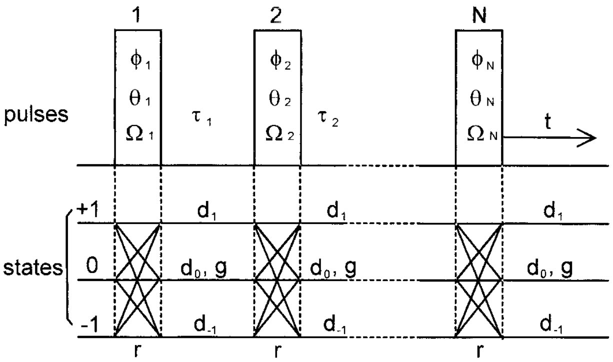

FIG. 2. *N*-pulse sequence and the magnetization pathways.

becomes more useful with increasing number of pulses, especially in predicting the number of echoes.

THE NUMBER OF ECHOES AFTER *N* PULSES

As mentioned above, an echo is formed if the scalar product of $\mathbf{m}^i$ and $\boldsymbol{\tau}^i$ is zero. Therefore, an echo time is given by the difference between the time remaining at the -1 state and that at the $+1$ state of the corresponding pathway. Let us consider a N-pulses sequence of which the intervals differ from each other as depicted in Fig. 2. To be most general, we further assume that the linear combinations of intervals are also different from each other. In this condition, it is guaranteed that there is no accidental disappearance of echo; that is, no echoes each of which is related with different main pathways appear at the same position.

Counting the number of echoes appearing after the final pulse is equivalent to counting the main pathway vectors forming echoes, because subpathway vectors cannot form echoes at separate positions. The main pathway vector consisting of all zeroes cannot generate an echo. The half of the remaining main pathway vectors can generate echoes after the final pulse because if a main pathway vector cannot satisfy Eq. [24] for positive t, a vector formed by reversing the sign of the elements except the last one can. The vector made in this way also belongs to the main pathway vectors. Each element of a main pathway vector can have three different values. Therefore, the total number of the main pathway vectors of N dimensions is 3^{N-1} considering only pathways ending with the $+1$ state. Subtracting the one consisting of only zeroes and dividing by 2 gives the total number of echoes after N pulses

$$E_N = \tfrac{1}{2}(3^{N-1} - 1). \qquad [34]$$

This formula gives the number of echoes $E_1 = 0$, $E_2 = 1$, $E_3 = 4$, $E_4 = 13$, $E_5 = 40$, and $E_6 = 121$ with 1, 2, 3, 4, 5, and 6 pulses, respectively, which disagree with Das's results, 0, 1, 4, 13, 39, and 112 (*2*).

This difference is due to Das's mistake in counting the number of echoes with his complicated method. In the following, we point out his mistake and show that the results are the same if the cases omitted in his analysis are included. He counted the number of echoes as follows. Designating y_n as the number of echoes expected after n pulses,

(a) number of primary echoes due to interaction directly between the nth pulse and any of the previous pulses $= {}_{n-1}C_1$;

(b) number of primary echoes due to interaction between the nth pulse and previous echoes $= y_{n-1} + y_{n-2} + \cdots + y_1$;

(c) number of stimulated echoes between the nth pulse and any two of the previous pulses $= {}_{n-1}C_2$;

(d) number of stimulated echoes between the nth and $(n-1)$th pulses and a previous echo $= y_{n-2} + y_{n-3} + \cdots + y_1$;

(e) number of quaternary mechanism echoes between the nth pulse and the three previous pulses $= {}_{n-1}C_3$;

(f) number of quaternary mechanism echoes between the previous echoes and the last three pulses $= y_{n-3} + y_{n-4} + \cdots + y_1$;

(g) number of $(n-1)$-mechanism echoes between the nth pulse and $(n-2)$ previous pulses $= {}_{n-1}C_{n-2}$;

(h) number of $(n-1)$-mechanism echoes between the last pulses and the previous echoes $= y_2 + y_1$;

(i) number of n-mechanism echoes $= {}_{n-1}C_{n-1} = 1$.

The above echo-forming mechanisms can be classified into two categories. One is due to the ''interaction'' between pulses, and the other between pulses and echoes. The number of echoes counted in terms (a), (c), (e), (g), and (i) belonging to the former is correct while in the terms (b), (d), (f), and (h) belonging to the latter some cases are missing. The number of echoes counted in (b) is correct. In (d), however, $y_{n-2} + y_{n-3} + \cdots + y_1$ is not the total number of echoes available by the mentioned mechanism. There are also ''stimulated echoes'' between the nth and $(n-2)$th pulses and a previous echo by the same mechanism. The number of these stimulated echoes is $y_{n-3} + y_{n-4} + \cdots + y_1$. Similarly, the number of echoes between the nth and $(n-3)$th pulses and a previous echo is $y_{n-4} + y_{n-5} + \cdots + y_1$. Proceeding this way, the total number of echoes expected in (d) is

$$\begin{aligned} & y_{n-2} + y_{n-3} + y_{n-4} + \cdots + y_1 \\ & \quad + y_{n-3} + y_{n-4} + \cdots + y_1 \\ & \qquad + y_{n-4} + \cdots + y_1 \\ & \qquad \vdots \\ & \qquad\qquad + y_1 \\ & = y_{n-2} + 2y_{n-3} + 3y_{n-4} + \cdots + (n-2)y_1. \end{aligned} \qquad [35]$$

A similar correction is necessary in (f). The ''quaternary mechanism echoes'' are also formed with the nth, $(n-1)$th, and $(n-3)$th pulses and nth, $(n-2)$th, and $(n-3)$th pulses as well as with the nth, $(n-1)$th, and $(n-2)$th pulses. The number of additional quaternary echoes between the previous echoes and three pulses including the last and $(n-3)$th pulses is $2(y_{n-4}+y_{n-5}+\cdots+y_1)$. Similarly, the number of quaternary mechanism echoes obtained by fixing the last and $(n-4)$th pulses is ${}_3C_1(y_{n-5}+y_{n-6}+\cdots+y_1)$. Proceeding in this way, the total number of echoes expected in (f) is

$$\begin{aligned} & y_{n-3}+y_{n-4}+y_{n-5}+\cdots+y_i+\cdots+y_1 \\ & \quad +2(y_{n-4}+y_{n-5}+\cdots+y_i+\cdots+y_1) \\ & \quad\quad +3(y_{n-5}+\cdots+y_i+\cdots+y_1) \\ & \quad\quad\quad \vdots \\ & \quad\quad\quad +(n-3)y_1 \\ & = y_{n-3}+3y_{n-4}+6y_{n-5}+\cdots+\frac{(n-i-2)(n-i-1)}{2}y_i+ \\ & \quad \cdots+\frac{(n-3)(n-2)}{2}y_1. \end{aligned} \quad [36]$$

This counting structure is applicable until (h). Consequently, the total number of echoes expected with the interaction between pulses and echoes is

$$\begin{aligned} & y_{n-1}+y_{n-2}+y_{n-3}+y_{n-4}+\cdots+y_1 \\ & \quad +y_{n-2}+2y_{n-3}+3y_{n-4}+\cdots+(n-2)y_1 \\ & \quad\quad +y_{n-3}+3y_{n-4}+\cdots+\frac{(n-3)(n-2)}{2}y_1 \\ & \quad\quad\quad \vdots \\ & \quad\quad\quad +y_1 \\ & = y_{n-1}+2y_{n-2}+4y_{n-3}+\cdots+2^{r-1}y_{n-r}+ \\ & \quad \cdots+2^{n-2}y_1=\sum_{r=1}^{n-1}2^{r-1}y_{n-r}. \end{aligned} \quad [37]$$

Here, we see that the coefficients of y_i constitute Pascal's triangle. Including the number of echoes due to the interaction between pulses, $2^{n-1}-1$, the total number of echoes is not

$$\begin{aligned} y_n &= {}_{n-1}C_1+{}_{n-1}C_2+\cdots+{}_{n-1}C_{n-1} \\ & \quad +[y_{n-1}+2y_{n-2}+\cdots+ry_{n-r} \\ & \quad\quad +\cdots+(n-1)y_1] \\ & = 2^{n-1}-1+[y_{n-1}+2y_{n-2}+\cdots+(n-1)y_1] \end{aligned} \quad [38]$$

given by Das, but is

$$y_n = 2^{n-1}-1+\sum_{r=1}^{n-1}2^{r-1}y_{n-r}. \quad [39]$$

By replacing n by $n+1$ in Eq. [39] and rearranging terms, we get the following recursion relation:

$$y_{n+1}=3y_n+1. \quad [40]$$

With $y_1=0$, this recursion relation gives the same formula as in Eq. [34]. The formula always holds true without special conditions on the intervals between pulses as Das imposed, unless echoes disappear accidently as mentioned above.

REFERENCES

1. E. L. Hahn, *Phys. Rev.* **80,** 580 (1950).
2. T. P. Das and D. K. Roy, *Phys. Rev.* **98,** 525 (1955).
3. A. L. Bloom, *Phys. Rev.* **98,** 1105 (1955).
4. D. E. Woessner, *J. Chem. Phys.* **34,** 2057 (1961).
5. R. Kaiser, E. Bartholdi, and R. R. Ernst, *J. Chem. Phys.* **60,** 2966 (1974).
6. S. Majumdar, S. C. Orphanoudakis, A. Gmitro, M. O'Donnell, and J. C. Gore, *Magn. Reson. Med.* **3,** 397 (1986).
7. D. G. Norris and P. Boernert, *J. Magn. Reson. A* **105,** 123 (1993).
8. S. Meiboom and D. Gill, *Rev. Sci. Instrum.* **29,** 688 (1958).
9. E. O. Stejskal and J. Schaefer, *J. Magn. Reson.* **14,** 160 (1974).
10. A. D. Bain, *J. Magn. Reson.* **56,** 418 (1984).
11. G. Bodenhausen, H. Kogler, and R. R. Ernst, *J. Magn. Reson.* **58,** 370 (1984).
12. G. Burns, ''Introduction to Group Theory with Applications,'' Academic Press, New York, 1977.
13. M. E. Rose, ''Elementary Theory of Angular Momentum,'' Wiley, New York, 1967.
14. R. Kaiser, *J. Magn. Reson.* **42,** 103 (1981).