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Simultaneous and Independent Rotations with Arbitrary Flip Angles and Phases for I, IS$^\alpha$, and IS$^\beta$ Spin Systems

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A new pulse sequence element for simultaneous and independent rotations with arbitrary flip angles and phases for isolated I, IS$^\alpha$, and IS$^\beta$ resonances without the use of selective radiofrequency pulses is introduced and experimentally demonstrated. S is a directly attached heteronucleus either at natural abundance or isotopically enriched. This pulse sequence element, dubbed TIG-BIRD (triselective independent gyrations BIRD), generalizes earlier elements like BIRD, TANGO, BANGO, and BIG-BIRD, the latter of which allows for arbitrary selection of flip angles and phases for I and IS spin systems without discriminating between IS$^\alpha$ and IS$^\beta$ resonances. For IS$^\alpha$ and IS$^\beta$ spin systems it also generalizes the spin-state-selective excitation (S^{3}E) element selectively exciting only one of the IS$^\alpha$ or IS$^\beta$ resonances. TIG-BIRD is a nonselective addition to the NMR toolkit which effects the equivalent of three independent selective rotations for I, IS$^\alpha$, and IS$^\beta$ resonances.

Key Words: **BIRD; selective rotations; nonselective pulses; $^1J_{IS}$ heteronuclear coupling constant; TIG-BIRD.**

A number of pulse sequences have been developed over the years which exploit the large one-bond heteronuclear coupling between protons directly attached to, e.g., ^{13}C or ^{15}N nuclei at either natural abundance or isotopically enriched for differentiating the protons directly bound to a heteronucleus from those not directly bound to a heteronucleus. For example, BIRD (*1*) was designed as a selective inversion element while TANGO (*2*) was proposed as a selective excitation or combined excitation-inversion element. These two elements were generalized in BANGO (*3*), which allows independent setting of arbitrary rotation angles of identical phase for the two classes of protons. Finally, a more recent pulse sequence element, BIG-BIRD (*4*), allows for arbitrary selection of flip angles and phases for I and IS spin systems but does not discriminate between IS$^\alpha$ and IS$^\beta$ resonances.

For protons directly bound to a heteronucleus, schemes have been designed to select only one component of the doublet by using heteronuclear coherence transfer echoes and pulsed field gradients (*5, 6*). Other more recent approaches, the spin-state-selective excitation (S^{3}E) (*7, 8*) and S^{3}CT (*9*) pulse sequence elements, allow both IS$^\alpha$ and IS$^\beta$ subspectra to be constructed from the same data set. A related experiment, TROSY (*10*), also aims at selecting only one of four correlations within a ^{15}N–^{1}H moiety which, for macromolecules, has the property of exhibiting a narrow peak at very high B_0 fields.

However, none of these techniques allow for arbitrary selection of the flip angle and phase for each individual component in I and IS spin systems. That becomes possible with our new pulse sequence element, TIG-BIRD (triselective independent gyrations BIRD), which enables simultaneous and independent rotation of I, IS$^\alpha$, and IS$^\beta$ resonances. We derive the new pulse sequence element using a vector model and demonstrate its essential features by a simple application.

The TIG-BIRD pulse sequence element, P, must as illustrated in Fig. 1a achieve simultaneous and independent rotations with arbitrary flip angles and phases for isolated I, IS$^\alpha$, and IS$^\beta$ resonances, i.e.,

$$\left\{\begin{array}{ll} \mathrm{I}: & (\gamma^{\mathrm{I}})_{\varphi^{\mathrm{I}}} \\ \mathrm{IS}^\alpha: & (\gamma^\alpha)_{\varphi^\alpha} \\ \mathrm{IS}^\beta: & (\gamma^\beta)_{\varphi^\beta} \end{array}\right. , \qquad [1]$$

where $(\gamma^{\mathrm{I}}, \gamma^\alpha, \gamma^\beta)$ are the flip angles and $(\varphi^{\mathrm{I}}, \varphi^\alpha, \varphi^\beta)$ the phases for I-spin magnetization of I, IS$^\alpha$, and IS$^\beta$ spin systems, respectively. Explicitly, this amounts to the transformations (*11*)

$$I_z \xrightarrow{P} I_z\sin(\gamma^{\mathrm{I}})\sin(\varphi^{\mathrm{I}}) - I_y\sin(\gamma^{\mathrm{I}})\cos(\varphi^{\mathrm{I}}) + I_z\cos(\gamma^{\mathrm{I}}) \qquad [2a]$$

$$I_zS^{\alpha,\beta} \xrightarrow{P} I_xS^{\alpha,\beta}\sin(\gamma^{\alpha,\beta})\sin(\varphi^{\alpha,\beta}) - I_yS^{\alpha,\beta}\sin(\gamma^{\alpha,\beta})\cos(\varphi^{\alpha,\beta}) + I_zS^{\alpha,\beta}\cos(\gamma^{\alpha,\beta}). \qquad [2b]$$

To derive TIG-BIRD we first ignore isolated I-spin magnetization and focus on an IS spin system with IS$^\alpha$ and IS$^\beta$ magnetizations. Following an illustrative and straightforward approach employed in the design of BIG-BIRD (*4*), we derive P^{-1}, i.e., a pulse sequence element which performs the opposite transformation of taking the magnetization vectors from their desired final positions back to the z axis, and then invert that pulse sequence. Five basic steps applied sequentially and

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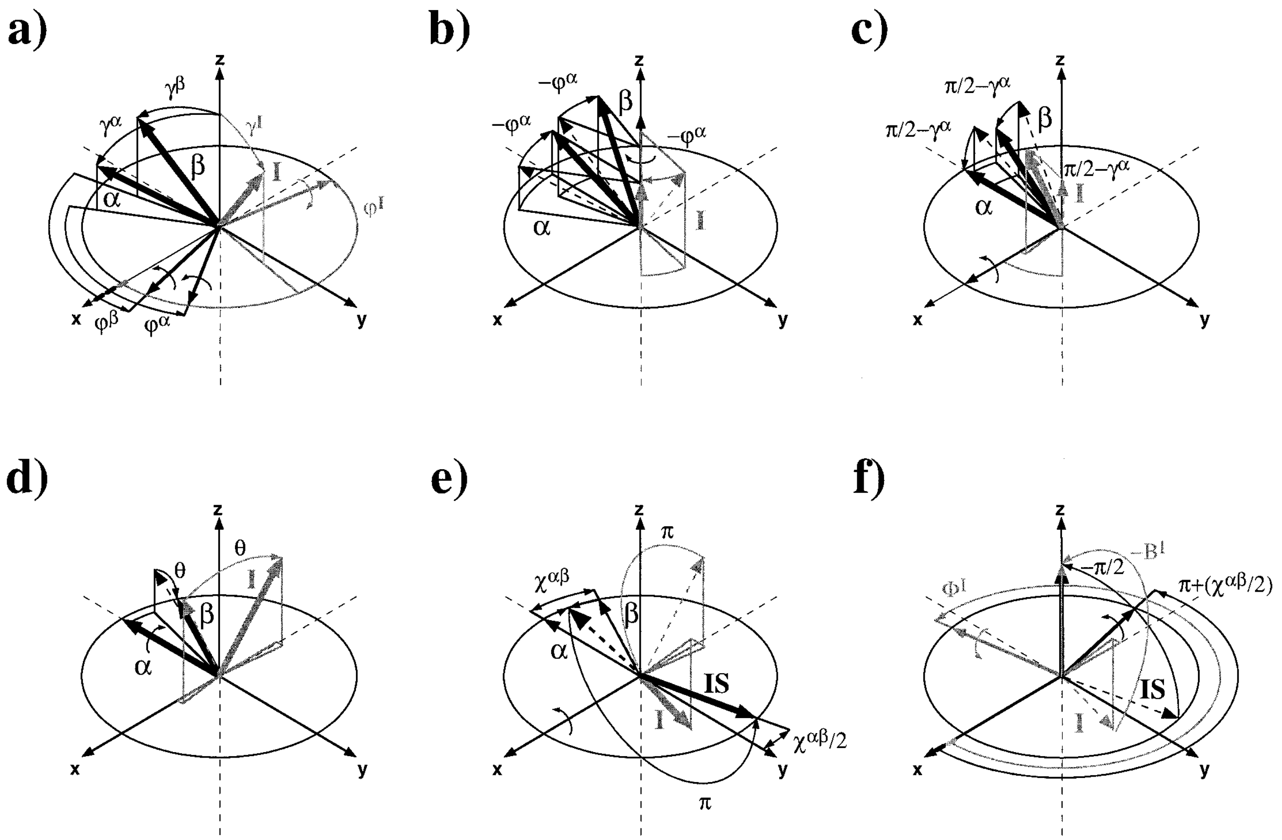

FIG. 1. Vector model representation of the basic steps involved in the design of the TIG-BIRD pulse sequence element. (a) The desired rotations of TIG-BIRD with I-spin z magnetization of I, IS$^\alpha$, and IS$^\beta$ spin systems being transformed by rotations of flip angles (γ^{I}, γ^α, γ^β) and phases (φ^{I}, φ^α, φ^β), respectively. (b)–(f) The five basic steps involved in the reverse derivation of TIG-BIRD (see text).

individually shown in Figs. 1b–1f will accomplish the goal: first, apply a rotation of angle $-\varphi^\alpha$ about the z axis (Fig. 1b) in order to bring IS$^\alpha$ magnetization into the yz plane; second, apply a rotation of angle ($\pi/2 - \gamma^\alpha$) about the x axis in order to align IS$^\alpha$ magnetization along the $-y$ axis (Fig. 1c); third, apply a rotation of angle θ about IS$^\alpha$ magnetization in order to bring IS$^\beta$ magnetization into the transverse plane (Fig. 1d); fourth, apply a spin-echo period of sufficient duration to allow heteronuclear coupling to refocus IS$^\alpha$ and IS$^\beta$ magnetization vectors along an axis in the transverse plane (Fig. 1e); finally, in the last step, apply a phase-shifted ($\pi/2$) rf pulse to flip both vectors back along the z axis (Fig. 1f). A pulse sequence effecting this series of transformations consists of a variable spin-echo delay surrounded by two pulses (*vide infra*). If I-spin systems also need to be manipulated independently, the simple ($\pi/2$) rf pulse in the last step must be replaced by an inverted BIG-BIRD pulse sequence which will bring IS$^\alpha$ and IS$^\beta$ as well as I-magnetization vectors back along the z axis (Fig. 1f). For the latter, TIG-BIRD will consist effectively of two modules, one of which will discriminate between IS$^\alpha$ and IS$^\beta$ vectors in an IS spin system while the other will discriminate between I and IS spin systems.

In the third step, IS$^\beta$ magnetization is rotated by an angle θ about IS$^\alpha$ magnetization which is collinear with the $-y$ axis, as shown in Fig. 1d. The angle θ required to bring IS$^\beta$ magnetization into the transverse plane can be determined from the set of equations

$$\begin{bmatrix} -\sin(\chi^{\alpha\beta}) \\ -\cos(\chi^{\alpha\beta}) \\ 0 \end{bmatrix} = \begin{bmatrix} c_\theta & 0 & -s_\theta \\ 0 & 1 & 0 \\ s_\theta & 0 & c_\theta \end{bmatrix} \begin{bmatrix} \sin(\gamma^\beta)\sin(\varphi^\beta - \varphi^\alpha) \\ -\cos(\chi^{\alpha\beta}) \\ -\cos(\chi^{\alpha'\beta}) \end{bmatrix}, \quad [3]$$

where $s_\theta = \sin(\theta)$ and $c_\theta = \cos(\theta)$. $\chi^{\alpha\beta}$ is the angle betwen IS$^\alpha$ and IS$^\beta$ magnetization vectors in their final state as defined in Eq. [2b] and is determined according to the formula for their scalar product

$$\cos(\chi^{\alpha\beta}) = \sin(\gamma^\alpha)\sin(\gamma^\beta)\cos(\varphi^\alpha - \varphi^\beta) + \cos(\gamma^\alpha)\cos(\gamma^\beta), \quad 0 \le \chi^{\alpha\beta} \le \pi \quad [4]$$

while

$$\cos(\chi^{\alpha'\beta}) = \sin(\gamma^\alpha + \pi/2)\sin(\gamma^\beta)\cos(\varphi^\alpha - \varphi^\beta) + \cos(\gamma^\alpha + \pi/2)\cos(\gamma^\beta). \quad [5]$$

An important requirement for the angle θ is to ensure that the total refocusing period never exceeds $1/(2J)$ (π angle). Hence the angle θ should be such that when IS^{β} magnetization is rotated into the transverse plane it should be in either the second or the third quadrant. This requirement is reflected in the signs of the trigonometric functions on the left-hand side of Eq. [3]. Solving Eq. [3] yields

$$\sin(\theta) = \frac{-\sin(\chi^{\alpha\beta})\cos(\chi^{\alpha'\beta})}{\sin^2(\gamma^{\beta})\sin^2(\varphi^{\beta} - \varphi^{\alpha}) + \cos^2(\chi^{\alpha'\beta})}, \qquad [6a]$$

$$\cos(\theta) = \frac{-\sin(\chi^{\alpha\beta})\sin(\gamma^{\beta})\sin(\varphi^{\beta} - \varphi^{\alpha})}{\sin^2(\gamma^{\beta})\sin^2(\varphi^{\beta} - \varphi^{\alpha}) + \cos^2(\chi^{\alpha'\beta})}, \qquad [6b]$$

which defines θ in the interval $0 \le \theta \le 2\pi$. Obviously $\theta = 0$ if $\chi^{\alpha\beta} = 0$ or π, since IS^{β} magnetization is aligned either parallel or antiparallel to IS^{α} magnetization.

In the fourth step, a spin-echo refocusing period which keeps heteronuclear coupling active but refocuses chemical shift evolution will align the IS^{α} and IS^{β} magnetization components which are separated by an angle $\chi^{\alpha\beta}$ (Fig. 1e) after the third step. That requires a pulse sequence element of the form

$$\frac{\Delta}{2} - (\pi)_x^{\mathrm{I,S}} - \frac{\Delta}{2}, \qquad [7]$$

where $\Delta = \chi^{\alpha\beta}/(2\pi J)$. Thereafter, IS^{α} and IS^{β} vectors are collinear and their orientation in the transverse plane is

$$\mathrm{I\{S\}}: \quad -I_x\sin(\chi^{\alpha\beta}/2) + I_y\cos(\chi^{\alpha\beta}/2). \qquad [8]$$

In the final step, a $(-\pi/2)_{\pi+(\chi^{\alpha\beta}/2)}$ pulse brings both vectors back along the z axis (Fig. 1f). We postpone the inversion of the pulse sequence derived and let it emerge as a special case of the full TIG-BIRD sequence which also independently manipulates isolated I-spin magnetization (*vide infra*).

Given the desired final state in Fig. 1a and expressed by Eq. [1], the orientation of isolated I-spin magnetization following the first four steps above is

$$\begin{aligned} I = {} & I_x[c_\theta\sin(\gamma^{\mathrm{I}})\sin(\varphi^{\mathrm{I}} - \varphi^{\alpha}) + s_\theta\cos(\chi^{\alpha'\mathrm{I}})] + I_y\cos(\chi^{\alpha\mathrm{I}}) \\ & - I_z[s_\theta\sin(\gamma^{\mathrm{I}})\sin(\varphi^{\mathrm{I}} - \varphi^{\alpha}) - c_\theta\cos(\chi^{\alpha'\mathrm{I}})], \end{aligned} \qquad [9]$$

where $\chi^{\alpha\mathrm{I}}$ and $\chi^{\alpha'\mathrm{I}}$ are defined in analogy to Eqs. [4] and [5] with the former being the angle between IS^{α} and I vectors in the final state as defined in Eq. [2], i.e.,

$$\begin{aligned} \cos(\chi^{\alpha\mathrm{I}}) = {} & \sin(\gamma^{\alpha})\sin(\gamma^{\mathrm{I}})\cos(\varphi^{\alpha} - \varphi^{\mathrm{I}}) \\ & + \cos(\gamma^{\alpha})\cos(\gamma^{\mathrm{I}}), \quad 0 \le \chi^{\alpha\mathrm{I}} \le \pi \end{aligned} \qquad [10]$$

$$\begin{aligned} \cos(\chi^{\alpha'\mathrm{I}}) = {} & \sin(\gamma^{\alpha} + \pi/2)\sin(\gamma^{\mathrm{I}})\cos(\varphi^{\alpha} - \varphi^{\mathrm{I}}) \\ & + \cos(\gamma^{\alpha} + \pi/2)\cos(\gamma^{\mathrm{I}}). \end{aligned} \qquad [11]$$

In order to bring all three magnetization vectors back to the z axis, an inverted BIG-BIRD pulse sequence of the form

$$(-\epsilon')_{\lambda'} - \frac{1}{2J} - (\pi)_x - \frac{1}{2J} - (-\epsilon)_{\lambda} \qquad [12]$$

is required. The flip angles (ϵ, ϵ') and phases (λ, λ') can be calculated from the desired rotations for IS and I spin systems as dictated by Eqs. [8] and [9]: for IS spin systems it must be a $(-\pi/2)_{\pi+(\chi^{\alpha\beta}/2)}$ rotation while for I spin systems it is a $(-B^{\mathrm{I}})_{\Phi^{\mathrm{I}}}$ rotation with the flip angle B^{I} and phase Φ^{I} expressed as (the negative flip angles serve a later convenience when P must be inverted)

$$\begin{aligned} \cos(B^{\mathrm{I}}) = {} & -s_\theta\sin(\gamma^{\mathrm{I}})\sin(\varphi^{\mathrm{I}} - \varphi^{\alpha}) + c_\theta\cos(\chi^{\alpha'\mathrm{I}}), \\ & 0 \le B^{\mathrm{I}} \le \pi \end{aligned} \qquad [13]$$

$$\begin{aligned} \sin(\Phi^{\mathrm{I}}) = {} & \frac{c_\theta\sin(\gamma^{\mathrm{I}})\sin(\varphi^{\mathrm{I}} - \varphi^{\alpha}) + s_\theta\cos(\chi^{\alpha'\mathrm{I}})}{\sin(B^{\mathrm{I}})}, \\ & 0 \le \Phi^{\mathrm{I}} \le 2\pi \end{aligned} \qquad [14a]$$

$$\cos(\Phi^{\mathrm{I}}) = \frac{-\cos(\chi^{\alpha\mathrm{I}})}{\sin(B^{\mathrm{I}})}, \quad 0 \le \Phi^{\mathrm{I}} \le 2\pi. \qquad [14b]$$

These five steps described above can now be concatenated,

$$\begin{aligned} P^{-1}: \quad & R_z(-\varphi^{\alpha}) - \left(\frac{\pi}{2} - \gamma^{\alpha}\right)_x^{\mathrm{I}} - (\theta)_{-y}^{\mathrm{I}} - \frac{\Delta}{2} - (\pi)_x^{\mathrm{I,S}} \\ & - \frac{\Delta}{2} - (-\epsilon')_{\lambda'}^{\mathrm{I}} - \frac{1}{2J} - (\pi)_x^{\mathrm{I,S}} - \frac{1}{2J} - (-\epsilon)_{\lambda}^{\mathrm{I}}, \end{aligned} \qquad [15]$$

with $R_z(-\varphi^{\alpha})$ designating a rotation of angle $-\varphi^{\alpha}$ about the z axis. The desired pulse sequence element P may now be determined by inverting P^{-1} in Eq. [15]:

$$\begin{aligned} P: \quad & (\epsilon)_{\lambda}^{\mathrm{I}} - \frac{1}{2J} - (\pi)_x^{\mathrm{I,S}} - \frac{1}{2J} - (\epsilon')_{\lambda'}^{\mathrm{I}} - \frac{\Delta}{2} - (\pi)_x^{\mathrm{I,S}} \\ & - \frac{\Delta}{2} - (\theta)_y^{\mathrm{I}} - \left(\frac{\pi}{2} - \gamma^{\alpha}\right)_{-x}^{\mathrm{I}} - R_z(\varphi^{\alpha}). \end{aligned} \qquad [16]$$

As a first step of simplification we seek a solution in which the third and second to last rotations are replaced according to

$$(\theta)_y^{\mathrm{I}} - \left(\frac{\pi}{2} - \gamma^{\alpha}\right)_{-x}^{\mathrm{I}} = (\mu)_{\psi}^{\mathrm{I}} R_z(\eta). \qquad [17]$$

A solution for Eq. [17] is possible based on the quaternion formalism (*12*, *13*)

$$\cos(\mu/2) = \frac{1}{\sqrt{2}}\sqrt{1 + \sin(\gamma^\alpha)\cos(\theta)} \quad [18]$$

$$\sin(\eta/2) = \frac{-s_t s_g}{\cos(\mu/2)} \quad [19a]$$

$$\cos(\eta/2) = \frac{c_t c_g}{\cos(\mu/2)} \quad [19b]$$

$$\sin(\psi) = \frac{c_t s_g \sin(\eta/2) + s_t c_g \cos(\eta/2)}{\sin(\mu/2)} \quad [20a]$$

$$\cos(\psi) = \frac{-c_t s_g \cos(\eta/2) + s_t c_g \sin(\eta/2)}{\sin(\mu/2)}, \quad [20b]$$

where $s_t = \sin(\theta/2)$, $c_t = \cos(\theta/2)$, $s_g = \sin(\pi/4-\gamma^\alpha/2)$, and $c_g = \cos(\pi/4-\gamma^\alpha/2)$. From Eqs. [18]–[20] the angle μ is defined in the interval $0 \leq \mu \leq \pi$ and the angles ψ, η are defined in the interval $0 \leq \psi, \eta \leq 2\pi$. Substituting Eq. [17] into Eq. [16], we obtain the following version of P:

$$P: \quad (\epsilon)^{\mathrm{I}}_{\lambda} - \frac{1}{2J} - (\pi)^{\mathrm{I,S}}_{x} - \frac{1}{2J} - (\epsilon')^{\mathrm{I}}_{\lambda'} - \frac{\Delta}{2} - (\pi)^{\mathrm{I,S}}_{x} - \frac{\Delta}{2} - (\mu)^{\mathrm{I}}_{\psi} - R_z(\eta + \varphi^\alpha). \quad [21]$$

Using the identities $(\zeta)_\epsilon R_z(\theta) \equiv R_z(\theta)(\zeta)_{\epsilon+\theta}$ and $(\pi)_\epsilon \equiv R_{-z}(2\epsilon)(\pi)_x$, Eq. [21] can be simplified to the final form

$$P: \quad R_z(\eta + \varphi^\alpha) - (\epsilon)^{\mathrm{I}}_{\lambda+\eta+\varphi^\alpha} - \frac{1}{2J} - (\pi)^{\mathrm{I,S}}_{x} - \frac{1}{2J} - (\epsilon')^{\mathrm{I}}_{\lambda'-\eta-\varphi^\alpha} - \frac{\Delta}{2} - (\pi)^{\mathrm{I,S}}_{x} - \frac{\Delta}{2} - (\mu)^{\mathrm{I}}_{\psi+\eta+\varphi^\alpha}. \quad [22]$$

When isolated I spin systems are irrelevant, a $(-\pi/2)_{\pi+(\chi^{\alpha\beta}/2)}$ pulse replaces the inverted BIG-BIRD pulse sequence element in Eq. [15] (*vide supra*). Following the same simplifications as above, Eq. [22] then reduces to

$$P: \quad R_z(\eta + \varphi^\alpha) - (\pi/2)^{\mathrm{I}}_{\pi-\eta-\varphi^\alpha+(\chi^{\alpha\beta}/2)} - \frac{\Delta}{2} - (\pi)^{\mathrm{I,S}}_{x} - \frac{\Delta}{2} - (\mu)^{\mathrm{I}}_{\psi+\eta+\varphi^\alpha}. \quad [23]$$

The z rotation $R_z(\eta + \varphi^\alpha)$ is irrelevant when P is used as an excitation element starting from longitudinal magnetization but causes a phase shift when the element is applied to other components of the density operator. The details of this, including a compensation scheme, will be covered in a separate publication. The pulse sequence elements of Eqs. [22] and [23] are illustrated in Fig. 2. As described above the full TIG-BIRD sequence in Fig. 2c is a combination of BIG-BIRD in Fig. 2a and α,β TIG-BIRD in Fig. 2b.

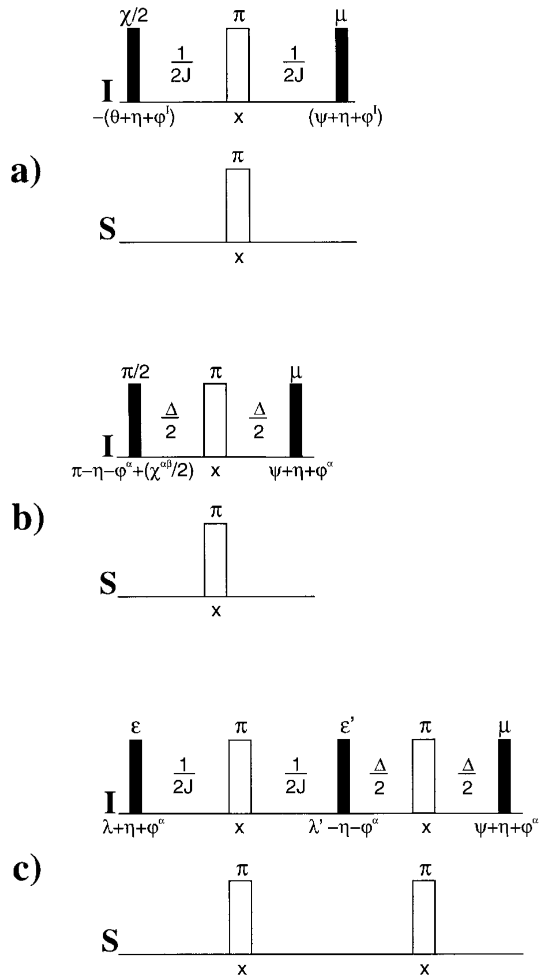

FIG. 2. Pulse sequence elements (a) BIG-BIRD (*4*); (b) α,β TIG-BIRD as expressed in Eq. [23] for arbitrary and independent manipulations of IS^α and IS^β resonances; and (c) full TIG-BIRD as expressed in Eq. [22] for triselective and independent rotations of I, IS^α, and IS^β spin systems. Flip angles and phases are shown above and below the pulses, respectively.

The TIG-BIRD pulse sequence element was tested on a sample of 1% iodomethane with about 60% ^{13}C labeling in $CDCl_3$. A series of one-dimensional spectra shown in Fig. 3 was recorded on a 500-MHz Varian UNITY*plus* spectrometer with different combinations of flip angles (γ^I, γ^α, γ^β) and phases (φ^I, φ^α, φ^β) for I-spin magnetization of I, IS^α, and IS^β spin systems with I = ^1H and S = ^{13}C. These results clearly demonstrate the ability of TIG-BIRD to arbitrarily

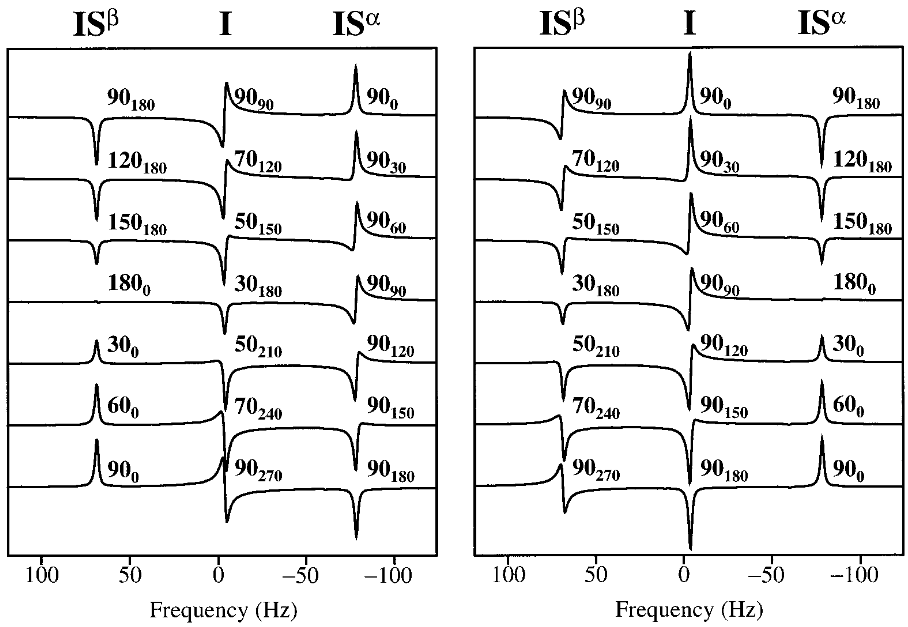

FIG. 3. One-dimensional TIG-BIRD ^{1}H NMR spectra of partially ^{13}C-enriched iodomethane in $CDCl_3$ recorded with different combinations of flip angles and phases for I-spin magnetization of I, IS^{α}, and IS^{β} spin systems. Each spectrum is labeled with its corresponding combination of $(\gamma^{I})_{\varphi^{I}}$, $(\gamma^{\alpha})_{\varphi^{\alpha}}$, and $(\gamma^{\beta})_{\varphi^{\beta}}$. Table 1 lists the flip angles and phases used in the TIG-BIRD pulse sequence element in Eq. [22]. As a note of caution we should mention that the sense of rf pulse phase shift is arbitrary on NMR instruments, as discussed by Levitt (*14*). On the 500-MHz Varian UNITY*plus* spectrometer used to record these spectra, the signs of all cumulative phases in Table 1, $(\lambda + \eta + \varphi^{\alpha})$, $(\lambda' - \eta - \varphi^{\alpha})$, $(\psi + \eta + \varphi^{\alpha})$, had to be inverted, i.e., a phase ϕ implemented as $2\pi - \phi$.

and independently vary the flip angles and phases in rotations for the three classes of protons. Table 1 lists the calculated flip angles and phases used in the pulse sequence elements of Eqs. [22] and [23] to achieve the desired rotations. Each spectrum represents four scans with independent $\{0,\pi\}$ two-step phase cycles on the two refocusing pulses on the I channel. All the spectra were processed in the same way, with the same phase correction and plotted with the same scaling factor.

In conclusion, we have introduced and demonstrated a new pulse sequence element, TIG-BIRD, which represents a non-selective addition to the NMR toolkit and effects the equivalent of three independent selective rotations of arbitrary flip angles and phases for I, IS^{α}, and IS^{β} resonances.

TABLE 1
Flip Angles and Phases in TIG-BIRD for a Series of Flip Angles (γ^{I}, γ^{α}, γ^{β}) and Phases (φ^{I}, φ^{α}, φ^{β}) for I-Spin Magnetization of I, IS^{α}, and IS^{β} Spin Systems

γ^{I}	φ^{I}	γ^{α}	φ^{α}	γ^{β}	φ^{β}	ϵ^{a}	$(\lambda + \eta + \varphi^{\alpha})$	ϵ'^{a}	$(\lambda' - \eta - \varphi^{\alpha})^{a}$	μ	$(\psi + \eta + \varphi^{\alpha})$	$J\Delta$
90°	90°	90°	0°	90°	180°	90°	270°	180°	90°	0°	0°	0.500
70°	120°	90°	30°	120°	180°	35.3°	305.9°	124.9°	44.7°	130.9°	300°	0.385
50°	150°	90°	60°	150°	180°	50.3°	30.5°	139.6°	342.4°	116.6°	330°	0.290
30°	180°	90°	90°	180°	0°	63.9°	172.9°	124.6°	257.3°	90°	0°	0.250
50°	210°	90°	120°	30°	0°	39.7°	251.7°	50.8°	299.1°	63.4°	210°	0.290
70°	240°	90°	150°	60°	0°	54.7°	258.3°	36.0°	268.6°	49.1°	240°	0.385
90°	270°	90°	180°	90°	0°	90°	90°	180°	270°	0°	180°	0.500
90°	0°	90°	180°	90°	90°	67.5°	337.5°	90°	292.5°	0°	180°	0.250
90°	30°	120°	180°	70°	120°	59.2°	39.9°	128.4°	258.3°	43.5°	232.5°	0.212
90°	60°	150°	180°	50°	150°	50.8°	144.3°	139.9°	201.1°	78.7°	249.7°	0.286
90°	90°	180°	0°	30°	180°	45.0°	195.0°	135.0°	165.0°	90°	270°	0.417
90°	120°	30°	0°	50°	210°	49.6°	104.1°	41.8°	93.7°	78.7°	110.3°	0.214
90°	150°	60°	0°	70°	240°	63.7°	128.0°	44.2°	111.4°	43.5°	127.5°	0.288
90°	180°	90°	0°	90°	270°	67.5°	157.5°	90°	112.5°	0°	0°	0.250

[a] If isolated I-spin magnetization is irrelevant (Eq. [23]), $\epsilon = 0°$, $\epsilon' = 90°$, and $\lambda' = \pi + (\chi^{\alpha\beta}/2)$.

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