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Construction of Universal Rotations from Point to Point Transformations

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For a desired range of offsets, universal rotations of arbitrary flip angle can be constructed based on point to point rotations of I_y with half the flip angle. This approach allows, for example, creation of broadband or bandselective refocussing pulses from broadband or bandselective excitation pulses. Furthermore, universal rotations about any axis can be obtained from point-to-point transformations that can easily be optimized using optimal control algorithms. The construction procedure is demonstrated on the examples of a broadband refocussing pulse, a broadband 120°_x rotation and a z-rotation with offset pattern.

Key Words: refocussing pulses; universal pulses; universal rotations; optimal control theory; z-rotations; pattern pulses.

INTRODUCTION

Two important classes of composite and shaped pulses are point-to-point rotations (PP pulses) and universal rotations (UR pulses). PP pulses (also denoted class B2 pulses [1]) are designed to rotate a magnetization vector from a given initial direction as closely as possible to a desired final direction, e.g. from the z axis to the x axis for excitation or from z to $-z$ for inversion pulses. In contrast, UR pulses (also denoted class A pulses [1], constant rotation pulses [2], general rotation pulses [3], plane rotation pulses [4] or simply universal pulses [5]) are designed to induce an effective rotation with a defined direction of the rotation axis and a defined rotation angle not only for a given initial vector orientation but for any arbitrary initial vector. Applications where UR pulses are required include refocusing and mixing pulses in two-dimensional experiments. The *de novo* design of UR pulses is generally assumed to be considerably harder than the design of robust PP pulses. However, here we present a surprisingly simple recipe for constructing a desired UR pulse from a PP pulse with half the flip angle. This allows one to draw from the vast literature on PP pulses [1, 2, 6, 7, 8, 9, 10] and to exploit efficient PP pulse optimization algorithms [11, 12, 13, 14, 15, 16, 17] for the design of UR pulses with unprecedented flexibility.

THEORY

In order to demonstrate the basic construction principle, we first consider the special case of UR rotations around the x axis before turning to general UR pulses with arbitrary rotation axes.

The unitary transformation $U_k(\alpha)$ corresponding to a rotation by angle α around axis k (equal to x, y, or z) is given by

$$U_k(\alpha) = \exp\{-\mathrm{i}\ \alpha\ I_k\}. \qquad (1)$$

We can decompose the rotation operator $U_x(\alpha)$ into two consecutive rotations of angle $\alpha/2$ around the x axis as

$$\begin{aligned} U_x(\alpha) &= U_x(\alpha/2)\, U_x(\alpha/2) \\ &= U_x(\alpha/2)\, \left[U_y(\pi)\, U_x(-\alpha/2)\, U_y^{-1}(\pi)\right] \\ &= \left[U_x(\alpha/2)\, U_y(\pi)\, U_x^{-1}(\alpha/2)\right] U_y^{-1}(\pi), \end{aligned} \qquad (2)$$

where in the second line we have used the well-known relation $U \exp\{A\}\, U^{-1} = \exp\{U A\, U^{-1}\}$ for $A = -\mathrm{i}\,(\alpha/2) I_x$ and $U = U_y(\pi)$ which yields $U_x(\alpha/2) = U_y(\pi)\, U_x(-\alpha/2)\, U_y^{-1}(\pi)$. In the last line we have simply regrouped the operators and written $U_x(-\alpha/2) = U_x^{-1}(\alpha/2)$. But this grouping now represents a rotation by $\alpha/2$ about the x axis, which is applied to the operator I_y in the exponent of $U_y(\pi)$. However, this result can be achieved by *any* PP rotation which has the same net effect, rotating I_y to an angle $\alpha/2$ above the y axis in the y, z plane (ie., $I_y \rightarrow I_y \cos\frac{\alpha}{2} + I_z \sin\frac{\alpha}{2}$). Hence, letting $V(\nu)$ represent the propagator of such a composite (or shaped) PP pulse $\mathbf{V}$, which is applied to a spin with a given offset ν, we can also express $U_x(\alpha)$ as

$$\begin{aligned} U_x(\alpha) &= \left[V(\nu)\, U_y(\pi)\, V^{-1}(\nu)\right] U_y^{-1}(\pi) \\ &= V(\nu)\, \left[U_y(\pi)\, V^{-1}(\nu)\, U_y^{-1}(\pi)\right]. \end{aligned} \qquad (3)$$

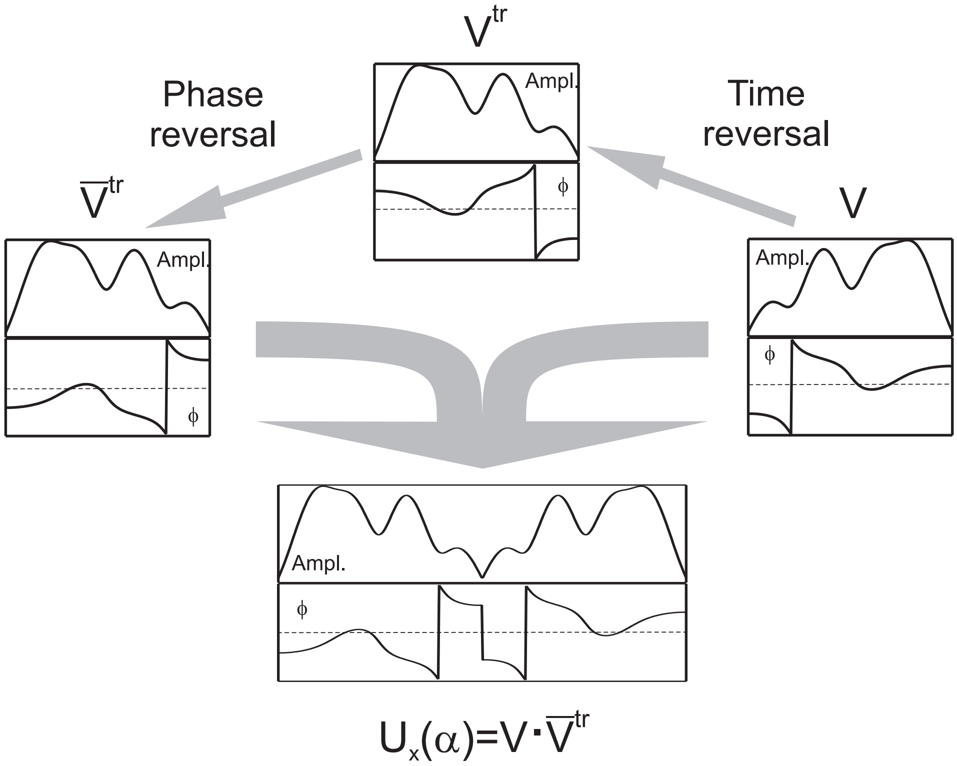

FIG. 1. Demonstration of the construction principle for universal rotation pulses. Starting from a point-to-point transformation pulse $\mathbf{V}$ that transforms I_y magnetization into $I_y \cos\,(\alpha/2) + I_z \sin\,(\alpha/2)$, the time and phase reversed pulse $\overline{\mathbf{V}}^{tr}$ is produced. The combined UR pulse consists of $\overline{\mathbf{V}}^{tr}$ followed by $\mathbf{V}$, effecting the rotation $U_x(\alpha) = V\,\overline{V}^{tr}$. The same procedure can be applied to produce UR pulses around an arbitrary rotation axis (see text for details).

We thus consider how $V^{-1}(\nu)$ transforms under a π rotation about the y axis. The following relations hold for *any* unitary transformation $W(\nu)$ effected by a composite pulse $\mathbf{W}$ at offset ν. For the *time-reversed* pulse $\mathbf{W}^{\rm tr}$, the propagator $W^{\rm tr}(-\nu)$ at offset $-\nu$ is [1]

$$W^{\rm tr}(-\nu) = U_z(\pi)\,W^{-1}(\nu)\,U_z^{-1}(\pi). \qquad (4)$$

For the *phase-reversed* pulse $\overline{\mathbf{W}}$, with the algebraic signs of all phases (expressed in rad or degrees) inverted, the resulting propagator at offset ν is [1]

$$\overline{W}(\nu) = U_x(\pi)\,W(-\nu)\,U_x^{-1}(\pi). \qquad (5)$$

Note that the symmetry relations [1] for phase-reversed pulses as defined above are different from the symmetry relations derived for 180° phase shifted pulses [18], which have been denoted "phase inverted" pulses. As a direct consequence of Eqs. (4) and (5), the propagator $\overline{W}^{\rm tr}(\nu)$ for the time- *and* phase-reversed pulse $\overline{\mathbf{W}}^{\rm tr}$ is given by

$$\begin{aligned}\overline{W}^{\rm tr}(\nu) &= U_x(\pi)\,W^{\rm tr}(-\nu)\,U_x^{-1}(\pi)\\ &= U_x(\pi)\left[U_z(\pi)\,W^{-1}(\nu)\,U_z^{-1}(\pi)\right]U_x^{-1}(\pi)\\ &= U_y(\pi)\;W^{-1}(\nu)\;U_y^{-1}(\pi), \qquad (6)\end{aligned}$$

where we have inserted Eq. (4) in the second line and used $U_x(\pi)\,U_z(\pi) = U_y(\pi)$. As this general relation also holds for the special case $W(\nu) = V(\nu)$, we can finally express Eq. (3) in the form

$$U_x(\alpha) \;=\; V(\nu)\,\overline{V}^{\rm tr}(\nu). \qquad (7)$$

Hence, in a desired range of offsets ν, a UR pulse corresponding to a rotation around the x axis by an angle α can be constructed based on a composite or shaped pulse $\mathbf{V}$ which simply effects a PP rotation from I_y to $(I_y \cos\frac{\alpha}{2} + I_z \sin\frac{\alpha}{2})$ in the desired range of offsets. First, the time and phase reversed PP pulse is applied, followed by $\mathbf{V}$. The phase reversed version of a 90°_y pulse is a 90°_{-y} pulse, where the sign of the pulse phase $\phi = \pi/2$ (corresponding to "y" in the usual short-hand notation) is changed to $\phi = -\pi/2$ (corresponding to "$-y$"). However, the phase reversed version of a 90°_x pulse is also a 90°_x pulse (not a 90°_{-x} pulse), because here $\phi = 0$ (corresponding to "x") remains $\phi = 0$ if the sign of ϕ is reversed. An explicit construction example is provided in Fig. 1.

As shown in Appendix A, it is straightforward to generalize the result of Eq. (7) for UR pulses with rotation angle α and any rotation axis, i.e. with arbitrary azimuthal an-

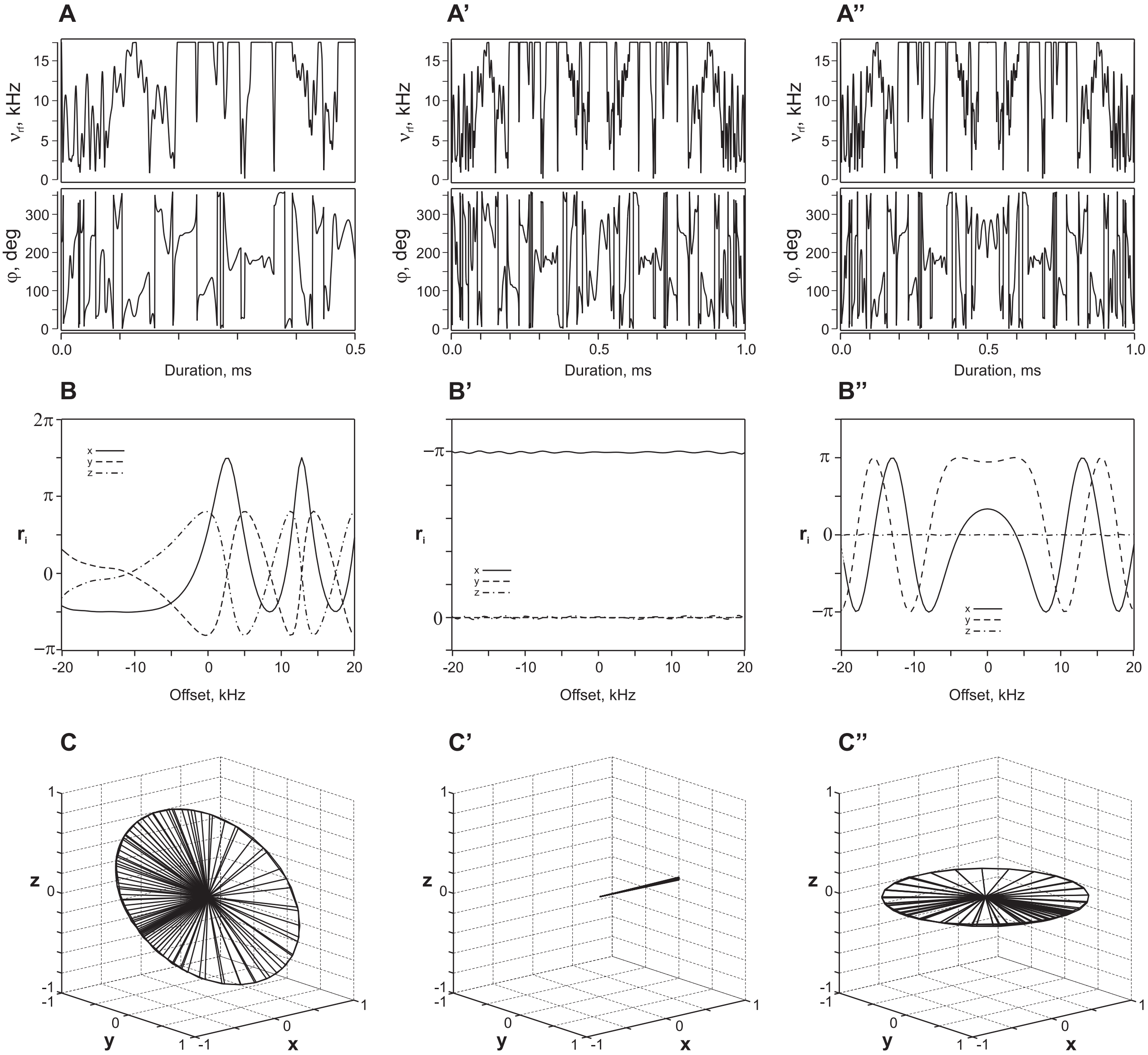

FIG. 2. Example for the construction of a refocussing pulse out of a previously published PP excitation pulse [13]. (A) Amplitude and phase of the original excitation pulse, (A′) the constructed refocussing pulse using the procedure described in Fig. 1, and (A″) a pulse constructed for refocussing using the procedure described in [19]. The corresponding offset profiles of the effective rotations are displayed in B,B′, and B″: x, y, and z components of the rotation vector $\vec{r} = \beta\vec{e}$, where β is the effective rotation angle and $\vec{e}$ is the unit vector pointing along the rotation axis, are given in radians. In C, C′, and C″ the rotation axes $\vec{e}$ are visualized in a 3D-plot for 100 offsets ν equally spaced in the range between ± 20 kHz. A universal 180°_x rotation is achieved in the entire offset range only for the refocussing pulse shown in A′. For excitation pulses the vectors lie in a tilted plane (c.f. Appendix B), while the pulse shown in A″ is a PP inversion pulse with rotation axes in the $x - y$ plane (c.f. Appendix C).

gle θ and phase φ. The construction of such a general UR pulse $U(\alpha, \theta, \varphi)$ can be summarized as follows:

1. Pick (or design [12, 13, 17]) a (composite or shaped) PP pulse $\mathbf{V}$ which effects the rotation

$$\begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \rightarrow \begin{pmatrix} -\sin\frac{\alpha}{2}\cos\theta \\ \cos\frac{\alpha}{2} \\ \sin\frac{\alpha}{2}\sin\theta \end{pmatrix} \tag{8}$$

for a desired range of offsets ν, where the initial vector corresponds to I_y and the final vector corresponds to $(I_y \cos\frac{\alpha}{2} + I_\gamma \sin\frac{\alpha}{2}) = (I_y \cos\frac{\alpha}{2} + I_z \sin\frac{\alpha}{2}\sin\theta - I_x \sin\frac{\alpha}{2}\cos\theta)$, c.f. Appendix A.

2. Construct a combined pulse $\mathbf{U}(\alpha, \theta, 0)$ consisting of the *time-reversed* and *phase-reversed* PP pulse $\overline{\mathbf{V}}^{\rm tr}$ followed by the PP pulse $\mathbf{V}$.

3. Create the desired UR pulse $\mathbf{U}(\alpha, \theta, \varphi)$ by shifting all phases of the individual pulse elements of $\mathbf{U}(\alpha, \theta, 0)$ by φ.

DISCUSSION

As a simple example, consider the construction of a refocussing pulse effecting a universal 180°_x rotation (i.e. a UR pulse with $\alpha = \pi$, $\theta = \pi/2$, and $\varphi = 0$) in the offset range $\nu_{\min} \leq \nu \leq \nu_{\max}$.

According to Eq. (8) in step 1, we first need to find a pulse $\mathbf{V}$ which effects the PP rotation

$$\begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \rightarrow \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \tag{9}$$

i.e. which flips I_y to I_z for the desired range of offsets. Suppose we are given a PP excitation pulse $\mathbf{W}$ which rotates I_z to $-I_y$ in the offset range $-\nu_{\max} \leq \nu \leq -\nu_{\min}$. From Eq. 4, it follows that the time-reversed pulse $\mathbf{W}^{\mathrm{tr}}$ corresponds to a pulse V which effects the required PP transformation from I_y to I_z (c.f. relation [9]) in the desired range $\nu_{min} \leq \nu \leq \nu_{max}$. In step 2, a UR pulse $\mathbf{U}(\pi, \pi/2, 0)$ can be constructed by first applying the pulse $\overline{\mathbf{V}}^{\mathrm{tr}} = \overline{\mathbf{W}}$, (the phase-reversed version of the excitation pulse $\mathbf{W}$), followed by $\mathbf{V} = \mathbf{W}^{\mathrm{tr}}$ (the time-reversed version of the excitation pulse $\mathbf{W}$). As in the given example $\varphi = 0$, step 3 has no effect. This procedure is illustrated in Fig. 2 for the case of a broadband PP excitation pulse $\mathbf{W}$ of 500 μs duration as previously optimized using optimal control theory [13] with $\nu_{\max} = -\nu_{\min} = 20$ kHz. Figure 2 A shows amplitude and phase of the pulse. Note that the pulse phase was shifted by $-\pi/2$ compared to Fig. 1 of [13] in order to effect a z to $-y$ rather than a z to x PP rotation. The combined UR 180°_x pulse consisting of $\overline{\mathbf{V}}^{\mathrm{tr}} = \overline{\mathbf{W}}$ followed by $\mathbf{V} = \mathbf{W}^{\mathrm{tr}}$ is shown in Fig. 2 A′. Figs. 2 B,C and 2 B′,C′ show the effective rotations as a function of offset for the PP pulse $\mathbf{W}$ and for the combined UR 180°_x pulse. As expected, the effected rotations of the constructed UR pulses closely approach the desired rotation in the active range of offsets, where the PP pulse is functional.

Note that the presented approach would yield an exact 180°_x UR pulse if the excitation pulse $\mathbf{W}$ would be perfect. This is in general not the case for a previously suggested approach [19] for the design of 180°_x UR pulses by applying an excitation pulse $\mathbf{W}$ (rather than $\overline{\mathbf{W}}$) followed by the time-reversed excitation pulse $\mathbf{W}^{\mathrm{tr}}$. For comparison, the resulting effective rotations are shown as a function of offset in Fig. 2 B″,C″. While the original excitation pulse has rotation axes distributed in a tilted plane as derived in Appendix B, the composite pulse consisting of the pulse $\mathbf{W}$ followed by $\mathbf{W}^{\mathrm{tr}}$ still shows a variety of rotation axes with respect to offset in the x, y plane, resulting in an inversion pulse (for a detailed derivation see Appendix C). Hence, the approach of Ref. [19] in general does not provide a functional refocussing pulse, but it is equivalent to the approach presented here for the special case of purely amplitude-modulated pulses with phase x or $-x$, where $\mathbf{W} = \overline{\mathbf{W}}$, i.e. if the excitation pulse is invariant under

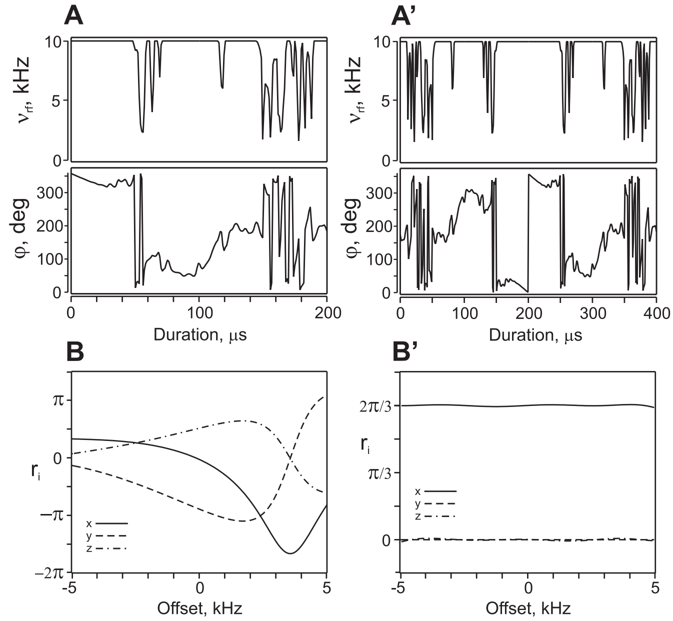

FIG. 3. Example for the construction of a UR pulse corresponding to an effective 120°_x rotation. Using optimal control methods as described in [12, 13, 14, 15], a PP pulse transforming I_y into $1/2 I_y + \sqrt{3}/2 I_z$ for the offset range of ± 5 kHz was optimized (A) and used for the construction of a 120°_x UR pulse (A′) using the procedure described in Fig. 1 and the text. In B and B′ the corresponding components of the rotation vectors $\vec{r}$ are shown as a function of offset.

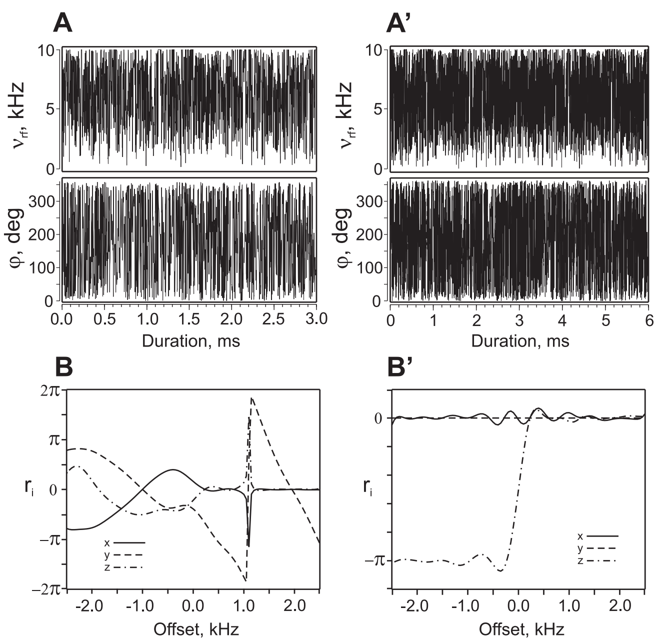

FIG. 4. Example for the construction of a bandselective 180°_z rotation pulse. With methods described in [12, 16] a bandselective PP transformation $I_y \rightarrow I_y$ for an offset range between -2500 Hz and 0 Hz and $I_y \rightarrow I_x$ for offsets between 0 Hz and 2500 Hz was optimized (A) and used for the construction of the bandselective z rotation pulse according to the procedure described in the text and Fig. 1.

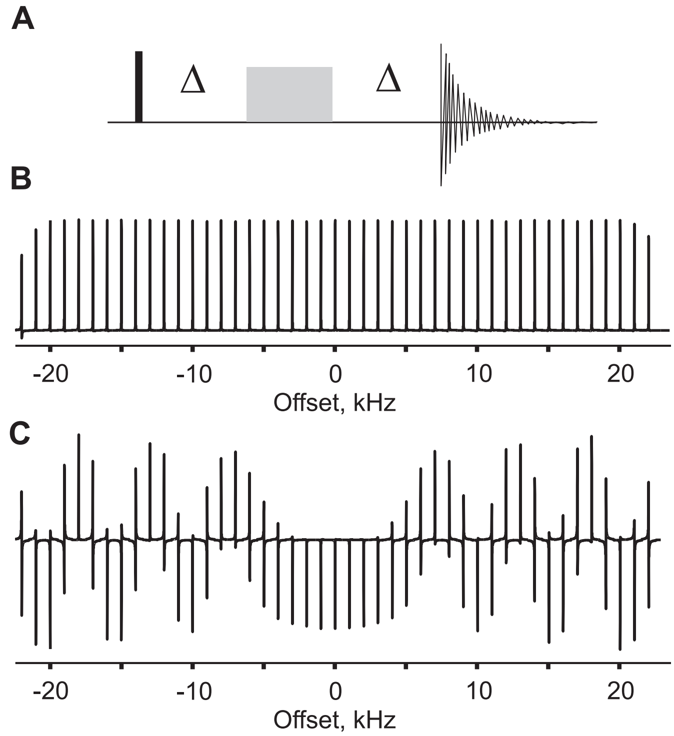

FIG. 5. Experimental validation of the refocussing pulses shown in Fig. 2A′. The pulse sequence uses an echo with delay $\Delta = 100$ ms after excitation (A). The offset of the shaped refocussing pulse was varied in increments of 1 kHz for the range of -22 kHz to 22 kHz with results shown in B. Excellent refocussing properties are obtained for a ±20 kHz offset range, for which the original excitation pulse (Fig. 2A) was optimized. The pulse constructed using the procedure described in [19] (c.f. Fig. 2″) produces the offset profile shown in C.

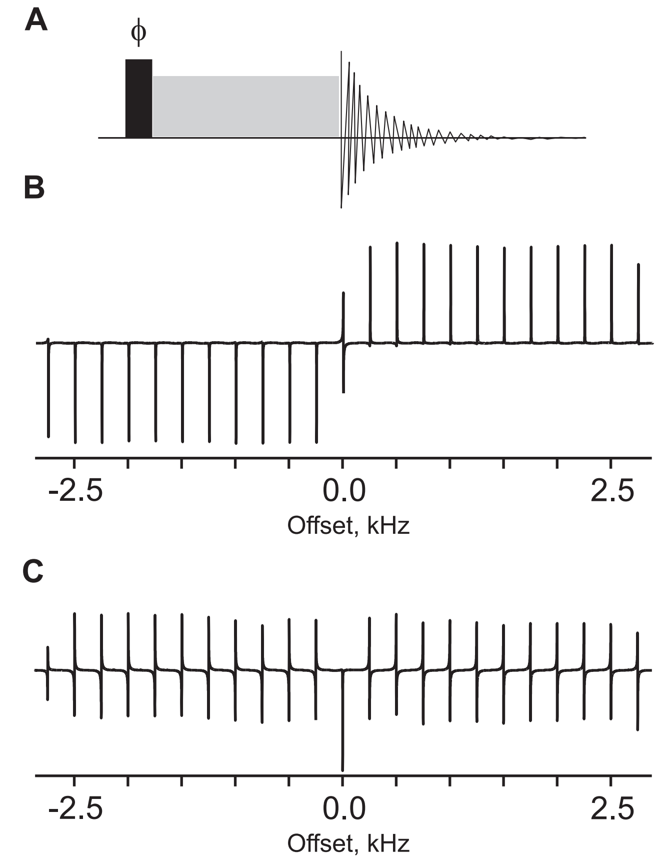

FIG. 6. Experimental validation of the bandselective 180°_z rotation pulse shown in Fig. 4A′. In the pulse sequence the shaped pulse was applied directly after excitation with phase ϕ and rf irradiation frequencies were shifted over an offset range of ±2500 Hz in steps of 250 Hz (A). While negative offset frequencies are basically untouched by the pulse, signals at offset frequencies between 500 Hz and 2500 Hz are inverted independent from the excitation phase ($\phi = x$ in B, $\phi = y$ in C).

phase reversal. Hence, time-symmetric phase-alternating composite 180°_x UR pulses result from phase-alternating 90° PP pulses. Conversely, 90° PP pulses can be obtained from the first half of symmetric phase-alternating composite 180°_x UR pulses, as previously shown in [20].

The general construction principle presented here, based on PP pulses of duration T, always results in UR pulses of duration $2T$ with symmetric rf amplitude, i.e. $\nu^{\rm rf}(t) = \nu^{\rm rf}(2T - t)$ where $\nu^{\rm rf}(t) = \sqrt{(\nu_x^{\rm rf}(t))^2 + (\nu_y^{\rm rf}(t))^2}$. For rotations with $\varphi = 0$, the x component of the rf amplitude $\nu_x^{\rm rf}$ is symmetric, i.e. $\nu_x^{\rm rf}(t) = \nu_x^{\rm rf}(2T - t)$, whereas the y component is antisymmetric with $\nu_y^{\rm rf}(t) = -\nu_y^{\rm rf}(2T - t)$. Note that pulses of this symmetry class are known to give net rotation axes in the xz plane [21].

In the following, two more pulses shall be examined in more detail to give examples for the broad flexibility and applicability of the proposed approach. First, the *de novo* construction of an UR pulse with arbitrary flip angle α shall be demonstrated on the example of a broadband 120°_x rotation. Using the optimal control based algorithm presented in earlier reports [12, 13], we initially optimized a PP pulse for the transfer $I_y \rightarrow \frac{1}{2} I_y + \frac{\sqrt{3}}{2} I_z$ or

$$\begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \rightarrow \begin{pmatrix} 0 \\ 1/2 \\ \sqrt{3}/2 \end{pmatrix}, \qquad (10)$$

effecting a 60°_x rotation for I_y-magnetization in an offset range of ±5 kHz as shown in Fig. 3 A. As expected, the PP pulse provides a variety of rotation axes (Fig. 3B). The UR pulse constructed out of the 60°_x PP pulse (Fig. 3A′), however, performs a uniform 120°_x rotation over the specified offset range (Fig. 3B′).

As a last example, the *de novo* construction of a bandselective 180°_z rotation will show the generality of the approach. The goal was to produce a pulse that has no effect on negative offsets, but works as a 180°_z rotation on positive offsets, which is equivalent to transforming the magnetization components according to $I_x \rightarrow -I_x$, $I_y \rightarrow -I_y$, and $I_z \rightarrow I_z$. An optimal control based algorithm [16] was used first to create a bandselective PP pulse starting from I_y with no rotation for the offset range 0 Hz to 2500 Hz and the transformation $I_y \rightarrow I_x$ for offsets between 0 Hz and -2500 Hz (Fig. 4A). From this PP pulse a UR pulse of twice the duration (Fig. 4A′) was then obtained with the identical procedure used before for the refocussing pulse and the 120°_x rotation. The theoretical performance of the pulse is displayed in Fig. 4B′.

Experimental results for the refocussing pulse (Fig. 2A′) and the bandselective 180°_z rotation (Fig. 4A′) are shown in Figs. 5 and 6. All experiments were acquired on a Bruker Avance 250 spectrometer with linearized amplifiers and SGU400 boards for pulse control using a copper sulfate doped 99.9 % D_2O sample. For validation of the refocussing pulse an echo experiment as shown in Fig. 5A was performed with offsets of the shaped pulse varied from -22 kHz to 22 kHz to cover and exceed its 40 kHz refocussing bandwidth. Over the whole bandwidth the theoretical performance is achieved. For comparison, Fig. 5C shows the effect of the pulse constructed according to [19].

The 180°_z UR pulse was applied directly after excitation with a 500 μs BEBOP pulse [13] in a series of 1D experiments with rf irradiation frequencies ranging from -2500 to 2500 Hz relative to the residual H_2O signal. The bandselective z rotation can be observed by either starting from I_x or I_y, which clearly distinguishes it from conventional 180° pulses with a rotation axis in the transverse plane.

CONCLUSION

A surprisingly simple method for the construction of universal rotation (UR) pulses from point-to-point (PP) pulses is introduced in this article. For a general rotation with an arbitrary flip angle α, a specific PP pulse with flip angle $\alpha/2$ is required. For example, refocussing pulses can be constructed from known excitation pulses in a straightforward way. However, since excitation pulses are usually optimized starting from I_z, they have to be time reversed first for constructing the UR pulse. The resulting refocussing pulses are only twice as long as the initial excitation pulses used for construction. With recent improvements in short broadband excitation pulses [12, 13, 14, 15] relatively short and robust refocussing pulses can be constructed using the presented approach. The proposed construction principle is very general and can be used for obtaining broadband as well as selective, or pattern-type UR pulses. Potential applications of robust pattern-type UR pulses include NMR imaging techniques and NMR-spectroscopy in inhomogeneous B_0-fields [22] and robust local operations in quantum information processing.

APPENDIX A: DECOMPOSITION OF ARBITRARY ROTATIONS INTO PP PULSES

Here we derive the decomposition of a general universal rotation (UR) into two point-to-point (PP) pulses of half the flip angle. A general UR can be characterized by the orientation of the rotation axis and by the rotation angle α around this axis. The orientation of the rotation axis is uniquely defined by the azimuthal angle θ (with $0 \le \theta \le \pi$) and the phase φ (with $0 \le \varphi \le 2\pi$). For the special case, where the rotation axis points along the x axis (i.e. $\theta = \pi/2$ and $\varphi = 0$), the decomposition was derived in the main text (Eqs. (1)-(7)). We first generalize this derivation for arbitrary θ (but still assuming $\varphi = 0$), which corresponds to a rotation axis in the x-z plane. The corresponding unitary operator is

$$U(\alpha,\theta) = \exp\{-\mathrm{i}\ \alpha\ I_\theta\} \tag{11}$$

with

$$I_\theta = I_z \cos\theta + I_x \sin\theta. \tag{12}$$

Using the identities

$$\exp\{-\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\} = \exp\{-\mathrm{i}\ \pi\ I_y\}\ \exp\{\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\}\ \exp\{\mathrm{i}\ \pi\ I_y\} \tag{13}$$

and

$$\exp\{-\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\}\ I_y\ \exp\{\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\} = I_y \cos\frac{\alpha}{2} + I_\gamma \sin\frac{\alpha}{2} \tag{14}$$

with

$$I_\gamma \ = \ \mathrm{i}[I_y, I_\theta] = I_z \sin\theta - I_x \cos\theta, \tag{15}$$

$U(\alpha,\theta)$ can be rewritten as

$$\begin{aligned} U(\alpha,\theta) &= \exp\{-\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\}\ \exp\{-\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\} \\ &= \exp\{-\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\}\ \exp\{-\mathrm{i}\ \pi\ I_y\}\ \exp\{\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\} \\ &\quad \exp\{\mathrm{i}\ \pi\ I_y\} \\ &= \exp\{-\mathrm{i}\ \pi\ (\exp\{-\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\}\ I_y\ \exp\{\mathrm{i}\ \frac{\alpha}{2}\ I_\theta\})\} \\ &\quad \exp\{\mathrm{i}\ \pi\ I_y\} \\ &= \exp\{-\mathrm{i}\ \pi\ (I_y \cos\frac{\alpha}{2} + I_\gamma \sin\frac{\alpha}{2})\} \\ &\quad \exp\{\mathrm{i}\ \pi\ I_y\}. \end{aligned} \tag{16}$$

Given any composite (or shaped) pulse effecting PP rotations $V_\theta(\nu)$ which transform I_y to $(I_y \cos\frac{\alpha}{2} + I_\gamma \sin\frac{\alpha}{2}) = (I_y \cos\frac{\alpha}{2} + I_z \sin\frac{\alpha}{2} \sin\theta - I_x \sin\frac{\alpha}{2} \cos\theta)$ for a range of offsets ν, i.e.

$$V_\theta(\nu)\ I_y\ V_\theta^{-1}(\nu) = I_y \cos\frac{\alpha}{2} + I_\gamma \sin\frac{\alpha}{2}, \tag{17}$$

Eq.[16] can be written in the form

$$\begin{aligned} U(\alpha,\theta) &= \exp\{-\mathrm{i}\ \pi\ (V_\theta(\nu)\ I_y\ V_\theta^{-1}(\nu))\}\ \exp\{\mathrm{i}\ \pi\ I_y\} \\ &= V_\theta(\nu)\ \exp\{-\mathrm{i}\ \pi\ I_y\ \}\ V_\theta^{-1}(\nu)\ \exp\{\mathrm{i}\ \pi\ I_y\} \\ &= V_\theta(\nu)\ \overline{V}_\theta^{\mathrm{tr}}(\nu), \end{aligned} \tag{18}$$

where in the final step we used Eq. (6) with $W(\nu) = V_\theta(\nu)$.

This decomposition of a UR pulse with a rotation axis in the x-z plane (i.e. $\varphi = 0$) can readily be generalized for arbitrary UR pulses with a unitary operator

$$\begin{aligned} U(\alpha,\theta,\varphi) &= \exp\{-\mathrm{i}\ \varphi\ I_z\}\ U(\alpha,\theta)\ \exp\{\mathrm{i}\ \varphi\ I_z\} \\ &= \exp\{-\mathrm{i}\ \varphi\ I_z\}\ V_\theta(\nu)\ \overline{V}_\theta^{\mathrm{tr}}(\nu)\ \exp\{\mathrm{i}\ \varphi\ I_z\} \\ &= \exp\{-\mathrm{i}\varphi I_z\}\ V_\theta(\nu)\ \exp\{\mathrm{i}\varphi I_z\} \\ &\quad \exp\{-\mathrm{i}\varphi I_z\}\ \overline{V}_\theta^{\mathrm{tr}}(\nu)\ \exp\{\mathrm{i}\varphi I_z\}. \end{aligned} \quad (19)$$

Hence, given a composite pulse corresponding to $U(\alpha,\theta) = V_\theta(\nu)\ \overline{V}_\theta^{\mathrm{tr}}(\nu)$ for $\varphi = 0$, the general UR pulse with $U(\alpha,\theta,\varphi)$ can be constructed simply by adding φ to the phase of each individual element of the composite pulse.

APPENDIX B: ROTATION VECTORS FOR PP PULSES

Consider any pair of initial and final vectors $\vec{v}_i$ and $\vec{v}_f$. The angle η between the two vectors is defined by $\cos\eta = \vec{v}_i \cdot \vec{v}_f$ with $0 \le \eta \le \pi$. For a PP rotation from $\vec{v}_i$ to $\vec{v}_f$, the rotation axis must be located in the plane through the origin which is orthogonal to $\vec{v}_i - \vec{v}_f$. The actual rotation angle β (i.e. the length of the rotation vector) depends on the orientation of the unit rotation vector $\vec{e}$ in this plane:

$$\cos\beta = -\frac{1-(1+\cos 2\gamma)\cos 2\frac{\eta}{2}}{1-\sin 2\gamma \cos 2\frac{\eta}{2}},$$

where γ is the angle between the unit vector $\vec{e}$ and the vector $\vec{v}_i \times \vec{v}_f$, with $\cos\gamma = \vec{e}\cdot(\vec{v}_i \times \vec{v}_f)$. The rotation angle β is positive for $|\gamma| \le \pi/2$ and negative for $|\gamma| > \pi/2$. For example, if $\vec{e}$ is parallel or antiparallel to $\vec{v}_i \times \vec{v}_f$ (i.e. $\gamma = 0$ or $\gamma = \pi$), the rotation angle β is η or $-\eta$, respectively. If $\vec{e}$ is colinear to $\vec{v}_i + \vec{v}_f$ (i.e. $\gamma = \pm\pi/2$), the rotation angle β is π.

APPENDIX C: ROTATION VECTORS FOR SYMMETRIZED PULSES ACCORDING TO REFERENCE [19]

Here we analyze the effect of an excitation pulse $\mathbf{W}$ followed by the time-reversed excitation pulse $\mathbf{W}^{\mathrm{tr}}$ as suggested in [19]. We assume that the PP pulse $\mathbf{W}$ transforms I_z to I_γ for a given range of offsets ν, i.e.

$$W(\nu)\ I_z\ W^{-1}(\nu) = I_\gamma, \quad (20)$$

where I_γ is in the transverse plane (e.g. $I_\gamma = I_y$). Subsequent application of $\mathbf{W}^{\mathrm{tr}}$ to I_γ yields

$$\begin{aligned} W^{\mathrm{tr}}(\nu) I_\gamma \{W^{\mathrm{tr}}(\nu)\}^{-1} &= \exp\{-\mathrm{i}\pi I_z\} W^{-1}(-\nu) \exp\{\mathrm{i}\pi I_z\} \\ &\quad I_\gamma \exp\{-\mathrm{i}\pi I_z\} W(-\nu) \exp\{\mathrm{i}\pi I_z\} \\ &= -\exp\{-\mathrm{i}\pi I_z\} W^{-1}(-\nu) \\ &\quad I_\gamma\ W(-\nu) \exp\{\mathrm{i}\pi I_z\}, \end{aligned} \quad (21)$$

where we used Eq. (4) and $\exp\{\mathrm{i}\pi I_z\} I_\gamma \exp\{-\mathrm{i}\pi I_z\} = -I_\gamma$. Eq. (21) can be further simplified, provided the excitation profile of the PP pulse $\mathbf{W}$ is symmetric with respect to offset. In this case, I_z is transformed to I_γ not only for offset ν but also for offset $-\nu$, i.e.

$$W(-\nu)\ I_z\ W^{-1}(-\nu) = I_\gamma. \quad (22)$$

Multiplying Eq. (22) from the left by $W^{-1}(-\nu)$ and from the right by $W(-\nu)$, we find

$$W^{-1}(-\nu)\ I_\gamma\ W(-\nu) = I_z \quad (23)$$

and Eq. (21) can be written as

$$\begin{aligned} W^{\mathrm{tr}}(\nu) I_\gamma \{W^{\mathrm{tr}}(\nu)\}^{-1} &= -\exp\{-\mathrm{i}\pi I_z\}\ I_z\ \exp\{\mathrm{i}\pi I_z\} \\ &= -I_z. \end{aligned} \quad (24)$$

Hence, for an excitation pulse $\mathbf{W}$ which transforms I_z to I_γ for offset ν and $-\nu$, the application of $\mathbf{W}$ followed by $\mathbf{W}^{\mathrm{tr}}$ results in a PP inversion pulse which transforms I_z to $-I_z$. However, in general this combined pulse is not a refocussing pulse because the effective axis of the π rotation can be located anywhere in the transverse plane, c.f. Fig. 2 C″. Only for the special case of a purely amplitude-modulated pulses $\mathbf{W}$ with phase x or $-x$, the resulting symmetric PP pulse is also a UR pulse with a unique rotation axis [23].

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