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Optimized Coherence Selection

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Abstract

A theoretical framework for optimized coherence order selection in regards to cogwheel phase cycling is developed based on analysis of solutions of the associated homogeneous linear Diophantine equation. The empirical Hughes-Carravetta-Levitt conjectures are derived. A general formula is constructed that is guaranteed to provide valid phase cycles. Optimality of this formula is proven for non-sparse sets of coherence levels with relatively prime lower absolute bounds. Smaller phase cycles can be constructed when these conditions do not hold. The resultant formula can be incorporated into NMR spectrometer software to automatically generate cogwheel phase cycles.

Graphical Abstract

Keywords

Cogwheel Phase Cycling; Coherence Order Selection; Linear Diophantine Equations; Mixed Radix Positional Encoding; Lattice Reduction

1. Introduction

Coherence order selection is a method of discriminating components of the density operator in a Nuclear Magnetic Resonance (NMR) experiment by shifting the phase of a coherence

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transfer propagator between successive scans [1, 2]. Briefly, a coherence transfer propagator with phase Φ resulting in a coherence order change p shifts the phase of the coherence component by $\Phi - p$. A coherence transfer pathway can be selected by cycling the phase of sequential coherence transfer propagators.

The NMR experiment consists of M coherence transfer blocks and the total length of the phase cycle is N . Successive steps in the phase cycle are designated $A(n)$. Right subscripts, l , enumerate successive coherence order change blocks. Superscripts, k , enumerate coherence order changes within each block. Coherence order changes intended for selection Δp_l^s exist in a set of coherence order changes Δp_l^k with range $(\Delta p_l^{min} \dots \Delta p_l^{max})$. The total number of pathways is $T \leq \prod_{l=0}^{M-1} \|\Delta p_l^{max} - \Delta p_l^{min}\|$, and individual coherence order changes in a pathway are indexed by k_l .

Provided that all coherence order changes are equally populated, the accumulated signal $A(n)$ at successive steps of the phase cycle is,

$$A(n) = \sum_{k_l=0}^T \exp\left(-i \frac{2\pi n}{N} \sum_{l=0}^{M-1} \phi_l \Delta p_l^{k_l}\right). \quad (1)$$

The coherence order change pathway can be selected by applying a matching phase cycle to the receiver, ϕ_{rec} ,

$$\phi_{rec} = - \sum_{l=0}^{M-1} \phi_l \Delta p_l^s \quad (2)$$

$$N = \sum_{n=0}^{N-1} A(n) \exp\left(-i \frac{2\pi n}{N} \phi_{rec}\right). \quad (3)$$

From this definition, the *receiver* is similar to a Discrete Fourier Transform.

Nesting is a method of cycling the *phase factors* ϕ of sequential coherence transfer propagators,

$$\phi_l = \frac{1}{N_l} \prod_{j=l}^{M-1} N_j \quad (4)$$

$$N_l = \|\Delta p_l^{max} - \Delta p_l^{min}\| \quad (5)$$

$$N = \prod_{l=0}^{M-1} N_l. \quad (6)$$

Nested phase cycling permits selection of all coherence order change pathways and is equivalent to *mixed radix positional number encoding*. Every coherence order change pathway has a unique angular frequency, c_n in $(0 \dots N-1)$,

$$\phi_0 \Delta p_0^{k_0} + \phi_1 \Delta p_1^{k_1} \dots + \phi_{M-1} \Delta p_{M-1}^{k_{M-1}} = c_n. \quad (7)$$

Without this requirement N can be made smaller initially by setting

$$N_l = 1 + \max(\|p_l^{min} - \Delta p_l^0\|, \|p_l^{max} - \Delta p_l^0\|).$$

Cogwheel phase cycling, introduced by Levitt and coworkers, further relaxes the nesting strategy and the requirement that all coherence order change pathways are selectable with the aim of obtaining a smaller phase cycle size, N [3]. This type of mixed radix positional number encoding only requires one pathway to have a unique representation.

Cogwheel phase cycling introduces winding numbers, v_l that modify the nesting strategy, where $\phi_l = v_l$. A winding number is also specified for the receiver, v_{rec} , to ensure selection. In practical NMR applications, the initial coherence order is $p_0 = 0$, and the final is $p_M = -1$. A solution can then be specified with delta winding numbers, $w_l = v_{l-1} - v_l$ [3]. Having defined $p_l = p_{l+1} - p_l$

$$v_0 \Delta p_0 + v_1 \Delta p_1 \dots + v_{M-1} \Delta p_{M-1} = -v_M \quad (8)$$

$$\begin{aligned} v_0(p_1 - 0) + v_1(p_2 - p_1) \dots \\ + v_{M-1}(-1 - p_{M-1}) = -v_M \end{aligned} \quad (9)$$

$$\begin{aligned} p_1(v_0 - v_1) + p_2(v_1 - v_2) \dots \\ + p_{M-1}(v_{M-2} - v_{M-1}) = v_{M-1} - v_M \end{aligned} \quad (10)$$

$$w_1 p_1 + w_2 p_2 \dots + w_{M-1} p_{M-1} = w_M = w_{rec}. \quad (11)$$

The empirical Hughes-Carravetta-Levitt (HCL) conjectures predict the optimal size of the cogwheel phase cycle, N_{pred} [4]. The HCL conjectures apply to the case where the desired

coherence order, $p_l^s = p_l^0$, is selected from a symmetric range, $(-p_l^{max} \dots p_l^{max})$, where p_l^{max} is the maximum coherence order present within each coherence order block. The expression for the optimal delta winding numbers ξ_l is observed to be valid when the numbers q_l are relatively prime [4],

$$q_l = p_l^{max} + 1 - |p_l^0| \quad (12)$$

$$Q = \prod_{l=1}^{M-1} q_l \quad (13)$$

$$\xi_l = \frac{\text{sign}(p_l^0)Q}{q_l} \quad (14)$$

$$N_{pred} = Q + \sum_{l=1}^{M-1} \left(\frac{2Q|p_l^0|}{q_l} \right). \quad (15)$$

Since the introduction of cogwheel phase cycling, progress has focussed on practical applications and computational aspects of generating cogwheel phase cycles, [5, 6, 7, 8]. The present work explores the theoretical rationale for cogwheel phase cycling and the HCL conjectures.

2. Optimized Phase Cycling

2.1. Definitions

For a coherence order block l , the desired coherence order, p_l^s , is selected from a range of coherence orders (p_l^{min}, p_l^{max}) , where $p_l^{min, max}$ are the smallest and largest coherence orders present, respectively. The range (p_l^{min}, p_l^{max}) does not need to be symmetric, and more generally may be sparse, in the sense that some coherence orders may be missing.

Centering each coherence order block about p_l^s introduces the numbers δ_l^k ,

$$\delta_l^k = p_l^k - p_l^s. \quad (16)$$

The range of δ_l^k for each coherence order block l is a set, $g_l = (\delta_l^{\min}, \delta_l^{\max})$, with minimum and maximum values that are the smallest and largest delta coherence orders present, respectively.

To select the $\{0, p_1^s, p_2^s, \dots, p_{M-1}^s, -1\}$ pathway the receiver should be tuned to w_{rec} while the delta winding numbers, w_l and optimal size of the phase cycle, N may be determined in the δ basis. This is true because if a pathway x has a unique value $x \equiv y \pmod{N}$, then $x + c \equiv y + c \pmod{N}$ must also be unique. Otherwise, for some other pathway z , $x + c \equiv z \pmod{N}$, which implies that x is not unique, $x \equiv z - c \pmod{N}$, with c determined up to modulo N .

In the δ basis, the $\{p_1^s, p_2^s, \dots, p_{M-1}^s\}$ pathway has an angular frequency of zero. Hence, the w_l should be chosen to ensure the zero pathway $\{0, 0, \dots, 0\}$ has a unique angular frequency among all available pathways $\{\delta_1^{k_1}, \delta_2^{k_2}, \dots, \delta_{M-1}^{k_{M-1}}\}$. The zero pathway is unique, when it is the only pathway with angular frequency of zero,

$$w_1 \delta_1 + w_2 \delta_2 \dots + w_{M-1} \delta_{M-1} = 0 \Leftrightarrow \delta_l = 0. \quad (17)$$

2.2. Motivation for the Solution

A homogeneous linear diophantine equation with non-negative integer coefficients has a trivial solution, namely all $\delta_l = 0$

$$w_1 \delta_1 \dots + w_{M-1} \delta_{M-1} = 0 \quad w_l \geq 0. \quad (18)$$

For non-trivial solutions, if at least one $\delta_l > 0$, then at least one $\delta_j < 0$. Hence, the trivial solution ($\forall \delta_l = 0$) will be unique, if $\forall \delta_l > 0$ or $\forall \delta_l < 0$. Equivalently, all δ_l can be *prevented* from attaining values of a given sign, as illustrated in Figure 1.

As a correlate, for a given p_l^s it is sufficient to *prevent* δ_l from attaining values in either $(\delta_l^{\min}, -1)$ or $(1, \delta_l^{\max})$, because the signs of the ranges may be inverted by inverting the sign w_l . Preventing all p_l^s from attaining values of a particular sign will ensure that the angular frequency of $\{p_1^s, p_2^s, \dots, p_{M-1}^s\}$ is unique.

In the preceding *preventing* a coherence order change block from attaining a certain value is used as a general concept. Prevention is accomplished by choice of the winding numbers (radix coefficients).

Without loss of generality, consider the solutions of δ_1 based on the choice of $w_{l=1}$, Eqs. 19, 20. In this example the w_l are setwise co-prime, so that greatest common divisor (gcd) of the entire set is 1, (however the gcd of a subset need not be 1). As a consequence of the Bézout Identity, the gcd of $w_{l=1}$ is the smallest value $w_1 \delta_1$ can obtain [9]. Briefly, the Bézout

Identity states that the sum of the products of integers (w_l) and integer constants (δ_l) must be a multiple of the gcd of the integers so the smallest non-zero value of their sum is the gcd.

$$w_1\delta_1 + w_2\delta_2 \cdots + w_{M-1}\delta_{M-1} = 0, \quad (19)$$

$$w_2\delta_2 \cdots + w_{M-1}\delta_{M-1} = -w_1\delta_1. \quad (20)$$

Because the w_l are setwise co-prime, w_1 and $\{w_{l \neq 1}\}$ contain no common factors. therefore, the smallest value of δ_1 is the gcd of $\{w_{l \neq 1}\}$. Consequently, to prevent δ_1 from attaining a range of values, it is sufficient to set the gcd of $w_{l \neq 1}$ to a value greater than maximum value of δ_1 . Based on the previous discussion, either positive or negative values of δ_1 need to be considered to choose the smallest absolute bound to δ_1 .

2.3. Unique Representation of a Pathway

The w_l can be chosen as according to the Chinese Remainder Theorem [9]. Given a set of relatively prime numbers h_l , a number x can be uniquely represented modulo $H = \prod_{l=1}^{M-1} h_l$ by a set of congruence relations $x \equiv a_l \pmod{h_l}$, denoted $x \equiv [a_l]_{h_l}$,

$$x = \sum_{l=1}^{M-1} a_l \frac{H}{h_l} \left[\left(\frac{H}{h_l} \right)^{-1} \right]_{h_l}. \quad (21)$$

In this construction the angular frequency of the pathway is equated to x . The congruence relations are applied to each set g_l and used to define the numbers h_l . The modular inverses $\left[\left(\frac{H}{h_l} \right)^{-1} \right]_{h_l}$ are multiplicative inverses of $\frac{H}{h_l}$ in the modular group $\mathbb{Z}/h_l\mathbb{Z}$. Here the a_l are equated to all the possible values δ_l in the set g_l with zero being unique,

$$\delta_l \equiv [0]_{h_l} \Leftrightarrow \delta_l = 0 \quad \text{for} \quad \delta_l \in g_l. \quad (22)$$

The w_l are equated to the coefficients of a_l in Eq. 21, namely,

$$w_l = \frac{H}{h_l} \left[\left(\frac{H}{h_l} \right)^{-1} \right]_{h_l}. \quad (23)$$

The theorem guarantees that $x = 0$ is unique modulo H , and only represented by $\delta_l = 0$. However, up to this point nothing is gained above the nesting strategy.

Two simplifications are possible in the special case where $\forall a_l = 0$. First, the modular inverses can be removed from the coefficients of the a_l in Eq. 21,

$$x = \sum_{i=1}^{M-1} a_i \frac{H}{h_i}. \quad (24)$$

The construction can be verified as a solution. Because the h_l are relatively prime, $h_l \neq j \mid \frac{H}{h_j}$ and $h_l \nmid \frac{H}{h_l}$.

Iterating on the numbers h_l , the only solution of $x = 0$ is given by all $a_l \equiv 0 \pmod{h_l}$, since for any h_l ,

$$0 = x \equiv \left[\sum_{l=1}^{M-1} a_l \frac{H}{h_l} \right]_{h_l} \quad (25)$$

$$0 = x \equiv \left[a_l \frac{H}{h_l} \right]_{h_l} + \left[\sum_{j \neq l} a_j \frac{H}{h_j} \right]_{h_l} \quad (26)$$

$$0 = x \equiv \left[a_l \frac{H}{h_l} \right]_{h_l} + 0. \quad (27)$$

Note that neither a_l nor $\frac{H}{h_l}$ have any common factors with h_l , thus requiring $a_l = 0$. Therefore the construction is sufficient to protect zero, if $[g_i]_{h_l} = (-\|h_l\| + 1, \|h_l\| - 1)$, because $\delta_l \equiv 0 \pmod{h_l}$ is unique in this range.

The second simplification is possible due to the argument in section 2.2. By analogy, it is necessary only to consider a_l of the same sign as $\frac{H}{h_l}$. Hence the h_l can be chosen so that zero is unique in $\delta_l \equiv 0 \pmod{h_l}$ for either positive or for negative δ_l .

By convention in this work the sign of the h_l is the sign of the δ_l which are prevented from completing the zero sum. This means that positive h_l prevent δ_l from completing the zero sum in the range $[(0 \dots h_l - 1)]_{h_l}$ while negative h_l prevent δ_l from completing the zero sum in the range $[(h_l + 1 \dots 0)]_{h_l}$. The h_l are then defined by the following property:

$$\delta_l \equiv [0]_{h_l} \Leftrightarrow \delta_l = 0 \quad \text{for} \quad \text{sign } h_l = \text{sign } \delta_l \in [g_l]_{h_l}. \quad (28)$$

Using the above definition of h_l leads to the definition of the delta winding numbers, w_l , chosen to maintain the trivial solution of the homogeneous linear equation Eq. 17, unique:

$$w_l = \frac{H}{h_l} = \prod_{j=1}^{M-1} \frac{h_j}{h_l}. \quad (29)$$

As a result, the coherence order pathway $\{0, p_1^s, p_2^s, \dots, p_{M-1}^s, -1\}$ will be unique. This construction is always valid when the chosen h_l are relatively prime. The process is illustrated in Figure 2.

2.4. Absent Pathway Angular Frequencies

From the preceding, w_l are chosen to ensure that the desired pathway $\{0, p_1^s, p_2^s, \dots, p_{M-1}^s, -1\}$ is unique. However, a given set of w_l applies to all coherence order pathways. This is true because any pathway can be expressed in the δ basis.

The set of delta winding numbers w_l based on the numbers h_l creates a neighborhood of protection for every coherence order pathway. If the conditions of the chosen h_l are fulfilled in this δ basis, then the pathway will be unique. The neighborhood of protection for each coherence order is $p_l \pm h_l$. Importantly, the angular frequency of a **unique** pathway that includes an **absent** coherence order will be **absent**.

2.5. Unique Representation Modulo N

A unique representation of zero modulo N requires that all multiples of the angular frequency N are absent. In general the number N is the angular frequency of some pathway $\{p_1^{k_1}, p_2^{k_2}, \dots, p_{M-1}^{k_{M-1}}\}$,

$$N = \sum_{l=1}^{M-1} w_l p_l^{k_l}. \quad (30)$$

The alternative, that there is no pathway with which N can be expressed implies that $\text{gcd}(w_l)$ does not divide N . Note that by the construction in section 2.3 the $\text{gcd}(w_l) = 1$. Furthermore, all multiples of $\text{gcd}(w_l) \cdot N$, should be expressible by some pathway $\{p_1^{k_1}, p_2^{k_2}, \dots, p_{M-1}^{k_{M-1}}\}$.

2.6. Example Construction of N

N can be constructed from the absolute bounds of sets g_l . These numbers are termed u_l and are depicted in Figure 3,

$$u_l = \max(\|\delta_l^{\min} - 1\|, \|\delta_l^{\max} + 1\|). \quad (31)$$

The use of the absolute upper bounds ensures that both positive and negative multiples of N will be absent. Moreover the use of the absolute upper bounds ensures that all multiples of N will be expressed with elements greater than the bounds of g_k which are hence absent, provided that the numbers $w_l u_l$ are the same sign. This arrangement ensures that every multiple kN cannot be expressed with elements of the sets g_k because the coherence orders of the pathway are greater than the upper bounds of the pathway g_k . The numbers u_l can further reduced if $[u_l]_{h_l}$ are absent for positive and negative multiples.

Note that with the w_l chosen as per the modified Chinese Remainder Theorem, any pathway is unique modulo H , within the neighborhood of protection for each coherence order block $p_l^{\chi_l} \pm h_l$. This includes the pathway composed of the numbers $u_l - h_l$ which are within the neighborhood of protection for u_l . This pathway is unique, although it is not necessarily absent. To be absent, one of the pathway elements must be absent.

Setting one of the pathway elements to a u_j ensures that the coherence pathway contains an absent value. The expression for N given M coherence order blocks is then,

$$N_M = w_j u_j + \sum_{l \neq j} w_l (u_l - h_l). \quad (32)$$

Using $w_l = \prod_{i=1}^{M-1} h_i / h_l$, the expression for N of size M is independent of the choice of j ,

$$N_M = \prod_{l=1}^{M-1} h_l \cdot \left(\sum_{l=1}^{M-1} \frac{u_l}{h_l} - M + 2 \right). \quad (33)$$

This equation is the main result of this work. The equation is always valid provided that the h_l are relatively prime. Note that if the smallest possible values of h_l are not relatively prime, they can be increased until they are equivalent to the u_l at which point the expression for N_M becomes equivalent to nesting, $N_M = \prod_{l=1}^{M-1} u_l$.

2.7. Optimality

Restrictions on the underlying sets are a critical consideration for optimality. Consider a construction for two coherence transfer blocks with chosen p_1, p_2 and w_1, w_2 that define N ,

$$N = w_1 p_1 + w_2 p_2, \quad (34)$$

and the degeneracy in p_j by steps in w_j and multiples nN ,

$$nN = w_1(np_1 + kw_2) + w_2(np_2 - kw_1). \quad (35)$$

For the construction to be valid, the numbers

$$(np_1 + kw_2) \notin g_1 \text{ and } (np_2 - kw_1) \notin g_2. \quad (36)$$

If the construction fails, integer solutions of n, k are required, for $\delta_1 \in g_1$ and $\delta_2 \in g_2$:

$$\begin{bmatrix} p_1 & w_2 \\ p_2 & -w_1 \end{bmatrix} \begin{bmatrix} n \\ k \end{bmatrix} = \begin{bmatrix} \delta_1 \\ \delta_2 \end{bmatrix}. \quad (37)$$

In solving this equation, note that the determinant $D = N = w_1 p_1 + w_2 p_2$ is non-zero in practical settings.

$$\begin{bmatrix} n \\ k \end{bmatrix} = \frac{1}{N} \begin{bmatrix} -w_1 & -w_2 \\ -p_2 & p_1 \end{bmatrix} \begin{bmatrix} \delta_1 \\ \delta_2 \end{bmatrix} \quad (38)$$

$$-n = \frac{w_1 \delta_1 + w_2 \delta_2}{w_1 p_1 + w_2 p_2} \quad (39)$$

$$k = \frac{\delta_2 p_1 - \delta_1 p_2}{w_1 p_1 + w_2 p_2} \quad (40)$$

Valid solutions of $n \in \mathbb{Z}_0$ and $k \in \mathbb{Z}$ recapitulate the original requirements, and show the degeneracy in n, k :

$$g_1 \ni np_1 + aw_2 = bp_1 + kw_2 \quad (41)$$

$$g_2 \ni np_2 - aw_1 = bp_2 - kw_1 \quad (42)$$

$$a, b \in \mathbb{Z}. \quad (43)$$

This problem is equivalent to lattice reduction with asymmetric bounds on the solution space.

2.8. Optimality of h_l

The construction of h_l and u_l chosen by the Chinese Remainder Theorem are a *minimal* construction when the sets are not sparse and the h_l are relatively prime. The construction (TC) is *minimal* in the sense that if any element is added to the boundary of sets g_h , then the construction will cease to be valid.

Consider some other construction (OC), with choice of w_l operating on some choice of p_l which construct the optimal N . The method is illustrated for two coherence transfer blocks:

$$w_1 p_1 + w_2 p_2 = 0 \pmod{N}. \quad (44)$$

The restriction of the w_l modulo h_l shows that one of the modular constructions must remain valid - the zero pathway, i.e., the trivial solution must be the only solution,

$$[w_1]_{h_2} p_1 + [w_2]_{h_1} p_2 = 0 \pmod{N} \quad (45)$$

$$(a_1 h_2) p_1 + (a_2 h_1) p_2 = 0 \pmod{N}. \quad (46)$$

However, because the construction is minimal, a consequence of the sets not being sparse, the modular reduction of the $[w_l]_{h_j \neq l}$ cannot protect zero, by inspection.

Hence, the w_l can be expressed as multiples of h_h Eq. 46. This construction (OC) is then equivalent to the TC, provided that the $(a_1 p_1, a_2 p_2)$ pathway is protected for all multiples. However, this simply amounts to a sequential re-ordering of the factors,

$$(h_2)(a_1 p_1) + (a_2 h_1) p_2 = 0 \pmod{N}. \quad (47)$$

Therefore, the h_l are an optimal choice, and the argument above can be repeated for M coherence transfer blocks, using modular restriction $[w_l]_{\prod_{h_j \neq l}}$.

As a consequence the u_l are an optimal choice: consider some sequential reduction in multiples from some large multiple kN where all $p_l > 0$. For any expression of one $p_l \ni g_h$, some pair can be re-expressed so that least one $p_j > g_h$ given degeneracy of p_j by w_j Eq. 36. The limiting case of this process is that the p_l are at the u_l boundary, a multiple which cannot be further reduced, while remaining protected.

Further, in the subsequent section optimality is shown directly through a complementary Frobenius approach and an alternative inductive proof is given in the appendix.

Hence, when the sets are not sparse, and the h_j are relatively prime, the construction using $w_l = \prod_{j=1}^{M-1} \frac{h_j}{h_l}$ (Eq. 33) is optimal. When these conditions do not hold, smaller N can be made from other constructions, as illustrated in Figure 4.

2.9. Frobenius approach

The construction (TC) also can be shown to be optimal through an approach using the Frobenius coin problem, which is illustrated for a case of 2 coherence transfer blocks.

For convenience in notation, consider the problem:

$$w_1\delta_1 + w_2\delta_2 = c_n. \quad (48)$$

The smallest and largest values c_n that can be achieved from the restriction of the sets g_l are:

$$w_1(h_1 - 1) + w_2(h_2 - 1) = -c_{min} \quad (49)$$

$$w_1(u_1 - 1) + w_2(u_2 - 1) = c_{max} \quad (50)$$

$$\text{with } c_{max} \geq |c_{min}|. \quad (51)$$

The optimal N is given by the smallest frequency $c_n = N$ such that every non-zero multiple of N is absent in the range $c_{min} \leq c_n \leq c_{max}$ (note that it is possible that $N = c_{max} + 1$).

By convention consider the expression for $N > 0$ and some number F ,

$$N = c_{max} - F. \quad (52)$$

The range of values of δ_l can be shifted to the range $\{0, \dots, p_l^{max} - p_l^{min}\} = \{0, \dots, \Delta p_l^{max}\}$ by subtracting c_{min} from Eq. 52:

$$Z = N - c_{min} = c_{max} - c_{min} - F \quad (53)$$

$$= w_1(p_1^{\max} - p_1^{\min}) + w_2(p_2^{\max} - p_2^{\min}) - F \quad (54)$$

$$= w_1\Delta p_1^{\max} + w_2\Delta p_2^{\max} - F. \quad (55)$$

Minimizing Z (and thereby minimizing N) is equivalent to finding the largest F that cannot be made by Eq. 48.

The well-known Frobenius coin problem [10] states that every $c_n > F$ can be made with δ_1 and δ_2 drawn from the positive integers, in which the Frobenius number is,

$$F = w_1w_2 - w_1 - w_2. \quad (56)$$

We now assert that every integer c_n satisfying $F < c_n < w_1\Delta p_1^{\max} + w_2\Delta p_2^{\max} - F$ can be made by Eq. 48 with δ_1 in the range $\{0, \dots, \Delta p_1^{\max}\}$.

To see this, consider a number made with $\delta_2 = \Delta p_2^{\max} + mw_1 + k$ outside the allowed range with $m \geq 0$ and $0 < k < w_1$:

$$c_n = w_1\delta_1 + w_2(\Delta p_2^{\max} + mw_1 + k) \quad (57)$$

$$= w_1(\delta_1 + [m+1]w_2) + w_2(\Delta p_2^{\max} + k - w_1). \quad (58)$$

in which $\Delta p_1^{\max} + k - w_1 < \Delta p_2^{\max}$ by construction under the constraint that $w_1 \leq \Delta p_2^{\max}$, allowed by Sec. 2.8; by similar reasoning, $w_2 \leq \Delta p_1^{\max}$.

If $\delta_1 + [m+1]w_2 < \Delta p_1^{\max}$, then c_n is made by values of δ_l in the allowed ranges. Suppose instead that $\delta_1 + [m+1]w_2 = \Delta p_1^{\max} + j$ is outside the allowed range with $j > 0$. Then,

$$c_n = w_1(\delta_1 + [m+1]w_2) + w_2(\Delta p_2^{\max} + k - w_1) \quad (59)$$

$$= w_1(\Delta p_1^{\max} + j) + w_2(2p^{\max} + k - w_1) \quad (60)$$

$$= w_1 \Delta p_1^{max} + w_2 \Delta p_2^{max} - w_1 w_2 + j w_1 + k w_2 \quad (61)$$

$$= w_1 \Delta p_1^{max} + w_2 \Delta p_2^{max} + (j-1)w_1 + (k-1)w_2 - F. \quad (62)$$

The smallest c_n that cannot be made is obtained for $j = k = 1$, satisfying the above relationship.

For completeness, the case $k = 0$ must also be considered:

$$c_n = w_1 \delta_1 + w_2 (\Delta p_2^{max} + m w_1) \quad (63)$$

$$= w_1 (\delta_1 + m w_2) + w_2 \Delta p_2^{max}. \quad (64)$$

Again, if $\delta_1 + m w_2 < \delta_1^{max}$, then c_n is made by values of δ_1 in the allowed ranges. Suppose instead that $\delta_1 + m w_2 = \Delta p_1^{max} + j$ is outside the allowed range with $j > 0$. Then,

$$c_n = w_1 (\delta_1 + m w_2) + w_2 \Delta p_2^{max} \quad (65)$$

$$= w_1 (\Delta p_1^{max} + j) + w_2 \Delta p_2^{max} \quad (66)$$

$$= w_1 \Delta p_1^{max} + w_2 \Delta p_2^{max} + j w_1. \quad (67)$$

The smallest c_n that cannot be made is obtained for $j = 1$, which is greater than c_{max} , again consistent with the above assertion.

Thus, every $c_{min} + F < c_n < c_{max} - F$ can be made with δ_l in the original ranges $\{\delta_l^{min}, \dots, \delta_l^{max}\}$. With the above results, the optimality of the choice of u_1, u_2 is demonstrated:

$$N = c_{max} - F \quad (68)$$

$$= c_{max} - w_1 w_2 + w_1 + w_2 \quad (69)$$

$$= w_1 u_1 + w_2 u_2 - w_1 w_2. \quad (70)$$

2.10. Derivation of the HCL Conjectures

The explicit formula, Equation 33, is equivalent to the Hughes-Carravetta-Levitt conjectures when the range of coherence orders is symmetric with $p_l^{max} = -p_l^{min}$. A derivation is given below with positive p_l^s so that $\|\delta_l^{min}\| > \|\delta_l^{max}\|$. The first candidate h_l is $h_l = \delta_l^{max} + 1$, and the construction is valid if the h_l are relatively prime. The absolute upper bounds are $u_l = -\delta_l^{min} + 1$. The choice of positive u_l provides a positive value of N ,

$$h_l = \delta_l^{max} + 1 = (p_l^{max} - p_l^s) + 1 = q_l \quad (71)$$

$$-u_l = \delta_l^{min} - 1 = -(-p_{l+}^{max} - p_{l+}^s) + 1 \quad (72)$$

$$\prod_{l=1}^{M-1} h_l = \prod_{l=1}^{M-1} q_l = Q \quad (73)$$

$$w_l = \frac{Q}{q_l} = \xi_l. \quad (74)$$

Substituting into the explicit formula, Equation 33, leads to N_{pred}

$$N = \prod_{l=1}^{M-1} h_l \cdot \left(\sum_{l=1}^{M-1} \frac{u_l}{h_l} - M + 2 \right) \quad (75)$$

$$N = \sum_{l=1}^{M-1} u_l \prod_{j=1}^{M-1} \frac{h_j}{h_l} - (M-2) \prod_{l=1}^{M-1} h_l \quad (76)$$

$$N = \sum_{l=1}^{M-1} (p_l^{max} + 1 + p_l^s) \frac{Q}{q_l} - (M-2)Q \quad (77)$$

$$N = \sum_{l=1}^{M-1} (q_l + 2p_l^s) \frac{Q}{q_l} - (M-2)Q \quad (78)$$

$$N = \sum_{l=1}^{M-1} (2p_l^s) \frac{Q}{q_l} + \sum_{l=1}^{M-1} Q - (M-2)Q \quad (79)$$

$$N = Q + \sum_{l=1}^{M-1} \left(\frac{2Qp_l^s}{q_l} \right) = N_{pred}. \quad (80)$$

2.11. FAM-RIACT-FAM Example

The FAM-RIACT-FAM sequence is chosen as an illustrative historical example to demonstrate how the method outlined in this work is applied to generate a valid cogwheel phase cycle. The initial applications of cogwheel phase cycling used numerical searches to obtain a reduced phase cycle size of $N=23$ [3, 11] or, subject to additional assumptions about sparsity, $N=11$, [5] compared to a conventional phase cycle with $N=35$. Without assumptions, the explicit method of Eq. 33 shows how to calculate $N=23$, Figure 5. The FAM-RIACT-FAM pulse sequence consists of three coherence order change blocks ($M=3$), where the desired pathway for selection is $p^s = \{0, 3, 1, -1\}$. The procedure starts by listing all the active coherence orders in each coherence order block. Then the coherence order blocks are centered around the coherence orders desired for selection, in this case (3,1). This is called the δ basis, where the coherence orders desired for selection are now (0,0). In the δ basis, the h_l are the smallest absolute value bounds, namely (1,3); and the u_l are the largest absolute value bounds, namely (7,5). The N is then formed by Eq. 33 as : $N = 3(7/1 + 5/3 - 3 + 2) = 21 + 5 - 3 = 23$. The delta-winding numbers are given by Eq. 29 as : $w_1 = h_2 = 3$ and $w_2 = h_1 = 1$. The receiver should be tuned to the frequency of the desired pathway $w_{rec} = w_1 p_1^s + w_2 p_2^s = 3.3 + 1.1 = 10$. The winding numbers are therefore $v = \{0, -3, -4, -14\}$, which differ by sign convention from the original description, namely COG23{0,3,4;14} [3].

2.12. Conclusion

This work exposes a property of number encoding that has an application to phase cycling in NMR experiments. A theoretical framework for cogwheel phase cycling and a derivation of the Hughes-Carravetta-Levitt conjectures are presented. A simple formula which is

guaranteed to work is presented, Eq. 33. The formula is more general than HCL conjectures, in that it can be applied to cases where the bounds of the coherence order blocks are not symmetric. While smaller N can sometimes be achieved under certain conditions, this formula is guaranteed to provide a valid phase cycle.

Given the similarity of coherence order selection to frequency labelling and the Discrete Fourier Transform, the application of recent advances in sparse signal recovery (i.e. compressed sensing) is a natural extension [12]. These methods may achieve smaller N particularly when sets are sparse, or some coherence orders have small amplitudes [5].

Lastly, due to the computational effort to calculate optimal phase cycles, NMR spectrometer software could be developed to calculate (based on the formula presented herein), a list of valid N from a user-input coherence order change table for selected elements of a pulse program.

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Appendix A. Proof of the choice of u_l by induction

It can be shown by induction that the choice of the pathway in Eqs. 32 and 33 composed of the u_l is optimal when the choice $w_l = \prod_{j=1}^{M-1} \frac{h_j}{h_l}$ is made with relatively prime h_l u_k and the sets g_l are not sparse.

The base case is $M = 2$, where only a single coherence transfer is present and $N_2 = u_1$, since this value is absent for all multiples (positive and negative). Given the prerequisites, the u_l pathway is assumed to lead to the smallest N of size M , namely N_M .

Suppose that N_{M+1} is the frequency of some other absent pathway r_k ,

$$N_{M+1} = \sum_{l=1}^M w_l r_l \quad (\text{A.1})$$

$$N_{M+1} = w_M r_M + \sum_{l=1}^{M-1} w_l r_l \quad (\text{A.2})$$

$$N_{M+1} = \prod_{l \neq M} h_l r_M + h_M R_M. \quad (\text{A.3})$$

R_M is the component of N_{M+1} which is of size M . Because N_{M+1} is an absent frequency, one of the r_l must be absent.

The r_M and R_M must be the same sign. Consider the converse, that they are opposite in sign, and re-write the above equation as,

$$N_{M+1} = \prod_{l \neq M} h_l r_M - h_M R_M \quad (\text{A.4})$$

$$N_{M+1} = \prod_{l \neq M} h_l (r_M - \alpha h_M) - h_M \left(R_M - \alpha \prod_{l \neq M} h_l \right). \quad (\text{A.5})$$

The number α can be used to reduce the values of r_M and R_M . If a number α exists such that $(r_M - \alpha h_M)$ and $(R_M - \alpha \prod_{l \neq M} h_l)$ are opposite sign, then r_M and R_M must be the same sign. Otherwise the sign of $(r_M - \alpha h_M)$ and $(R_M - \alpha \prod_{l \neq M} h_l)$ must be the same for all α .

Set α such that $(r_M - \alpha h_M)$ is not absent, but that $(r_M - (\alpha + 1)h_M)$ is opposite in sign from r_M . This is possible because by construction $h_M \leq u_M$, and u_M is the upper bound of the set, and thus a value of α must exist such that $r_M - \alpha h_M$ is within the set. Equivalently $(R_M - \alpha \prod_{l \neq M} h_l)$ and $(R_M - (\alpha + 1) \prod_{l \neq M} h_l)$ must be opposite sign.

Because R_M is composed of relatively prime w_b , $(R_M - \alpha \prod_{l \neq M} h_l)$ should be expressible with some r'_l to make R'_M . Hence, R'_M and $(R'_M - \prod_{l \neq M} h_l)$ must be opposite sign. This implies that $R'_M - \prod_{l \neq M} h_l$

Note that because N is absent and $(r_M - \alpha h_M)$ is not absent, R'_M must be absent. However, every number between $0 \cdot \prod_{l \neq M} h_l$ can be made from r_l in $-u_l \leq -h_l < r_l < h_l \leq u_l$ because the set $\mathcal{g}_l$ is not sparse. This is due to the modified Chinese Remainder Theorem, where from the congruence relations, the number x is unique modulo $H = \prod h_l$. If a number c cannot be made modulo H , then $x + c \equiv x \pmod{H}$.

Therefore $(R'_M - \alpha \prod_{l \neq M} h_l)$ cannot be absent. As a consequence, r_M and R_M must be the same sign. Because both are positive, the smallest N is made with the smallest values of r_M and R_M .

$$N_{M+1} = w_M r_M + h_M R_M. \quad (\text{A.6})$$

We reduce the representation of the pathway, and set the number α so that $r_M - \alpha h_M$ is not absent, but that $r_M - (\alpha + 1)h_M$ is absent. Again, this is possible because by construction $h_M \leq u_M$.

$$N_{M+1} = w_M(r_M - \alpha h_M) + h_M(R_M + \alpha w_M). \quad (\text{A.7})$$

Because $r_M - \alpha h_M$ is not absent, then $R_M + \alpha w_M = R'_M$ must be absent,

$$N_{M+1} = w_M(r_M - \alpha h_M) + h_MR'_M. \quad (\text{A.8})$$

From the induction hypothesis the smallest value of R'_M is N_M since the reduction can be performed for positive and negative multiples of N_{M+1} .

Note that $r_M - (\alpha - 1)h_M$ is absent by construction. The smallest non-absent value is the upper bound of the set: $u_M = r_M - (\alpha - 1)h_M$. Substituting $u_M - h_M = r_M - \alpha h_M$, leads to the expression for N_{M+1} ,

$$N_{M+1} = w_M(u_M - h_M) + w_R R'_M \quad (\text{A.9})$$

$$N_{M+1} = w_M(u_M - h_M) + w_R N_M \quad (\text{A.10})$$

$$N_{M+1} = \prod_{l=1}^{M-1} h_l(u_M - h_M) + h_M \prod_{l=1}^{M-1} h_l \cdot \left(\sum_{l=1}^{M-1} \frac{u_l}{h_l} - M + 2 \right) \quad (\text{A.11})$$

$$N_{M+1} = \prod_{l=1}^M h_l \left(\frac{u_M}{h_M} - 1 \right) + \prod_{l=1}^M h_l \cdot \left(\sum_{l=1}^{M-1} \frac{u_l}{h_l} - M + 2 \right) \quad (\text{A.12})$$

$$N_{M+1} = \prod_{l=1}^M h_l \cdot \left(\sum_{i=1}^M \frac{u_i}{h_i} - M + 1 \right). \quad (\text{A.13})$$

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Highlights

1. Formal procedure for determining cogwheel phase cycles.
2. Proof that cogwheel phase cycles are optimal under many conditions in NMR.
3. Results allow automated generation of cogwheel phase cycles.

Figure 1: Unique Representation of Zero. Consider the sum of two variables ranging in $(2, -\infty)$ and $(4, -\infty)$. It is sufficient to prevent positive values from being attained so as to keep the representation of the zero-valued sum $(0 + 0)$ unique. All negative values in the ranges may be allowed.

Figure 2:

Determination of h_l in the δ basis. Coherence order blocks are converted to the δ basis, and subsequently h_l are defined for each block. In the above example two h_l (arrows) bound each coherence order set. Because the set is allowed to have missing coherence orders, there may be other h_l , such as $h_l = 3$ for the second coherence order block. Additionally, values beyond the bounds of the coherence order set are also h_l , for example 6 and -4 for the second coherence order block.

Figure 3:

An example of u_l . The u_l (circles) are determined based on the available h_l (arrows). Because the set is allowed to have missing coherence orders, there may be other u_l , such as $u_2 = 3$ for the second coherence order block. In this example, $h_1 = 2$ and $h_2 = 3$, $w_1 = h_2$ and $w_2 = h_1$, $u_1 = 3$ and $u_2 = 3$, and $N = w_1 (u_1 - h_1) + w_2 u_2 = 3 + 6 = 9$. Alternatively, a nesting strategy provides $N = 18$.

Figure 4:

Example showing that the algorithm is not optimal when sets are sparse. Allowing $w_1 = h_2 = 4$ and $w_2 = h_1 = -5$ operating on $u_1 = 1$ and $u_2 = -2$, results in $N = w_1 u_1 + w_2 u_2 = 4 \cdot 1 + -5 \cdot -2 = 14$. However, for this example, it is possible to choose $w_1 = 2$ and $w_2 = 1$ operating on $p_1 = 5$ and $p_2 = 3$, resulting in $N = 13$.

Figure 5:

FAM-RIACT-FAM example. The sequence diagram is adapted from [3]. The pathway intended for selection is $p^s = \{0, 3, 1, -1\}$ (ovals), which is selected from pathways with $p_1 = (-3, 3)$ and $p_2 = (-3, 3)$. The process starts by converting the coherence orders to the δ basis. The h_l (arrows) are calculated in the δ basis: $h_1 = 1$ and $h_2 = 3$, which are relatively prime, allowing the calculation to proceed. The delta-winding numbers are then w_l are $w_1 = h_2 = 3$ and $w_2 = h_1 = 1$. The u_l (circles) are the absolute value bounds, $u_1 = 7$ and $u_2 = 5$. By Eq. 33, $N = 3(7/1 + 5/3 - 3 + 2) = 23$.