# Wigner numbers

Cite as: J. Chem. Phys. **151**, 244122 (2019); https://doi.org/10.1063/1.5135721 Submitted: 07 November 2019 . Accepted: 06 December 2019 . Published Online: 30 December 2019

Wesley D. Allen 问



### ARTICLES YOU MAY BE INTERESTED IN

Adventures in DFT by a wavefunction theorist The Journal of Chemical Physics **151**, 160901 (2019); https://doi.org/10.1063/1.5116338

### In search of Coulson's lost theorem

The Journal of Chemical Physics 151, 151101 (2019); https://doi.org/10.1063/1.5128624

Fantasy versus reality in fragment-based quantum chemistry The Journal of Chemical Physics **151**, 170901 (2019); https://doi.org/10.1063/1.5126216





J. Chem. Phys. **151**, 244122 (2019); https://doi.org/10.1063/1.5135721 © 2019 Author(s).

# Wigner numbers

Cite as: J. Chem. Phys. 151, 244122 (2019); doi: 10.1063/1.5135721 Submitted: 7 November 2019 • Accepted: 6 December 2019 • Published Online: 30 December 2019



### Wesley D. Allen<sup>a)</sup> 问

### AFFILIATIONS

Center for Computational Quantum Chemistry, University of Georgia, Athens, Georgia 30622, USA and Allen Heritage Foundation, Dickson, Tennessee 37055, USA

<sup>a)</sup>Electronic mail: wdallen@uga.edu

### ABSTRACT

All reduced Wigner rotation matrix elements  $d_{M'M}^{J}(\theta)$  can be evaluated very efficiently as a linear combination of either  $\cos(N\theta)$  or  $\sin(N\theta)$  terms as N runs in unit steps from either 0 or  $\frac{1}{2}$  to J. Exact, infinite-precision formulas are derived here for the Fourier coefficients in these  $d_{M'M}^{J}(\theta)$  expressions by finding remarkable analytic solutions for the normalized eigenvectors of arbitrarily large matrices that represent the  $\hat{J}_{Y}$  angular momentum operator in the basis of  $\hat{J}_{Z}$  eigenstates. The solutions involve collections of numbers  $W_{m,n}^{J}$  for  $(m, n) = (J-M, J-N) \in [0, 2J]$  that satisfy the recursion relation  $(m + 1)W_{m+1,n}^{J} - 2(J - n)W_{m,n}^{J} + (2J - m + 1)W_{m-1,n}^{J} = 0$ . These quantities, designated here as *Wigner numbers*, are proved to be integers that exhibit myriad intriguing mathematical properties, including various closed combinatorial formulas, (M, N) sum rules, three separate M-, N-, and J-recursion relations, and a large-J limiting differential equation whose applicable solutions are products of a polynomial and a Gaussian function in the variable  $z = -2^{1/2}(J + 1)^{-1/2}M$ . Accordingly, the Wigner numbers constitute a new thread of mathematics extending outside the context of their immediate discovery. In the midst of the  $W_{m,n}^{J}$  proofs, a class of previously unknown combinatorial summation identities is also found from Wigner number orthonormalization conditions.

Published under license by AIP Publishing. https://doi.org/10.1063/1.5135721

### INTRODUCTION

The theory of angular momentum in quantum mechanics is permeated by the Wigner name. Of particular interest are the Wigner rotation matrices,  $D^{J}_{M'M}(\phi, \theta, \chi) = e^{-iM'\phi} d^{J}_{M'M}(\theta) e^{-iM\chi}$ , which find wide application in fields such as molecular spectroscopy, quantum chemistry, nuclear structure, scattering theory, quantum metrology, geodesy, and the theory of irreducible tensor operators.<sup>1–4</sup> These functions depend on the Euler rotation angles ( $\phi$ ,  $\theta$ ,  $\chi$ ) and are indexed by the quantum numbers for angular momentum  $J \in \{0, \frac{1}{2}, 1, \frac{3}{2}, 2, \frac{5}{2}, 3, ...\}$  and its Z-components  $(M', M) \in \{-J, -J + 1, ...\}$ ..., J - 1, J}. The reduced Wigner matrices  $d^{J}_{M'M}(\theta)$  can be obtained by several established means, including explicit expressions, differential equations, recursion relations, projection methods, and boson operator techniques.<sup>4</sup> New research regarding these matrices continues to be published in recent years.<sup>5-19</sup> The phase conventions for  $d'_{M'M}(\theta)$  vary in the literature and thus warrant careful attention.

Equivalent to the original Wigner formula is the expression<sup>21</sup>

$$d_{M'M}^{J}(\theta) = \sqrt{(J+M')!(J-M')!(J+M)!(J-M)!} \sum_{\sigma=\max[0,-(M'+M)]}^{\min[J-M',J-M]} \times \frac{(-1)^{J-M-\sigma} [\cos(\frac{1}{2}\theta)]^{M'+M+2\sigma} [\sin(\frac{1}{2}\theta)]^{2J-M'-M-2\sigma}}{\sigma!(M+M'+\sigma)!(J-M'-\sigma)!(J-M-\sigma)!},$$
(1)

in which  $\sigma$  is an integer index. The hypergeometric function  $_2F_1$  provides another explicit  $d^{J}_{M'M}(\theta)$  equation,

$$d_{M'M}^{J}(\theta) = \frac{(-1)^{M'-M}}{(M'-M)!} \sqrt{\frac{(J-M)!(J+M')!}{(J+M)!(J-M')!}} \times \left[\cos\left(\frac{1}{2}\theta\right)\right]^{2J+M-M'} \left[\sin\left(\frac{1}{2}\theta\right)\right]^{M'-M} \times {}_{2}F_{1}\left[M'-J, -M-J, M'-M+1; -\tan^{2}\left(\frac{1}{2}\theta\right)\right], \quad (2)$$

which assumes that  $M' \ge M$ . A related expression,

$$d_{M'M}^{J}(\theta) = \sqrt{\frac{(J-M)!(J+M)!}{(J+M')!(J-M')!} \left[\cos\left(\frac{1}{2}\theta\right)\right]^{M+M'} \left[\sin\left(\frac{1}{2}\theta\right)\right]^{M-M'}}{\times P_{J-M}^{(M-M',M+M')}(\cos\theta)},$$
(3)

invokes the Jacobi polynomials  $P_{J-M}^{(M-M^\prime,M+M^\prime)}$  given by

$$P_{m}^{(p,q)}(x) = 2^{-m}(m+p)!(m+q)! \sum_{\sigma=\max[0,-q]}^{\min[m+p,m]} \times \frac{(x+1)^{\sigma}(x-1)^{m-\sigma}}{\sigma!(m+p-\sigma)!(q+\sigma)!(m-\sigma)!},$$
(4)

when *p*, *q*, and *m* are integers. Alternative  $d^{I}_{M'M}(\theta)$  formulas exist that are analogous to Eqs. (2) and (3) but contain no half-argument  $(\frac{1}{2}\theta)$  trigonometric functions.<sup>9</sup>

Two principal recursion relations for reduced Wigner matrices are

$$2(M\cos\theta - M')d^{J}_{M',M}(\theta) = \sqrt{(J-M)(J+M+1)}\sin\theta d^{J}_{M',M+1}(\theta) + \sqrt{(J+M)(J-M+1)}\sin\theta d^{J}_{M',M-1}(\theta)$$
(5)

and

$$(M - M') \cot\left(\frac{1}{2}\theta\right) d^{J}_{M',M}(\theta) = \sqrt{(J + M)(J - M + 1)} d^{J}_{M',M-1}(\theta) + \sqrt{(J + M')(J - M' + 1)} d^{J}_{M'-1,M}(\theta).$$
(6)

Another recursion formula<sup>2,8,16</sup> exploited in the fast multipole method is derived from Eq. (6) in the supplementary material. The  $d_{M'M}^{J}(\theta)$  functions can also be built up iteratively by applying the differential equations,

$$\sqrt{(J \neq M')(J \pm M' + 1)} d^{J}_{M' \pm 1,M}(\theta)$$
  
=  $\csc \theta (M - M' \cos \theta) d^{J}_{M',M}(\theta) \pm \frac{d}{d\theta} d^{J}_{M',M}(\theta)$  (7)

and

$$\sqrt{(J \mp M)(J \pm M + 1)d_{M',M\pm 1}^{J}(\theta)}$$
$$= \csc\theta (M\cos\theta - M')d_{M',M}^{J}(\theta) \mp \frac{d}{d\theta}d_{M',M}^{J}(\theta).$$
(8)

Finally, an elegant but inefficient projection operator scheme gives the entire  $(2J + 1) \times (2J + 1)$  reduced Wigner matrix as

$$\mathbf{d}^{J}(\theta) = \sum_{N=-J}^{J} e^{-iN\theta} \prod_{\substack{N'=-J\\ (\neq N)}}^{J} \left( \frac{\mathbf{J}_{Y}^{(J)} - N'\mathbf{I}^{(J)}}{N - N'} \right), \tag{9}$$

where  $\mathbf{J}_{\mathbf{Y}}^{(J)}$  is the matrix of the  $\hat{J}_{\mathbf{Y}}$  operator in the basis of  $\hat{J}_{\mathbf{Z}}$  eigenstates and  $\mathbf{I}^{(J)}$  is the corresponding identity matrix.

None of the equations given above provides an entirely satisfactory means of ascertaining reduced Wigner rotation matrices for arbitrary *J*. In particular, the original Wigner formula suffers from well-documented catastrophic numerical errors for large values of the angular momentum quantum number because loss of significance occurs when large terms of the opposite sign in Eq. (1) nearly cancel one another.<sup>10,11</sup> To circumvent this large-number problem, recursion algorithms have often been employed. Since these recursion methods also require remedies for numerical instability, a number of procedures have been developed.<sup>8,14–19</sup> Nevertheless, the most effective general approach for  $d^{J}_{M'M}(\theta)$  evaluation appears to be Fourier representation.

ARTICLE

Two recent papers<sup>10,11</sup> on Fourier series for reduced Wigner matrices are especially noteworthy. Tajima<sup>10</sup> derived a closed combinatorial formula for  $d^{J}_{M'M}(\theta)$  Fourier coefficients that involves double summations, but the equation suffers from serious numerical errors in ordinary floating-point calculations. Symbolic manipulation software proved prohibitively time consuming for obtaining exact summation results, and thus, high-precision arithmetic using up to 74 digits for  $J \le 100$  was carried out to obtain practical numerical solutions. The time required to calculate the Fourier coefficients for all  $d'_{M'M}(\theta)$  elements for a given J rose steeply with the angular momentum quantum number, and for J = 100, this task required 43 h of central processing unit (CPU) time on a 3.3 GHz Intel core-i7 3960X processor. Feng and co-workers<sup>11</sup> addressed the Fourier coefficient inefficiency and numerical instability problems via a straightforward approach utilizing standard numerical methods to obtain the eigenvectors of the  $\hat{J}_{Y}$  operator in the basis of  $\hat{J}_{Z}$  eigenstates. They reported accurate evaluation of  $d^{J}_{M'M}(\theta)$  matrices and their derivatives for J values up to a few thousand. Notwithstanding the overall success of the method, a numerical issue persists in that large relative errors occur in peripheral (M', M) cases for which  $d_{M'M}^{I}(\theta)$  is already minute.

As detailed below, the Fourier series approach to  $d'_{M'M}(\theta)$ evaluation is readily apparent from the defining equations for the Wigner rotation matrices. Executing the procedure for very small values of J has been a pedagogical exercise in some graduate quantum mechanics classes for decades. From the perspective of analytic mathematics, extension of the method to practical systems has been blocked by the need to find the eigenvectors of the matrix of the  $\hat{J}_{\rm Y}$  operator. In response, this paper reports exact, analytic solutions for the normalized  $\hat{J}_{\rm Y}$  eigenvectors that hold for any value of J. These solutions involve combinatorial factors and a new class of integers  $W_{m,n}^J$  named here Wigner numbers. Rigorous mathematical proofs and analyses are devoted in this paper to discovering the properties of Wigner numbers, and the wealth of their characteristics gives them a life of their own. A new thread of mathematics is thereby developed that may find application well beyond the exact and universal determination of  $d'_{M'M}(\theta)$ functions.

## WIGNER ROTATION MATRICES FROM $\hat{J}_{\rm Y}$ EIGENVECTORS

The Wigner rotation matrices are defined as matrix elements of the operator  $\hat{R}(\phi, \theta, \chi)$  for Euler-angle rotations in the basis of  $\hat{J}_Z$  eigenstates,

$$D_{M'M}^{J}(\phi,\theta,\chi) = \langle JM' | \hat{R}(\phi,\theta,\chi) | JM \rangle$$
  
=  $\langle JM' | e^{-i\phi\hat{j}_{\chi}} e^{-i\theta\hat{j}_{\chi}} e^{-i\chi\hat{j}_{\chi}} | JM \rangle$   
=  $e^{-iM'\phi} e^{-iM\chi} d_{M'M}^{J}(\theta).$  (10)

The reduced  $d^{J}_{M'M}(\theta)$  functions are given by

$$d_{M'M}^{J}(\theta) = \langle JM' | e^{-i\theta \hat{J}_{Y}} | JM \rangle = \sum_{N=-J}^{J} C_{M',N}^{J} (C_{M,N}^{J})^{*} e^{-iN\theta},$$
(11)

where  $C'_{M,N}$  denotes the eigenvector coefficients for eigenvalue N of the matrix  $\langle JM' | \hat{J}_Y | JM \rangle$ . These coefficients are precisely those computed by Feng *et al.*<sup>11</sup> using standard numerical methods. The largenumber problem in evaluating Wigner reduced rotation matrices is solved by Eq. (11) because the  $C'_{M,N}$  coefficients are elements of normalized eigenvectors, and thus, the magnitude of the summand in these equations cannot exceed unity.

Standard ladder-operator techniques in angular momentum theory  $^{1-3}$  provide the matrix elements,

$$\langle JM | \hat{J}_{Y} | JM' \rangle = \frac{1}{2i} \sqrt{(J - M + 1)(J + M)} \, \delta_{M - 1, M'} - \frac{1}{2i} \sqrt{(J + M + 1)(J - M)} \, \delta_{M + 1, M'},$$
 (12)

and hence the eigenvectors of the  $\hat{J}_{Y}$  matrix satisfy

$$\frac{1}{2i}\sqrt{(J-M+1)(J+M)} C^{J}_{M-1,N} - NC^{J}_{M,N} -\frac{1}{2i}\sqrt{(J+M+1)(J-M)} C^{J}_{M+1,N} = 0.$$
(13)

A recursion procedure for the eigenvector coefficients is established by adding the boundary conditions  $C_{J+1,N}^{I} = C_{-J-1,N}^{I} = 0$ . We require the eigenvectors to be normalized and choose the phases such that if *J* is integral,  $C_{J,N}^{I}$  is positive real, and if *J* is half-integral,  $C_{J,N}^{I}$  is positive imaginary. The  $C_{M,N}^{I}$  coefficients obey the following symmetry properties:

$$C_{M,N}^{J} = (-1)^{M+N-J} C_{-M,N}^{J} = (-1)^{J-M} C_{M,-N}^{J}$$
$$= (-1)^{2J} (C_{M,-N}^{J})^{*} = (-1)^{M+J} (C_{M,N}^{J})^{*}, \qquad (14)$$

as readily seen from Eq. (13).

Equation (14) yields the following  $d_{M'M}^J(\theta)$  symmetry relations straightforwardly:

$$d_{M'M}^{J}(\theta) = d_{-M,-M'}^{J}(\theta) = (-1)^{M-M'} d_{MM'}^{J}(\theta)$$
$$= d_{MM'}^{J}(-\theta) = (-1)^{J+M'} d_{-M',M}^{J}(\theta - \pi).$$
(15)

From Eqs. (11) and (14),

$$d_{M'M}^{J}(\theta) = (-1)^{M+J} \sum_{N=-J}^{J} C_{M',N}^{J} C_{M,N}^{J} e^{-iN\theta},$$
(16)

which can be reduced to trigonometric functions by defining

$$N_0 = \begin{cases} 0 & J \text{ integral,} \\ \frac{1}{2} & J \text{ odd half - integral.} \end{cases}$$
(17)

In particular, Eq. (16) combined with Eq. (14) yields

$$d_{M'M}^{J}(\theta) = (-1)^{M+J} \sum_{N=N_{0}}^{J} \frac{1}{1+\delta_{N,0}} \Big( C_{M',N}^{J} C_{M,N}^{J} e^{-iN\theta} + C_{M',-N}^{J} C_{M,-N}^{J} e^{iN\theta} \Big) \Longrightarrow$$
(18)

$$d_{M'M}^{J}(\theta) = (-1)^{M+J} \sum_{N=N_0}^{J} \frac{C_{M',N}^{J} C_{M,N}^{J}}{1+\delta_{N,0}} \Big[ e^{-iN\theta} + (-1)^{M'-M} e^{iN\theta} \Big]$$
(19)

because J - M must be an integer, whence  $(-1)^{M'+M-2J} = (-1)^{M'-M}$ . From Eq. (19), we conclude that

$$d_{M'M}^{J}(\theta) = 2(-1)^{M+J} \sum_{N=N_0}^{J} \frac{C_{M',N}^{J} C_{M,N}^{J}}{1 + \delta_{N,0}} \cos(N\theta) \quad (M' - M \text{ even})$$
(20)

and

$$d_{M'M}^{J}(\theta) = -2i(-1)^{M+J} \sum_{N=N_0}^{J} C_{M',N}^{J} C_{M,N}^{J} \sin(N\theta) \quad (M' - M \text{ odd}).$$
(21)

### $\hat{J}_X$ AND $\hat{J}_Y$ EIGENVECTORS FROM WIGNER NUMBERS

The eigenvector coefficients can be represented as

$$C_{M,N}^{J} = i^{J-M+2N_0} \alpha_N^{J} W_{J-M,J-N}^{J} \sqrt{(J+M)!(J-M)!}, \qquad (22)$$

in which

$$\alpha_N^J = \frac{1}{2^J \sqrt{(J+N)!(J-N)!}}$$
(23)

is a normalization constant whose form is proved below. The first step to determine the unknown quantities  $W_{J-M,J-N}^{J}$  is to insert Eq. (22) into Eq. (13) and then divide out  $i^{J-M+2N_0}\alpha_N^J$ . Subsequently, all factorial terms can be collected under square roots, and the common factor  $\frac{1}{2}\sqrt{(J+M)!(J-M)!}$  can be factored out.

The resulting recursion formula for  $M \in [-J, J]$  is

$$(J - M + 1) W_{J-M+1,J-N}^{J} - 2NW_{J-M,J-N}^{J} + (J + M + 1) W_{J-M-1,J-N}^{J} = 0,$$
(24)

provided that the boundary conditions

$$W'_{0,J-N} = 1 \text{ and } W'_{-1,J-N} = 0$$
 (25)

are affixed. The associated properties arising from Eq. (14) are that  $W^J_{J-M,J-N}$  must be real and

$$W_{J-M,J-N}^{J} = (-1)^{J-M} W_{J-M,J+N}^{J} = (-1)^{J-N} W_{J+M,J-N}^{J}$$
$$= (-1)^{M+N} W_{J+M,J+N}^{J}.$$
 (26)

The indices in Eq. (24) can be streamlined by designating m = J - Mand n = J - N so that

$$(m+1) W_{m+1,n}^{J} - 2(J-n) W_{m,n}^{J} + (2J-m+1) W_{m-1,n}^{J} = 0$$
(27)

for (m, n) = 0, 1, ..., 2J. This recursion equation is consistent with  $W_{m,n}^J = 0$  whenever *m* lies outside the range [0, 2J]. The  $W_{m,n}^J$ 

J. Chem. Phys. **151**, 244122 (2019); doi: 10.1063/1.5135721 Published under license by AIP Publishing quantities satisfying Eq. (27) are designated here as Wigner numbers. It is transparent from the recursive formula that  $W_{m,n}^{J}$  is always a rational number, but further investigation discussed below reveals that all  $W_{m,n}^{J}$  values are in fact integers.

The matrix elements of the  $\hat{J}_X$  operator are given by

$$\langle JM | \hat{J}_{X} | JM' \rangle = \frac{1}{2} \sqrt{(J - M + 1)(J + M)} \, \delta_{M-1,M'} + \frac{1}{2} \sqrt{(J + M + 1)(J - M)} \, \delta_{M+1,M'}.$$
 (28)

Hence, the eigenvector coefficients corresponding to eigenvalue *P* are determined by

$$\sqrt{(J-M+1)(J+M)} C_{M-1,P}^{J} - 2PC_{M,P}^{J} + \sqrt{(J+M+1)(J-M)} C_{M+1,P}^{J} = 0.$$
 (29)

The same approach employed above for the  $\hat{J}_{\rm Y}$  eigenvectors yields the solution to Eq. (29) as

$$C_{M,P}^{J} = \frac{i^{2N_0} W_{J-M,J-P}^{J}}{2^J} \sqrt{\frac{(J+M)!(J-M)!}{(J+P)!(J-P)!}},$$
(30)

where  $W_{I-M,I-P}^{J}$  is also a Wigner number.

## WIGNER ROTATION MATRICES FROM WIGNER NUMBERS

Because  $(-1)^{2N_0} i^{2J-M'-M} = (-1)^{J-M'+2N_0} i^{M'-M}$ , the product of two eigenvector coefficients from Eq. (22) can be written as

$$C^{J}_{M',N}C^{J}_{M,N} = (-1)^{J-M'+2N_0} i^{M'-M} n^{J}_{M'} n^{J}_{M} c^{J}_{M'M}(N), \qquad (31)$$

where

$$c_{M'M}^{J}(N) = \frac{W_{J-M',J-N}^{J}W_{J-M,J-N}^{J}}{(J+N)!(J-N)!}$$
(32)

and

$$n_M^J = 2^{-J} \sqrt{(J+M)!(J-M)!}.$$
 (33)

If M' - M = 2p is even, then

$$d_{M'M}^{J}(\theta) = 2(-1)^{p} n_{M'}^{J} n_{M}^{J} \sum_{N=N_{0}}^{J} \frac{c_{M'M}^{J}(N)}{1+\delta_{N,0}} \cos(N\theta).$$
(34)

Otherwise, if M' - M = 2p - 1 is odd, then

$$d_{M'M}^{J}(\theta) = 2(-1)^{p} n_{M'}^{J} n_{M}^{J} \sum_{N=N_{0}}^{J} c_{M'M}^{J}(N) \sin(N\theta).$$
(35)

Equations (34) and (35) show that  $d'_{M'M}(\theta)$  can always be represented without approximation as a linear combination of a finite number of either  $\cos(N\theta)$  or  $\sin(N\theta)$  terms. An additional merit is that once this representation is known, then arbitrary-order derivatives of  $d'_{M'M}(\theta)$  can be derived trivially, in contrast to the circumstances of Eq. (1). The Fourier coefficients in the  $d^{J}_{M'M}(\theta)$  expressions are given exactly by Eqs. (32) and (33), averting the need for numerical determination of  $\hat{J}_{Y}$  eigenvectors. The requisite Wigner numbers can be computed very efficiently by the recursion formulas [Eq. (24) or (27)], although explicit closed-form expressions for these integers exist, as derived below. These calculations should be executed using integer arithmetic rather than floating-point operations in order to prevent possible loss of precision. It is apparent from Eqs. (32)–(35) that a convenient and maximally reduced way to exactly express and record Fourier series for the Wigner rotation matrices is

$$d_{M'M}^{J}(\theta) = \sqrt{\rho_0^{J,M'M}} \sum_{N=N_0}^{J} \frac{\rho_{N,1}^{J,M'M}}{\rho_{N,2}^{J,M'M}} f(N\theta),$$
(36)

where all of the  $\rho^{J,M'M}$  quantities are integers and  $f(x) = [\cos(x), \sin(x)]$ , whenever M' - M is (even, odd). In fact, our exhaustive tests lead to the following conjectures for J > 0: the prime factorization of  $\rho_0^{J,M'M}$  contains 2 no more than once,  $\rho_{N,1}^{J,M'M}$  is an odd number, and  $\rho_{N,2}^{J,M'M}$  equals 2 raised to a non-negative integral power less than 2*J*.

For all  $d'_{M'M}(\theta)$  cases with  $J \leq 50$ , our Fourier representations were confirmed to be mathematically identical to those given by *Mathematica* based on Eq. (1);<sup>22</sup> however, onerous trigonometric substitutions implemented within the FullSimplify command of *Mathematica* are necessary to demonstrate this equivalence. In addition, our Fourier coefficients reproduce to standard double precision all the numerical values for  $J \leq 50$  given as supplementary data by Tajima.<sup>10</sup> To illustrate the infinite precision of our method, Table I lists some example terms appearing in Eq. (36) for selected  $d'_{M'M}(\theta)$  cases with J = 80. Although these terms have been maximally reduced, integers are found therein with as many as 44 digits. Some example sets of  $\rho^{J,M'M}$  integers are given in the supplementary material for J = 500, 800, 1000, and 5000; in the latter case, the integers have as many as 2294 digits.

TABLE I. Some exact Fourier contributions [Eq	q. (36)] to selected $d_{\lambda}^{J}$	$f_{MM}(\theta)$ matrix elements for J = 80.
---	--	--

(M', M) = (-14, 68)	$2^{-155}(16084549869640104875)\sqrt{839949057558148542034690372778065}\cos(73\theta)$
(M', M) = (-21, 64)	$2^{-150}(123880549096465021374191311135)\sqrt{20649343335219713852083693347}\sin(31\theta)$
(M', M) = (-80, 78)	$-2^{-154}(50147594357793823553977616732735263859755855)\sqrt{795}\cos(3\theta)$
$(M^{\prime},M)=(1,0)$	$-2^{-153}(8066175552027489352731945121584871325410005)\sqrt{5}\sin(8\theta)$
(M', M) = (-52, 64)	$2^{-152}(42211582681175906077299959132386535235)\sqrt{21288074485}\cos(28\theta)$

#### SUM RULES FOR WIGNER NUMBERS

The  $C_{M,N}^{J}$  coefficients comprise the orthonormal eigenvectors of the Hermitian matrix  $\langle JM' | \hat{J}_{Y} | JM \rangle$ . Thus, the  $C_{M,N}^{J}$  quantities collectively form a unitary matrix; whence,

$$\sum_{M=-J}^{J} C_{M,N'}^{J} (C_{M,N}^{J})^{*} = \delta_{N',N}$$
(37)

and

$$\sum_{N=-J}^{J} C_{M',N}^{J} (C_{M,N}^{J})^{*} = \delta_{M',M}.$$
(38)

Substituting Eq. (22) into Eqs. (37) and (38) provides two sum rules for Wigner numbers,

$$\sum_{M=-J}^{J} W_{J-M,J-N'}^{J} W_{J-M,J-N}^{J} (J+M)! (J-M)! = \frac{\delta_{N,N'}}{\left(\alpha_{N}^{J}\right)^{2}}$$
(39)

and

$$\sum_{N=-J}^{J} W_{J-M',J-N}^{J} W_{J-M,J-N}^{J} (\alpha_{N}^{J})^{2} = \frac{\delta_{M',M}}{(J+M)!(J-M)!}.$$
 (40)

If Eq. (23) is adopted for the normalization constants  $\alpha_N^J$ , these sum rules become

$$\sum_{M=-J}^{J} W_{J-M,J-N'}^{J} W_{J-M,J-N}^{J} (J+M)! (J-M)! = 4^{J} (J+N)! (J-N)! \,\delta_{N',N}$$
(41)

and

$$\sum_{N=-J}^{J} \frac{W'_{J-M',J-N}W'_{J-M,J-N}}{4^{J}(J+N)!(J-N)!} = \frac{\delta_{M',M}}{(J+M)!(J-M)!}.$$
 (42)

### NORMALIZATION AND *N*-RECURSION OF WIGNER NUMBERS

The customary raising  $(\hat{J}_+)$  and lowering  $(\hat{J}_-)$  operators for the eigenstates of  $\hat{J}_Z$  are ubiquitous in angular momentum theory. Cyclic permutation of the (X, Y, Z) axes in the expressions for  $\hat{J}_{\pm}$  yields corresponding ladder operators  $\hat{J}_{\pm}^{(Y)}$  for the  $\hat{J}_Y$  component. Specifically,

$$\hat{J}_{\pm}^{(Y)} = \hat{J}_{Z} \pm i\hat{J}_{X} = \hat{J}_{Z} \pm \frac{1}{2}i\hat{J}_{+} \pm \frac{1}{2}i\hat{J}_{-}, \qquad (43)$$

whose effects on the eigenstates of  $\hat{J}_{\rm Y}$  and  $\hat{J}_{\rm Z}$ , respectively, are given by

$$\hat{f}_{\pm}^{(Y)}|JN\rangle = \sqrt{(J \mp N)(J \pm N + 1)} |J, N \pm 1\rangle$$
(44)

and

$$\hat{J}_{\pm}^{(Y)} | JM' \rangle = M' | JM' \rangle \pm \frac{1}{2} i \sqrt{(J - M')(J + M' + 1)} | J, M' + 1 \rangle$$
  
 
$$\pm \frac{1}{2} i \sqrt{(J + M')(J - M' + 1)} | J, M' - 1 \rangle.$$
 (45)

In the representation of the eigenstates of  $\hat{J}_{\rm Y}$  in the  $|JM'\rangle$  basis,

 $\hat{J}^{(Y)}_{\pm}$  can be applied to both sides via Eqs. (44) and (45) to yield

$$\sum_{M'=-J}^{J} \left\{ \begin{bmatrix} \sqrt{(J \mp N)(J \pm N + 1)}C_{M',N\pm 1}^{J} - M'C_{M',N}^{J} \\ \#\frac{1}{2}iC_{M',N}^{J}\sqrt{(J - M')(J + M' + 1)} \\ \#\frac{1}{2}iC_{M',N}^{J}\sqrt{(J - M')(J - M' + 1)} \\ \#\frac{1}{2}iC_{M',N}^{J}\sqrt{(J + M')(J - M' + 1)} \\ \end{bmatrix} = 0.$$

$$(47)$$

Projection of  $\langle JM |$  onto Eq. (47) produces

$$\sum_{M'=-J}^{J} \left\{ \begin{bmatrix} \sqrt{(J \mp N)(J \pm N + 1)}C_{M',N\pm 1}^{J} - M'C_{M',N}^{J} \end{bmatrix} \delta_{M,M'} \\ \mp \frac{1}{2}iC_{M',N}^{J}\sqrt{(J - M')(J + M' + 1)} \delta_{M,M'+1} \\ \mp \frac{1}{2}iC_{M',N}^{J}\sqrt{(J + M')(J - M' + 1)} \delta_{M,M'-1} \end{bmatrix} = 0.$$
(48)

The Kronecker delta factors in Eq. (48) cause the summation to collapse into three terms, providing an initial *N*-recursion formula,

$$\sqrt{(J \mp N)(J \pm N + 1)C'_{M,N\pm 1}}$$
  
=  $M C^{J}_{M,N} \pm \frac{1}{2}iC^{J}_{M-1,N}\sqrt{(J - M + 1)(J + M)}$   
 $\pm \frac{1}{2}iC^{J}_{M+1,N}\sqrt{(J + M + 1)(J - M)}.$  (49)

Equation (49) can be converted into a Wigner number relation by inserting Eq. (22) for the eigenvector coefficients and dividing out  $\sqrt{(I+M)!(I-M)!}$  from both sides. The result can be written as

$$R_{N,\pm}^{J}W_{J-M,J-N\mp 1}^{J} = MW_{J-M,J-N}^{J} \mp \frac{1}{2}(J-M+1)W_{J-M+1,J-N}^{J} \\ \pm \frac{1}{2}(J+M+1)W_{J-M-1,J-N}^{J},$$
(50)

where

$$R_{N,\pm}^{J} = \sqrt{(J \mp N)(J \pm N + 1)} \left(\frac{\alpha_{N\pm1}^{J}}{\alpha_{N}^{J}}\right) = \frac{\beta_{N\pm1}^{J}}{\beta_{N}^{J}} \left(J \mp N\right)$$
(51)

and

$$\beta_N^J = \alpha_N^J \sqrt{(J+N)!(J-N)!}.$$
(52)

The  $W_{J-M+1,J-N}^{J}$  term can be eliminated from the right side of Eq. (50) via Eq. (24); hence,

$$(J \neq N)\beta_{N\pm 1}^{J}W_{J-M,J-N\mp 1}^{J} = (M \neq N)\beta_{N}^{J}W_{J-M,J-N}^{J}$$
$$\pm (J + M + 1)\beta_{N}^{J}W_{J-M-1,J-N}^{J}.$$
 (53)

In the particular case of M = J and  $N \neq \pm J$ , Eq. (53) yields  $\beta_{N\pm 1}^{J} W_{0,J-N\mp 1}^{J} = \beta_{N}^{J} W_{0,J-N}^{J}$ . Remembering Eq. (25), we conclude that  $\beta_{N\pm 1}^{J} = \beta_{N}^{J} = \beta_{N}^{J}$ , demonstrating that this quantity is independent of *N*. Therefore, the  $\beta^{J}$  terms can be divided out of Eq. (53) to obtain

$$(J \neq N) W_{J-M,J-N+1}^{J} = (M \neq N) W_{J-M,J-N}^{J} \pm (J + M + 1) W_{J-M-1,J-N}^{J}.$$
(54)

By adding the plus and minus variants of Eq. (54), we arrive at a final *N*-recursion formula,

$$(2J-n)W_{m,n+1}^{J}-2(J-m)W_{m,n}^{J}+nW_{m,n-1}^{J}=0,$$
 (55)

where m = J - M and n = J - N as before. According to Eq. (97) presented below,

$$W_{m,0}^{J} = \begin{pmatrix} 2J\\m \end{pmatrix},\tag{56}$$

which is sufficient to start a recursive algorithm based on Eq. (55) with n = 0 because the term involving  $W_{m,-1}^{f}$  vanishes.

To ascertain  $\beta^{J}$ , Eq. (40) can be applied when M' = M = J,

$$\sum_{N=-J}^{J} \left( W_{0,J-N}^{J} \right)^{2} \left( \alpha_{N}^{J} \right)^{2} = \left( \beta^{J} \right)^{2} \sum_{N=-J}^{J} \frac{1}{(J+N)!(J-N)!} = \frac{1}{(2J)!}.$$
 (57)

However, elementary sum rules<sup>23</sup> for binomial coefficients yield

$$\sum_{N=-J}^{J} \frac{1}{(J+N)!(J-N)!} = \frac{1}{(2J)!} \sum_{N=-J}^{J} \binom{2J}{J+N}$$
$$= \frac{1}{(2J)!} \sum_{q=0}^{2J} \binom{2J}{q} = \frac{4^{J}}{(2J)!}.$$
(58)

Combining Eqs. (57) and (58) reveals that  $\beta^J = 2^{-J}$ , and placing this simple result into Eq. (52) proves Eq. (23). This general derivation of the normalization constant is meritorious not merely for elegance because direct attempts to evaluate  $\alpha_N^J$  on a case-by-case basis encounter daunting algebraic complexity.

### WIGNER NUMBERS AS INTEGERS FROM J-RECURSION

Developing a *J*-recursion formula for the Wigner numbers requires evaluation of the change in  $W_{m,n}^J$  occurring when the quantum number *J* is incremented by the minimum allowed amount of  $\frac{1}{2}$ . Specifically, an equation must be found for the quantity,

$$D_{m,n}^{J} = W_{m,n}^{J+\frac{1}{2}} - W_{m,n}^{J}.$$
 (59)

Starting with the *M*-recursion formula for  $W_{m,n}^{J+\frac{1}{2}}$  obtained by making the replacement  $J \rightarrow J + \frac{1}{2}$  in Eq. (27) and then employing Eq. (59), we arrive at

$$(m+1) W_{m+1,n}^{J} - (2J+1-2n) W_{m,n}^{J} + (2J+2-m) W_{m-1,n}^{J} + (m+1) D_{m+1,n}^{J} - (2J+1-2n) D_{m,n}^{J} + (2J+2-m) D_{m-1,n}^{J} = 0.$$
(60)

Furthermore, Eq. (27) can be utilized again to eliminate  $W_{m+1,n}^{J}$  from Eq. (60) and ascertain

$$W_{m-1,n}^{J} - W_{m,n}^{J} + (m+1) D_{m+1,n}^{J} - (2J+1-2n) D_{m,n}^{J} + (2J+2-m) D_{m-1,n}^{J} = 0.$$
(61)

An incisive rearrangement of terms in Eq. (61) gives

$$m D_{m+1,n}^{J} - (2J - 2n) D_{m,n}^{J} + (2J + 2 - m) D_{m-1,n}^{J}$$
  
=  $D_{m,n}^{J} - W_{m-1,n}^{J} - D_{m+1,n}^{J} + W_{m,n}^{J}.$  (62)

The right side of Eq. (62) vanishes manifestly if  $D_{m,n}^{J} = W_{m-1,n}^{J}$  for all *m*; moreover, this equivalence also causes the left side of Eq. (62) to vanish as a consequence of Eq. (27). Hence, a remarkably simple solution is discovered for *J*-recursion,

ARTICLE

$$W_{m,n}^{J+\frac{1}{2}} = W_{m,n}^{J} + W_{m-1,n}^{J},$$
(63)

which is valid for  $m \in [0, 2J + 1]$  and  $n \in [0, 2J]$ , recognizing that  $W_{-1,n}^{J} = W_{2J+1,n}^{J} = 0$ . An additional *J*-recursion equation is required to completely

An additional *J*-recursion equation is required to completely determine all  $W_{m,n}^{J+\frac{1}{2}}$  values because the n = 2J + 1 case is not covered by Eq. (63). For this purpose, we go to Eq. (54), make the replacement  $J \rightarrow J + \frac{1}{2}$ , and choose the lower sign to find that

$$(2J+1-n)W_{m,n+1}^{J+\frac{1}{2}} = (2J+1-m-n)W_{m,n}^{J+\frac{1}{2}} - (2J-m+2)W_{m-1,n}^{J+\frac{1}{2}}.$$
(64)

Equation (63) can be used to eliminate  $W_{m,n}^{J+\frac{1}{2}}$  and  $W_{m-1,n}^{J+\frac{1}{2}}$  from the right side of Eq. (64), providing

$$(2J+1-n)W_{m,n+1}^{J+\frac{1}{2}} = (2J+1-m-n)W_{m,n}^{J} - (n+1)W_{m-1,n}^{J} - (2J-m+2)W_{m-2,n}^{J}.$$
(65)

According to Eq. (27), the replacement  $(2J - m + 2) W_{m-2,n}^J \rightarrow (2J - 2n) W_{m-1,n}^J - m W_{m,n}^J$  can be made on the right side of Eq. (65), and subsequent extensive cancellation of terms reveals that

$$W_{m,n+1}^{J+\frac{1}{2}} = W_{m,n}^{J} - W_{m-1,n}^{J},$$
(66)

with the same (m, n) ranges of validity as in Eq. (63). Setting n = 2J in Eq. (66) provides the quantities  $W_{m,2J+1}^{J+\frac{1}{2}}$  that are not given by Eq. (63).

The complementary recursion relations of Eqs. (63) and (66) allow all Wigner numbers to be built up by successive additions from the root quantity  $W_{0,0}^0 = 1$ . This fact proves that all Wigner numbers are integers, a nontrivial conclusion from the perspective of the defining condition alone [Eq. (27)]. To illustrate the characteristics of Wigner numbers, arrays of  $W_{m,n}^J$  values are given in Table II for J = 5 and  $J = \frac{11}{2}$ , while more extensive tabulations appear in the supplementary material for all allowed *J* values between 0 and 12.

### COMBINATORIAL FORMULAS FOR WIGNER NUMBERS

The first combinatorial equation proved here for the Wigner numbers takes the form

$$W_{m,J-N}^{J} = \sum_{q=0}^{m} {\binom{N}{q}} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q) \right\rfloor} A_{q,p}^{m} {\binom{J}{p}},$$
(67)

in which the  $A_{q,p}^m$  coefficients are chosen to satisfy the defining recursion relation [Eq. (27)]. For convenience in formal manipulations,  $A_{q,p}^m$  can be set to 0 if either of the subscripts lies outside the ranges  $0 \le q \le m$  and  $0 \le p \le \lfloor \frac{1}{2}(m-q) \rfloor$ . Inserting Eq. (67) into Eq. (27) gives

т <u>г</u>													
) = 5		0	1	2	3	4	n 5	6	7	8	9	10	
	0	1	1	1	1	1	1	1	1	1	1	1	
	1	10	8	6	4	2	0	-2	-4	-6	-8	-10	
	2	45	27	13	3	-3	-5	-3	3	13	27	45	
	3	120	48	8	-8	-8	0	8	8	-8	-48	-120	
	4	210	42	-14	-14	2	10	2	-14	-14	42	210	
т	5	252	0	-28	0	12	0	-12	0	28	0	-252	
	6	210	-42	-14	14	2	-10	2	14	-14	-42	210	
	7	120	-48	8	8	-8	0	8	-8	-8	48	-120	
	8	45	-27	13	-3	-3	5	-3	-3	13	-27	45	
	9	10	-8	6	-4	2	0	-2	4	-6	8	-10	
	10	1	-1	1	-1	1	-1	1	-1	1	-1	1	
$J = \frac{11}{2}$							п						
Ξ.		0	1	2	3	4	5	6	7	8	9	10	11
	0	1	1	1	1	1	1	1	1	1	1	1	1
	1	11	9	7	5	3	1	-1	-3	-5	-7	-9	-11
	2	55	35	19	7	-1	-5	-5	-1	7	19	35	55
	3	165	75	21	-5	-11	-5	5	11	5	-21	-75	-165
	4	330	90	-6	-22	-6	10	10	-6	-22	-6	90	330
т	5	462	42	-42	-14	14	10	-10	-14	14	42	-42	-462
	6	462	-42	-42	14	14	-10	-10	14	14	-42	-42	462
	7	330	-90	-6	22	-6	-10	10	6	-22	6	90	-330
	8	165	-75	21	5	-11	5	5	-11	5	21	-75	165
	9	55	-35	19	-7	-1	5	-5	1	7	-19	35	-55
	10	11	-9	7	-5	3	-1	-1	3	-5	7	-9	11

TABLE II. Array	s of Wigner numbers	$W'_{m,n}$ for $J = 5$ and $\cdot$	<u>.</u>
-----------------	---------------------	------------------------------------	----------

$$(m+1)\sum_{q=0}^{m+1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q+1) \right\rfloor} A_{q,p}^{m+1} \binom{J}{p} - (m-1)\sum_{q=0}^{m-1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q-1) \right\rfloor} A_{q,p}^{m-1} \binom{N}{q} = 2\sum_{q=0}^{m} N\binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q) \right\rfloor} A_{q,p}^{m} \binom{J}{p} - 2\sum_{q=0}^{m-1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q-1) \right\rfloor} A_{q,p}^{m-1} J\binom{J}{p}.$$
(68)

The general theorem

$$Z\begin{pmatrix} Z\\r \end{pmatrix} = r\begin{pmatrix} Z\\r \end{pmatrix} + (r+1)\begin{pmatrix} Z\\r+1 \end{pmatrix}$$
(69)

can be used for both (Z, r) = (N, q) and (Z, r) = (J, p) on the right side of Eq. (68). Thus,

$$(m+1) \sum_{q=0}^{m+1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q+1) \right\rfloor} A_{q,p}^{m+1} \binom{J}{p} - (m-1) \sum_{q=0}^{m-1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q-1) \right\rfloor} A_{q,p}^{m-1} \binom{J}{p}$$
$$= 2 \sum_{q=0}^{m} \left[ q\binom{N}{q} + (q+1)\binom{N}{q+1} \right] \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q) \right\rfloor} A_{q,p}^{m} \binom{J}{p}$$
$$- 2 \sum_{q=0}^{m-1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q-1) \right\rfloor} A_{q,p}^{m-1} \left[ p\binom{J}{p} + (p+1)\binom{J}{p+1} \right],$$
(70)

which can be rearranged to

$$\sum_{q=0}^{m+1} \binom{N}{q} \sum_{p=0}^{\lfloor \frac{1}{2}(m-q+1) \rfloor} \binom{J}{p} [(m+1)A_{q,p}^{m+1} - (m-1)A_{q,p}^{m-1} - 2qA_{q,p}^{m} + 2pA_{q,p}^{m-1}]$$

$$= 2\sum_{q=0}^{m} (q+1)\binom{N}{q+1} \sum_{p=0}^{\lfloor \frac{1}{2}(m-q) \rfloor} A_{q,p}^{m} \binom{J}{p} - 2\sum_{q=0}^{m-1} \binom{N}{q} \sum_{p=0}^{\lfloor \frac{1}{2}(m-q-1) \rfloor} A_{q,p}^{m-1} (p+1)\binom{J}{p+1}.$$
(71)

Finally,

$$\sum_{q=0}^{m+1} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q+1) \right\rfloor} \binom{J}{p} \left[ (m+1)A_{q,p}^{m+1} - (m-1)A_{q,p}^{m-1} - 2qA_{q,p}^{m} - 2qA_{q-1,p}^{m} + 2pA_{q,p}^{m-1} + 2pA_{q,p-1}^{m-1} \right] = 0$$
(72)

is obtained by reindexing the summations on the right side of Eq. (71) and employing the aforementioned  $A_{q,p}^m$  boundaries to bring all summation limits into confluence.

In order to satisfy Eq. (72) for all N and J, the  $A_{q,p}^m$  coefficients must obey

$$(m+1)A_{q,p}^{m+1} = (m-2p-1)A_{q,p}^{m-1} + 2q(A_{q,p}^m + A_{q-1,p}^m) - 2pA_{q,p-1}^{m-1}.$$
 (73)

This condition can be simplified by making the substitution

$$A_{q,p}^{m} = 2^{q} (-1)^{p} B_{m-2p,q}$$
(74)

to obtain

$$(s+1)B_{s+1,q} = (s-1)B_{s-1,q} + q(2B_{s,q} + B_{s,q-1}),$$
 (75)

where s = m - 2p. It is straightforward to show from the definition of binomial coefficients that the desired solution to Eq. (75) is

$$B_{s,q} = \binom{s-1}{q-1}.$$
(76)

Therefore, for  $0 \le q \le m$  and  $0 \le p \le \left| \frac{1}{2}(m-q) \right|$ ,

$$A_{q,p}^{m} = 2^{q} (-1)^{p} \binom{m-2p-1}{q-1}.$$
 (77)

In summary, the Wigner numbers are given explicitly by the combinatorial formula,

$$W_{mJ-N}^{J} = \sum_{q=0}^{m} 2^{q} \binom{N}{q} \sum_{p=0}^{\left\lfloor \frac{1}{2}(m-q) \right\rfloor} (-1)^{p} \binom{m-2p-1}{q-1} \binom{J}{p}, \quad (78)$$

which works even for negative *N* and half-integral *J*. In fact, Eq. (78) is valid for continuous values of *N*; hence, it allows  $W_{m,J-N}^{J}$  to be differentiated with respect to *N*. The q = 0 terms in the summation

for 
$$W_{m,J-N}^J$$
 are not problematic because  $\begin{pmatrix} m-2p-1\\-1 \end{pmatrix} = \delta_{m,2p}$ .

The order of summations in Eq. (78) can be reversed upon careful analysis of the index ranges to obtain a second combinatorial formula,  $W_{m,J-N}^{J} = \sum_{p=0}^{\left\lfloor \frac{1}{2}m \right\rfloor} (-1)^{p} {J \choose p} \sum_{q=0}^{m-2p} 2^{q} {N \choose q} {m-2p-1 \choose q-1}.$  (79)

Moreover, as proved in the supplementary material,

$$\sum_{q=0}^{s} 2^{q} \binom{N}{q} \binom{s-1}{q-1} = (2N)_{2} F_{1}(1-N,1-s,2,2) = 2P_{N-1}^{(1,-N-s)}(-3)$$
(80)

for s > 0, where  ${}_{2}F_{1}$  denotes the (Gaussian) hypergeometric function and  $P_{m}^{(p,q)}(x)$  is a Jacobi polynomial. Therefore, Eq. (79) yields a third combinatorial relation that is best represented by the following pair of equations:

$$W_{2k,J-N}^{J} = (-1)^{k} {J \choose k} + 2 \sum_{p=0}^{k-1} {J \choose p} (-1)^{p} P_{N-1}^{(1,2p-2k-N)} (-3)$$
(81)

and

$$W_{2k+1,J-N}^{J} = 2\sum_{p=0}^{k} {J \choose p} (-1)^{p} P_{N-1}^{(1,2p-2k-N-1)} (-3), \qquad (82)$$

in which  $k \in [0, J]$  and  $k \in [0, J - 1]$  for the even and odd *m* cases, respectively.

These combinatorial formulas can be profitably reduced for cases in which N is a small integer. Such results generated from Eqs. (81) and (82) can be cast into the following forms:

$$W_{2k,J-N}^{J} = (-1)^{k} {\binom{J}{k}} \left\{ 1 - \frac{2k\varepsilon_{N+1}}{J+\delta_{J,0}} - \frac{K(J-N)!}{2J!} \times \left[ (J-2k)\varepsilon_{N+1} + \varepsilon_{N} \right] P_{\mathrm{E},N}(J,K) \right\}$$
(83)

and

$$W_{2k+1,J-N}^{J} = 2(-1)^{k} \binom{J-1}{k} \left\{ N \left( 1 - \frac{2k\varepsilon_{N}}{J-1+\delta_{J,1}} \right) - \frac{L(J-N)!}{(J-1)!} \left[ (J-2k-1)\varepsilon_{N} + \varepsilon_{N+1} \right] P_{O,N}(J,L) \right\}, \quad (84)$$

J. Chem. Phys. **151**, 244122 (2019); doi: 10.1063/1.5135721 Published under license by AIP Publishing **TABLE III.** Polynomials  $P_{E,N}(J, K)$  and  $P_{O,N}(J, L)$  specifying the Wigner numbers  $W_{m,J-N}^{J}$  for N = 0-9.

N	$P_{\mathrm{E},N}(J,K)$	$P_{\mathrm{O},N}(J,L)$
0	0	0
1	0	0
2	1	0
3	2	1
4	$4(J^2 - J + 2) - K$	2
5	$2(3J^2-5J+20)-2K$	$5J^2 - 19J + 28 - L$
6	$9J^4 - 30J^3 + 191J^2 - 138J + 184$ $-2K(3J^2 - 3I + 20) + K^2$	$8(J^2-5J+12)-2L$
7	$\begin{array}{c} 4(3J^4 - 14J^3 + 133J^2 - 154J + 392) \\ -2K(5J^2 - 7J + 70) + 2K^2 \end{array}$	$2(7J^4 - 70J^3 + 361J^2 - 762J + 696) -L(7J^2 - 29J + 82) + L^2$
8	$\frac{16(J^6 - 7J^5 + 91J^4 - 205J^3 + 796J^2 - 484J + 528)}{-4K(5J^4 - 14J^3 + 191J^2 - 150J + 616)} \\ + 8K^2(J^2 - J + 14) - K^3$	$4(5J^4 - 62J^3 + 431J^2 - 1206J + 1704) -4L(3J^2 - 15J + 62) + 2L^2$
9	$4(5J^{6} - 45J^{5} + 777J^{4} - 2403J^{3} + 13578J^{2} - 12888J + 26176) -6K(5J^{4} - 18J^{3} + 327J^{2} - 354J + 2128) +2K^{2}(7J^{2} - 9J + 168) - 2K^{3}$	$ \begin{array}{r} 6(5J^6-93J^5+977J^4-5123J^3+16226J^2-25960J+18624)\\ -L(27J^4-270J^3+1961J^2-5230J+8496)\\ +L^2(9J^2-39J+184)-L^3 \end{array} $

where

$$\varepsilon_p = \frac{1}{2} \left[ 1 + (-1)^p \right] = \begin{cases} 1 & p \text{ even} \\ 0 & p \text{ odd} \end{cases},$$
(85)

$$K = 16k(J - k),$$
 (86)

$$L = 16k(J - k - 1), \tag{87}$$

and  $P_{E,N}(J, K)$  and  $P_{O,N}(J, L)$  are polynomials listed in Table III for N = 0-9. A benefit of the results in Table III is that the leading Fourier coefficients in Eqs. (34) and (35) for the Wigner rotation matrices and be readily computed for arbitrarily large integral values of J, M', and M.

These explicit Wigner number formulas give rise to a new class of summation identities as an unexpected benefit. Setting N' = 0 in the sum rule of Eq. (41) gives

$$\sum_{k=0}^{J} W_{2k,J}^{J} W_{2k,J-N}^{J} (2J-2k)! (2k)! = 4^{J} (J!)^{2} \delta_{N,0};$$
(88)

moreover,

$$W_{2k,J}^{J} = (-1)^{k} {J \choose k}$$
 and  $W_{2k+1,J}^{J} = 0$  (89)

are obtained from Eqs. (83) and (84) when N = 0. Combining Eqs. (83), (88), and (89) produces the following summation condition:

$$\sum_{k=0}^{J} {\binom{J}{k}}^2 (2J - 2k)! (2k)! S_N(J,k) = 4^J (J!)^2 \,\delta_{N,0}, \qquad (90)$$

in which

$$S_{N}(J,k) = (J - 2k\varepsilon_{N+1})\frac{(J-1)!}{(J-N)!} - \frac{1}{2}K[(J-2k)\varepsilon_{N+1} + \varepsilon_{N}]P_{\mathrm{E},N}(J,K).$$
(91)

The  $S_N(J, k)$  quantities can be recast in the form

 $S_N(J,k) = \frac{J!}{(J-N)!} + \sum_{p=1}^N b_{N,p} k^p,$ (92)

where the  $b_{N,p}$  coefficients are defined for  $N \ge 1$  as polynomials in *J* that are independent of *k*. The diagonal coefficients are merely  $b_{N,N} = (-1)^N 2^{2N-1}$ , whereas the off-diagonal  $b_{N,p}$  coefficients are tabulated in the supplementary material for  $2 \le N \le 9$  and  $1 \le p \le N - 1$ .

Upon inserting Eq. (92) into Eq. (90) and defining

$$T_{J,p} = \frac{1}{4^{J}(J!)^{2}} \sum_{k=0}^{J} (4k)^{p} {\binom{J}{k}}^{2} (2J - 2k)! (2k)!,$$
(93)

the summation condition becomes

$$\frac{J!}{(J-N)!} T_{J,0} + \sum_{p=1}^{N} 4^{-p} b_{N,p} T_{J,p} = \delta_{N,0}.$$
(94)

for N = 0 and

Equation (94) yields

$$T_{J,N} = 2(-1)^{N+1} \left[ \frac{J!}{(J-N)!} + \sum_{p=1}^{N-1} T_{J,p} 4^{-p} b_{N,p} \right]$$
(96)

for N > 0. Iterative application of Eq. (96) for successively larger values of N produces the set of results reported in Table IV. These remarkable summation identities appear to not have been published heretofore, and their existence would be an enigma without knowledge of the mathematical machinery of Wigner numbers. Analogously, a previous paper<sup>7</sup> on the mathematical form of Wigner rotation matrix elements reported closed forms for a series of simpler summations involving products of two binomial coefficients.

 $T_{I,0} = 1$ 

(95)

TABLE IV. A class of summation identities arising from Wigner number orthonormalization.

$$T_{J,p} = \frac{1}{4^{J}(!!)^{2}} \sum_{k=0}^{J} (4k)^{p} {\binom{J}{k}}^{2} (2J-2k)! (2k)! = \frac{1}{2^{J}} \sum_{k=0}^{J} (4k)^{p} \frac{(2J-2k-1)!!(2k-1)!!}{k!(J-k)!}$$

$$T_{J,0} = 1$$

$$T_{J,1} = 2J$$

$$T_{J,2} = 2J(3J + 1)$$

$$T_{J,3} = 4J^{2}(5J + 3)$$

$$T_{J,4} = 2J(35J^{3} + 30J^{2} + J - 2)$$

$$T_{J,5} = 4J^{2}(63J^{3} + 70J^{2} + 5J - 10)$$

$$T_{J,5} = 4J^{2}(63J^{3} + 70J^{2} + 5J - 10)$$

$$T_{J,6} = 2J(462J^{5} + 630J^{4} + 70J^{3} - 150J^{2} - 4J + 16)$$

$$T_{J,7} = 4J^{2}(858J^{5} + 1386J^{4} + 210J^{3} - 490J^{2} - 28J + 112)$$

$$T_{J,8} = 2J(6435J^{7} + 12012J^{6} + 2310J^{5} - 5880J^{4} - 525J^{3} + 2268J^{2} + 36J - 272)$$

$$T_{J,9} = 4J^{2}(12155J^{7} + 25740J^{6} + 6006J^{5} - 16632J^{4} - 2037J^{3} + 9660J^{2} + 324J - 2448)$$

A final combinatorial formula for the Wigner numbers is motivated by analyses<sup>10,15,24</sup> that have related the coefficients in a Fourier representation of  $d^{I}_{M'M}(\theta)$  to special values of these functions given by Eq. (1) for  $\theta = \frac{1}{2}\pi$ . Comparing our Eqs. (34) and (35) to Eqs. (11)–(13) of Ref. 10 or Eq. (5) of Ref. 24 leads to the following ansatz:

$$W_{m,n}^{J} = \sum_{\sigma=\max[0,m-n]}^{\min[m,2J-n]} (-1)^{m+\sigma} {2J-n \choose \sigma} {n \choose m-\sigma}$$
(97)

or its alternative form

$$W_{m,n}^{J} = (-1)^{m} n! (2J - n)! \sum_{\sigma = \max[0,m-n]}^{\min[m,2J-n]} \times \frac{(-1)^{\sigma}}{\sigma! (2J - n - \sigma)! (m - \sigma)! (n - m + \sigma)!}.$$
 (98)

These formulas can be proved by showing that Eq. (98) obeys the *J*-recursion conditions for Wigner numbers. Outside the summation limits of Eq. (98), at least one factorial argument is a negative integer, giving an unbound quantity in the denominator and hence zero contribution to the sum; therefore, summation limits need not be considered in the proof.

For an incremented J, Eq. (98) provides

$$W_{m,n}^{J+\frac{1}{2}} = (-1)^{m} n! (2J-n)! \sum_{\sigma} \frac{(-1)^{\sigma}}{\sigma! (2J-n-\sigma)! (m-\sigma)! (n-m+\sigma)!} \times \left[ \frac{(2J-n+1)}{(2J-n-\sigma+1)} \right].$$
(99)

The term in brackets in Eq. (99) is equivalent to  $1 + \sigma(2J - n - \sigma + 1)^{-1}$  so that

$$W_{m,n}^{J+\frac{1}{2}} = W_{m,n}^{J} + (-1)^{m} n! (2J-n)! \sum_{\sigma} \times \frac{(-1)^{\sigma}}{(\sigma-1)! (2J-n-\sigma+1)! (m-\sigma)! (n-m+\sigma)!}.$$
 (100)

Setting  $\sigma \rightarrow v + 1$  to reindex the summation yields

$$W_{m,n}^{J+\frac{1}{2}} = W_{m,n}^{J} + (-1)^{m-1} n! (2J-n)! \sum_{\nu} \times \frac{(-1)^{\nu}}{\nu! (2J-n-\nu)! (m-1-\nu)! (n-m+1+\nu)!}.$$
 (101)

Because the summation on the right side of Eq. (101) gives  $W_{m-1,n}^J$ , Eq. (63) is satisfied. Now returning to Eq. (98),

$$W_{m,n+1}^{J+\frac{1}{2}} = (-1)^{m} n! (2J-n)! \sum_{\sigma} \frac{(-1)^{\sigma}}{\sigma! (2J-n-\sigma)! (m-\sigma)! (n-m+\sigma)!} \\ \times \left[\frac{n+1}{(n-m+1+\sigma)}\right].$$
(102)

The term in brackets in Eq. (102) equals  $1 + (m - \sigma)(n - m + 1 + \sigma)^{-1}$ ; thus,

$$W_{m,n+1}^{J+\frac{1}{2}} = W_{m,n}^{J} + (-1)^{m} n! (2J-n)! \sum_{\sigma} \\ \times \frac{(-1)^{\sigma}}{\sigma! (2J-n-\sigma)! (m-\sigma-1)! (n-m+1+\sigma)!}.$$
 (103)

The summation on the right side of Eq. (103) is equal to  $-W_{m-1,n}^{J}$ , showing that Eq. (66) is also satisfied. Finally, Eq. (98) clearly reproduces the root quantity  $W_{0,0}^{0} = 1$ . In summary, all the *J*-recursion conditions are obeyed, proving that Eqs. (97) and (98) are valid combinatorial formulas for Wigner numbers.

#### WIGNER NUMBERS AS N-POLYNOMIALS

A different representation of the Wigner numbers can be derived by repeatedly applying the *M*-recursion formula [Eq. (27)] with successively smaller values of *m*. To demonstrate the approach, we adopt the shorthand notation u = 2N, v = 2J + 2, and  $w_m = W_{m,n}^J$  and write the recursion relation as

ARTICLE

$$mw_m = uw_{m-1} + (m - v)w_{m-2}.$$
 (104)

Using the same equation to replace  $w_{m-1}$  on the right side of Eq. (104) gives

$$m(m-1)w_m = \left[u^2 + (m-1)(m-\nu)\right]w_{m-2} + u(m-1-\nu)w_{m-3}.$$
(105)

Iterating this procedure two more time yields

$$m(m-1)(m-2)w_{m}$$
  
=  $u[u^{2} + (m-1)(m-v) + (m-2)(m-1-v)]w_{m-3}$   
+  $[u^{2} + (m-1)(m-v)](m-2-v)w_{m-4}$  (106)

and then

$$m(m-1)(m-2)(m-3)w_{m} = \begin{bmatrix} u^{4} + u^{2}(m-1)(m-v) + u^{2}(m-2)(m-1-v) \\ + u^{2}(m-3)(m-2-v) + (m-1)(m-3)(m-y)(m-2-v) \end{bmatrix} w_{m-4} \\ + \begin{bmatrix} u^{2} + (m-v)(m-1) + (m-2)(m-1-v) \end{bmatrix} (m-3-v) uw_{m-5}.$$
(107)

After a total of m - 1 iterations in this manner, the Wigner numbers appearing on the right side of the equation for  $w_m$  become  $w_0 = 1$  and  $w_{-1} = 0$ . The coefficient of the former divided by m! thus comprises a formula for  $w_m$  as a polynomial in u. This formula can be written in full notation as

$$W_{m,J-N}^{J} = \frac{1}{m!} \sum_{q=0}^{\left\lfloor \frac{1}{2}m \right\rfloor} G_{m,q}^{J} (2N)^{m-2q},$$
(108)

where the structure of the  $G_{m,q}^{J}$  coefficient, evident after iterating well past Eq. (107), is

$$G_{m,q}^{J} = \sum_{k_1 \ge k_2 \ge \dots \ge k_q \ge 2q}^{m} \prod_{j=1}^{q} (k_j - 2j + 1)(k_j - 2j - 2J).$$
(109)

An alternative approach to finding N-polynomials for Wigner numbers is to represent  $G^{\rm J}_{m,q}$  as

$$G_{m,q}^{J} = \frac{2^{q}m!}{(2q+1)!(m-2q)!}Q_{m,q}^{J}$$
(110)

within Eq. (108); hence,

$$W_{2k,J-N}^{J} = 4^{k} \sum_{q=0}^{k-1} \frac{Q_{2k,q}^{J} N^{2(k-q)}}{2^{q} (2q+1)! (2k-2q)!} + \frac{2^{k} Q_{2k,k}^{J}}{(2k+1)!}$$
(111)

and

$$W_{2k+1,J-N}^{J} = 2^{2k+1} \sum_{q=0}^{k} \frac{Q_{2k+1,q}^{J} N^{2(k-q)+1}}{2^{q} (2q+1)! (2k-2q+1)!}.$$
 (112)

Plugging Eq. (110) into Eq. (108) and inserting the result for  $W_{m,J-N}^{J}$  into Eq. (27) provides

$$\binom{m+1}{\sum_{q=0}^{\lfloor \frac{1}{2}(m+1) \rfloor}} \frac{2^{m+1-q}Q_{m+1,q}^{J}N^{m-2q+1}}{(2q+1)!(m-2q+1)!} - \sum_{q=0}^{\lfloor \frac{1}{2}m \rfloor} \frac{2^{m-q+1}Q_{m,q}^{J}N^{m-2q+1}}{(2q+1)!(m-2q)!} + (2J-m+1) \sum_{q=0}^{\lfloor \frac{1}{2}(m-1) \rfloor} \frac{2^{m-q-1}Q_{m-1,q}^{J}N^{m-2q-1}}{(2q+1)!(m-2q-1)!} = 0.$$
(113)

Two modifications can be made to collect terms in Eq. (113). First, the upper index on the second summation can be changed from  $\lfloor \frac{1}{2}m \rfloor$  to  $\lfloor \frac{1}{2}(m+1) \rfloor$ . If *m* is even, this change is immaterial; if *m* is odd, then we encounter the case  $q = \frac{1}{2}(m+1)$ , which does not appear in Eq. (108) so that we can define  $G_{m,\frac{1}{2}(m+1)}^{J} = 0$  and  $Q_{m,\frac{1}{2}(m+1)}^{J} = 0$  under these circumstances. Second, the lower limit on the third summation can be extended to q = -1, recognizing that  $[(-1)!]^{-1} = 0$ ; then, this summation can be reindexed with the replacement  $q \rightarrow q + 1$ . The net result of these modifications is

$$\begin{bmatrix} \frac{1}{2}(m+1) \\ \sum_{q=0} \end{bmatrix} [(m+1)Q_{m+1,q}^{J} + q(2q+1)(2J-m+1)Q_{m-1,q-1}^{J} \\ -(m-2q+1)Q_{m,q}^{J}] \frac{2^{m-q+1}N^{m-2q+1}}{(2q+1)!(m-2q+1)!} = 0.$$
(114)

The solution for  $Q_{m,q}^{I}$  which satisfies Eq. (114) is clearly

$$(m+1)Q_{m+1,q}^{J} = Q_{m,q}^{J}(m-2q+1) + q(2q+1)(m-2J-1)Q_{m-1,q-1}^{J}$$
(115)

for  $0 \le q \le \lfloor \frac{1}{2}(m+1) \rfloor$  and all  $m \ge -1$ , with the initial conditions  $Q_{m,0}^{J} = 1$  and  $Q_{m,-1}^{J} = 0$ . Table V collects explicit algebraic  $Q_{m,q}^{J}$  formulas for q = 1-5.

J. Chem. Phys. **151**, 244122 (2019); doi: 10.1063/1.5135721 Published under license by AIP Publishing

TABLE V. Leading coefficients for the representation of Wigner numbers as N-polynomials.

$$\begin{split} Q_{m,1}^{J} &= m - 3J - 2 \\ Q_{m,2}^{J} &= 5J(3J - 2m + 5) + \frac{1}{3}(m - 4)(5m - 7) \\ Q_{m,3}^{J} &= 105J^{2}(m - J - 3) - 7J(5m^{2} - 32m + 42) + \frac{1}{9}(m - 6)(35m^{2} - 147m + 124) \\ Q_{m,4}^{J} &= 315J^{3}(3J - 4m + 14) + 63J^{2}(10m^{2} - 74m + 117) - 2J(70m^{3} - 819m^{2} + 2747m - 2565) \\ &+ \frac{1}{15}(m - 8)(175m^{3} - 1470m^{2} + 3509m - 2286) \\ Q_{m,5}^{J} &= 3465J^{4}(5m - 3J - 20) - 1155J^{3}(10m^{2} - 84m + 155) + 55J^{2}(70m^{3} - 924m^{2} + 3587m - 4020) \\ &- \frac{11}{3}J(175m^{4} - 3220m^{3} + 19679m^{2} - 46484m + 34980) + \frac{1}{9}(m - 10)(385m^{4} - 5390m^{3} + 24959m^{2} - 44242m + 24528) \end{split}$$

### ASYMPTOTIC DIFFERENTIAL EQUATION FOR WIGNER NUMBERS

By viewing *m* as a continuous quantity,  $W_{m\pm 1,n}^{J}$  can be expressed as the series expansion

$$W_{m\pm 1,n}^{J} = W_{n}(J;m) \pm W_{n}'(J;m) + \frac{1}{2}W_{n}''(J;m) \\ \pm \frac{1}{6}W_{n}'''(J;m) + \frac{1}{24}W_{n}^{(4)}(J;m) + \cdots.$$
(116)

Using Eq. (116) to eliminate  $W_{m\pm 1,n}^{J}$  from Eq. (27) provides the following differential equation:

$$(n+1)W_n(J;m) + (m-J)W'_n(J;m) + \frac{1}{2}(J+1)W''_n(J;m) = -\frac{1}{6}(m-J)W''_n(J;m) - \frac{1}{24}(J+1)W_n^{(4)}(J;m).$$
(117)

In terms of the variable,

$$z = \frac{\sqrt{2}(m-J)}{\sqrt{(J+1)}} = \frac{-\sqrt{2}M}{\sqrt{(J+1)}},$$
(118)

Eq. (117) takes the form

$$(n+1)W_n(J;z) + zW'_n(J;z) + W''_n(J;z) = -\frac{1}{6(J+1)} \Big[ 2zW''_n(J;z) + W^{(4)}_n(J;z) + \cdots \Big].$$
(119)

In the limit of large *J*, the right side of Eq. (119) goes to 0. Thus, a Wigner-number approximation  $\tilde{W}_n(J;z)$  is obtained as a solution to a second-order ordinary differential equation,

$$(n+1)\tilde{W}_n(J;z) + z\tilde{W}'_n(J;z) + \tilde{W}''_n(J;z) = 0.$$
(120)

Into this asymptotic equation, we can substitute

$$\tilde{W}_n(J;z) = \omega_{n,0} U_n(z) e^{-\frac{1}{2}z^2},$$
(121)

in which  $\omega_{n,0}$  is a constant, and thus arrive at a differential equation for  $U_n(z)$ ,

$$U_n''(z) - zU_n'(z) + nU_n(z) = 0.$$
(122)

J. Chem. Phys. **151**, 244122 (2019); doi: 10.1063/1.5135721 Published under license by AIP Publishing The series solution

$$U_n(z) = \sum_{p=0}^{\infty} a_{n,p} z^p \tag{123}$$

yields the function  $U_n(z)$  that modulates the Gaussian factor in the asymptotic form of the Wigner numbers. Employing Eq. (123) within Eq. (122) produces the condition

$$\sum_{p=0}^{\infty} \left[ a_{n,p} (n-p) + a_{n,p+2} (p+1) (p+2) \right] z^p = 0, \qquad (124)$$

which requires the coefficients to obey the recursion relation

$$a_{n,p+2} = \frac{a_{n,p}(p-n)}{(p+1)(p+2)}.$$
(125)

Equation (125) shows that the power series for  $U_n(z)$  truncates when p = n; moreover, each coefficient is independent of its immediate predecessor. Accordingly,  $U_n(z)$  must be either an even or odd polynomial of order *n*. If the coefficient of the highest power of *z* in  $U_n(z)$  is chosen to be 1, then Eq. (125) yields the following explicit formulas for  $0 \le k \le \lfloor \frac{1}{2}n \rfloor$ :

$$a_{n,2k} = \frac{(-1)^{n/2-k} n!}{(2k)!(n-2k)!!} \quad (n \text{ even})$$
(126)

and

$$a_{n,2k+1} = \frac{(-1)^{(n-1)/2-k} n!}{(2k+1)!(n-2k-1)!!} \quad (n \text{ odd}).$$
(127)

Table VI presents the  $U_n(z)$  polynomials for n = 0-9. It is evident from Table VI and straightforward to prove from Eqs. (126) and (127) that the  $U_n(z)$  polynomials obey the simple recursion relation

$$U_{n+1}(z) = zU_n(z) - nU_{n-1}(z).$$
(128)

The class of functions  $\tilde{W}_n(J; z)$  becomes completely specified if each  $\omega_{n,0}$  constant in Eq. (121) is chosen to reproduce the nonzero  $W_{m,n}^J$  integer whose corresponding *m* value is closest to or coincides with the central point at z = 0. Even at low *J*, these  $\tilde{W}_n(J; z)$  functions provide a valuable means of interpreting the  $W_{m,n}^J$  quantities and suggest a continuous representation  $W_n(J;z)$  of the Wigner numbers that is exact and physically sound. In particular, the asymptotic form of  $\tilde{W}_n(J;z)$  motivates the basis set expansion

**TABLE VI**. Polynomials modulating the Gaussian factor in the asymptotic form of the Wigner numbers.

п	$U_n(z)$
0	1
1	Z
2	$z^2-1$
3	$z^{3} - 3z$
4	$z^4 - 6z^2 + 3$
5	$z^5 - 10z^3 + 15z$
6	$z^6 - 15z^4 + 45z^2 - 15$
7	$z^7 - 21z^5 + 105z^3 - 105z$
8	$z^8 - 28z^6 + 210z^4 - 420z^2 + 105$
9	$z^9 - 36z^7 + 378z^5 - 1260z^3 + 945z$

$$W_n(J;z) = \sum_{p=0}^{2(J+\mu)} c_{pn} z^p e^{-\frac{1}{2}z^2},$$
 (129)

where the coefficients  $c_{pn}$  are chosen to reproduce the  $W_{m,n}^J$  integers at the points  $z_m$ . This approach proves superior to spline interpolation by producing results that are devoid of superfluous undulations and more faithful to the asymptotic profiles.

The expansion coefficients are solutions to a linear system of equations,

$$W_{m,n}^{J} = \sum_{p=0}^{2(J+\mu)} F_{mp} c_{pn},$$
(130)

involving a matrix F whose elements are

$$F_{mp} = z_m^p e^{-\frac{1}{2}z_m^2}.$$
 (131)

The fitting points comprise the genuine Wigner integers for  $0 \le m \le 2J$  augmented by damping values  $W_{m,n}^J = 0$  for  $-\mu \le m \le -1$ and  $2J + 1 \le m \le 2J + \mu$ . The  $2\mu$  additional points prevent any artificial, unphysically large oscillations in  $W_n(J; z)$  in the large |z| regions where the distribution should decay to zero. Our investigations found that  $\mu = \lfloor \frac{1}{2}J \rfloor$  is an efficacious choice for this purpose. In summary, the expansion coefficients in Eq. (129) are determined as

$$c_{pn} = \sum_{m=-\mu}^{2J+\mu} (F^{-1})_{pm} W^{J}_{m,n}.$$
 (132)

Extra high numerical precision is usually required to invert **F** accurately. Because the  $W_n(J; z)$  functions have symmetry, Eq. (129) will include only even or odd powers of *z*. Therefore, the set of fitting points can be restricted to  $m \ge J$  with concomitant reduction in the dimension of the **F** matrix by a factor of 2.

Illustrative  $W_n(J; z)$  functions that define the Wigner numbers for continuous values of m are plotted in Fig. 1 for  $J = \frac{21}{2}$  and n = 0, 1, 2, and 3; scaling by the corresponding  $W_{m,n}^J$  point of largest magnitude is performed therein to allow all the curves to appear on the same figure. The  $W_n(J; z)$  functions display a striking resemblance to quantum-mechanical stationary-state wave functions that would appear in a symmetric binding potential. In particular, as n increases, the  $W_n(J; z)$  curves incrementally develop more nodes and alternate between even and odd functions; in this respect, n acts like a vibrational quantum number. The  $W_n(J; z)$  functions resemble but are not equivalent to harmonic-oscillator wave functions; although both contain overarching Gaussian factors, the modulating polynomials are different. In proceeding from the central value of m, the  $U_n(z)$  polynomials yield local extrema that successively diminish in magnitude, quite unlike the profiles given by the Hermite polynomials.

Representative plots are presented in Fig. 2 that compare the exact  $W_n(J; z)$  and asymptotic  $\tilde{W}_n(J; z)$  functions for J = 50 and n = 0, 5, and 10. Even for such a modest J, the near coincidence of  $W_n(J; z)$  and  $\tilde{W}_n(J; z)$  across the range of m is remarkable for the lowest n values. As n increases, the accord between the two functions starts to diminish. For n values significantly larger than shown in Fig. 2, the exact and asymptotic curves maintain very similar shapes but begin to exhibit substantial phase shifts.

**FIG. 1.** For the  $J = \frac{21}{2}$  case, scaled Wigner numbers are plotted as continuous functions of *m* for the lowest four values of the quantum number *n*; corresponding scaled  $W_{m,n}^J$  integers are shown as discrete points.



J. Chem. Phys. **151**, 244122 (2019); doi: 10.1063/1.5135721 Published under license by AIP Publishing



**FIG. 2.** For the J = 50 case, the exact  $W_n(J;z)$  (colored traces) and asymptotic  $\tilde{W}_n(J;z)$  (black curves) functions are compared for the n = 0, 5, and 10 quantum numbers.

#### SUMMARY

The key findings in this paper have been obtained by rigorous mathematical proofs and analyses. For all allowed values of the angular momentum quantum number J, a sequence of integers  $W'_{m,n}$  for  $(m, n) \in [0, 2J]$  can be generated via *M*-recursion [Eq. (27)] by starting with the boundary conditions  $W_{0,n}^J = 1$  and  $W_{-1,n}^J = 0$ . These solutions constitute an alluring class of quantities designated here as Wigner numbers. The normalized eigenvectors of the  $\hat{J}_X$  and  $\hat{J}_Y$  operators represented in the basis of  $\hat{J}_Z$  eigenstates are given exactly in terms of Wigner numbers by Eqs. (30) and (22), respectively, which are efficacious for arbitrarily large J. Thus, Wigner numbers obviate the need to ever diagonalize  $\hat{J}_X$  and  $\hat{J}_Y$  matrices by numerical methods. Moreover, by means of Eqs. (32)–(35), the  $W_{m,n}^{I}$  integers allow any reduced Wigner rotation matrix element  $d^{J}_{M'M}(\theta)$  to be obtained as a linear combination of either  $\cos(N\theta)$  or  $\sin(N\theta)$  terms as N runs in unit steps from either 0 or  $\frac{1}{2}$  to J. These exact, infiniteprecision Fourier representations that involve only |J + 1| terms fully eliminate longstanding problems in computing  $d'_{M'M}(\theta)$  elements for large *J* values. For J > 0, the Fourier coefficients threat ments for large *J* values. For J > 0, the Fourier coefficients threat appear to always take the simple form  $(\rho_0^{J,M'M})^{1/2} \rho_{N,1}^{J,M'M} (\rho_{N,2}^{J,M'M})^{-1}$ , where  $\rho_0^{J,M'M}$  is an integer whose prime factorization contains 2 no more than once,  $\rho_{N,1}^{J,M'M}$  is an odd integer, and  $\rho_{N,2}^{J,M'M}$  equals 2 raised to a non-negative integral power less than 2J.

The Wigner numbers exhibit a wide variety of notable mathematical properties. The  $W_{m,n}^{J}$  integers obey the *m*- and *n*-sum rules specified by Eqs. (41) and (42). Remarkably, the defining  $W_{m,n}^{J}$  recursion equation can be solved to obtain various explicit combinatorial formulas, Eqs. (78), (79), (81), and (82); the first two solutions entail double summations over a product of three binomial coefficients, while the last two equations require single summations over a binomial coefficient multiplied by a Jacobi polynomial of the form  $P_{N-1}^{(1,q)}(x)$  evaluated at x = 3. Equations (81) and (82) can be profitably reduced for cases in which *N* is a small integer to provide Eqs. (83)–(87) and Table III that compactly formulate the Wigner numbers  $W_{m,J-N}^J$  for the cases N = 0-9. An unexpected consequence of Eqs. (41), (83), and (84) is the class of novel summation identities shown in Table IV. The most efficient combinatorial formula is Eq. (97), although none of the explicit  $W_{m,n}^J$  equations can match the speed of the *M*-recursion approach. Finally, the Wigner numbers can be written explicitly as *N*-polynomials as in Eq. (108) or Eqs. (111) and (112); the polynomial coefficients can be obtained by either Eq. (109) or Eq. (115) and Table V.

The Wigner numbers not only obey the defining *M*-sequence [Eq. (27)] but also follow the *N*-recursion formula of Eq. (55). Moreover, the *J*-recursion relations of Eqs. (63) and (66) allow all Wigner numbers to be constructed by successive additions from the root quantity  $W_{0,0}^0 = 1$ , proving that all  $W_{m,n}^J$  values are integers, a property not at all evident from Eq. (27) or the first three combinatorial formulas. The Wigner numbers can be generalized into continuous functions  $W_n(J; z)$  that depend on the variable  $z = 2^{1/2}(J + 1)^{-1/2}(m - J)$  and resemble quantum-mechanical stationary-state wave functions bound by a symmetric potential. In the limit of large *J*, these functions satisfy a second-order differential equation and become products of a polynomial  $U_n(z)$  (Table VI) and a Gaussian factor  $e^{-\frac{1}{2}z^2}$ .

In order to confirm the extensive derivations presented in this paper, exhaustive testing of the new formulas has been carried out over all possible (m, n) combinations for all integral and half-integral J values up to 50. The *Mathematica* program WignerNumbers written for this purpose is provided in the supplementary material to facilitate independent validation of our final equations. The program can generate complete  $W_{m,n}^J$  matrices for J as large as 1200 in less than one CPU minute on a 2.8 GHz processor. Nevertheless, the most efficient means of deriving a specific  $d_{M'M}^J(\theta)$  element is to only compute the J-M' and J-M rows of the  $W_{m,n}^J$  matrix using N-recursion based on Eqs. (55) and (56). Even for J = 25000, the program yields in this manner the Fourier coefficient integers of Eq. (36) for any particular  $d_{M'M}^J(\theta)$  element in less than 8 CPU minutes. In practical applications that require  $d_{M'M}^J(\theta)$  elements to be calculated for *J* values less than a few hundred, all the Fourier coefficients in Eq. (36) can be easily precomputed by means of Wigner numbers and stored on disk or in memory as floating-point numbers. In the *J* = 200 example, the double-precision Fourier coefficients of all unique  $d_{M'M}^J(\theta)$  elements require as little as 32 MB of storage. Hence, for any argument  $\theta$ , each  $d_{M'M}^J(\theta)$  quantity is given as a simple dot product of a precomputed Fourier coefficient vector and a vector of trigonometric values  $\cos(N\theta)$  or  $\sin(N\theta)$  for  $N = N_0$  to *J*. From the perspective of accuracy and efficiency, the mathematics presented in this paper represents a full solution to the problem of computing the Wigner rotation matrices originally introduced 92 years ago.

### SUPPLEMENTARY MATERIAL

See the supplementary material for representative Fourier contributions to selected reduced Wigner matrix elements (Table S1), coefficients determining the summation identities  $T_{J,N}$  (Table S2), proof of Eq. (80), an alternative recursion formula for reduced Wigner matrices, tabulations of Wigner numbers, and the Wigner-Numbers computer program.

### REFERENCES

<sup>1</sup>M. E. Rose, *Elementary Theory of Angular Momentum* (Wiley, New York, 1957).
 <sup>2</sup>A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, NJ, 1960).

<sup>3</sup> R. N. Zare, Angular Momentum: Understanding Spatial Aspects in Chemistry and Physics (Wiley, New York, 1988).

<sup>4</sup>M. A. Morrison and G. A. Parker, "A guide to rotations in quantum mechanics," Aust. J. Phys. **40**, 465–497 (1987).

<sup>5</sup>K.-M. Chen and C.-C. Pei, "An addition theorem of Wigner rotation matrices and its application in photodissociation processes," Chem. Phys. Lett. **124**, 365– 369 (1986).

<sup>6</sup>A. Ernesti and H. J. Korsch, "Factorization relations and Wigner's rotation matrices," J. Phys. A: Math. Gen. 25, L817–L821 (1992).

<sup>7</sup>S.-T. Lai, P. Palting, and Y.-N. Chiu, "On the closed form of Wigner rotation matrix elements," J. Math. Chem. **19**, 131–145 (1996).

<sup>8</sup>H. Dachsel, "Fast and accurate determination of the Wigner rotation matrices in the fast multipole method," J. Chem. Phys. **124**, 144115 (2006).

<sup>9</sup>G. Aubert, "An alternative to Wigner d-matrices for rotating real spherical harmonics," AIP Adv. **3**, 062121 (2013).

 $^{10}$  N. Tajima, "Analytical formula for numerical evaluations of the Wigner rotation matrices at high spins," Phys. Rev. C **91**, 014320 (2015).

<sup>11</sup>X. M. Feng, P. Wang, W. Yang, and G. R. Jin, "High-precision evaluation of Wigner's *d* matrix by exact diagonalization," Phys. Rev. E **92**, 043307 (2015).

<sup>12</sup>P. P. Man, "Wigner active and passive rotation matrices applied to NMR tensor," Concepts Magn. Reson., Part A **45A**, e21385-1–e21385-64 (2017).

<sup>13</sup>S. E. Hoffmann, "Uniform analytic approximations of Wigner rotation matrices," J. Math. Phys. 59, 022102 (2018).

<sup>14</sup>C. A. White and M. Head-Gordon, "Rotating around the quartic angular momentum barrier in fast multipole method calculations," J. Chem. Phys. 105, 5061–5067 (1996).

<sup>15</sup>T. Risbo, "Fourier transform summation of Legendre series and D-functions," J. Geodesy **70**, 383–396 (1996).

<sup>16</sup>M. A. Blanco, M. Flórez, and M. Bermejo, "Evaluation of the rotation matrices in the basis of real spherical harmonics," J. Mol. Struct.: THEOCHEM **419**, 19–27 (1997).

<sup>17</sup>J. Ivanic and K. Ruedenberg, "Rotation matrices for real spherical harmonics. Direct determination by recursion," J. Phys. Chem. **100**, 6342–6347 (1996);
 J. Phys. Chem. A **102**, 9099–9100 (1998).

<sup>18</sup>C. H. Choi, J. Ivanic, M. S. Gordon, and K. Ruedenberg, "Rapid and stable determination of rotation matrices between spherical harmonics by direct recursion," J. Chem. Phys. **111**, 8825 (1999).

<sup>19</sup>G. Prézeau and M. Reinecke, "Algorithm for the evaluation of reduced Wigner matrices," Astrophys. J., Suppl. Ser. **190**, 267–274 (2010). <sup>20</sup>The  $d_{M'M}^{j}(\theta)$  phase conventions in Refs. 1, 3, and 4 are the same as those

<sup>20</sup> The  $d'_{M'M}(\theta)$  phase conventions in Refs. 1, 3, and 4 are the same as those adopted here, except that the signs of  $d^2_{1,0}$ ,  $d^2_{-1,0}$ ,  $d^2_{0,1}$ , and  $d^2_{0,-1}$  are different in Ref. 3. The  $d'_{M'M}(\theta)$  matrices of Ref. 2 and *Mathematica* are transposed relative to ours, and thus corresponding elements differ by a factor of  $(-1)^{M-M'}$ .

<sup>21</sup>The customary Wigner formula<sup>3,10,11</sup> is recovered by the following steps: (1) make the replacements  $M \to -M$  and  $\theta \to \theta - \pi$  in Eq. (1); (2) utilize the trigonometric identities  $\cos\left[\frac{1}{2}(\theta - \pi)\right] = \sin\left(\frac{1}{2}\theta\right)$  and  $\sin\left[\frac{1}{2}(\theta - \pi)\right] = -\cos\left(\frac{1}{2}\theta\right)$ ; (3) employ  $d_{M',-M}^{I}(\theta - \pi) = (-1)^{J-M} d_{M'M}^{J}(\theta)$ ; and (4) simplify the phase factors.

<sup>22</sup>The reduced Wigner matrices provided in version 10.3.0.0 of *Mathematica* seem to be derived from Eq. (1) by the following steps: factor out the product  $\cos^p(\frac{1}{2}\theta)\sin^q(\frac{1}{2}\theta)$  with the largest possible integral values of p and q from the summation; and then inside the remaining terms in the summation make the sequential substitutions  $\cos^2(\frac{1}{2}\theta) \rightarrow 1 - \sin^2(\frac{1}{2}\theta)$  and  $\sin^2(\frac{1}{2}\theta) \rightarrow \frac{1}{2}(1 - \cos\theta)$ . <sup>23</sup>*Handbook of Mathematical Functions*, 10th printing, edited by M. Abramowitz and I. A. Stegun (U.S. National Bureau of Standards, 1972); Equation 3.1.6, p. 10. <sup>24</sup>K. M. Huffenberger and B. D. Wandelt, "Fast and exact spin-s spherical harmonic transforms," Astrophys. J., Suppl. Ser. **189**, 255–260 (2010).