Wigner representation of the rotational dynamics of rigid tops

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We propose a methodology to design Wigner representations in phase spaces with nontrivial topology having evolution equations with desired mathematical properties. As an illustration, two representations of molecular rotations are developed to facilitate the analysis of molecular alignment in moderately intense laser fields, reaction dynamics, scattering phenomena, and dissipative processes.

I. INTRODUCTION

The dynamics of complex quantum systems on the border between classical and quantum mechanics is relevant to a variety of fields, including quantum optics and information, structural analysis, studies of matter waves, and mechanisms of chemical reactions (see, e.g., [1–5]). The details of these dynamics can be experimentally traced with up to attosecond resolution, owing to the advances in quantum state preparation and transient probing [6–9]. However, specialized models are needed to numerically access this regime. Attractive approaches are based on semiclassical propagation of the Wigner function [10–13], including phase integral methods [14] and a large family of initial value representations and their related techniques and extensions [5,15,16].

The idea underlying all these approaches is to find a computationally efficient way to expand any given exact generator of quantum motion in a rapidly converging series [17]. However, the mathematical form of the exact generator of motion can be substantially altered by changing the topology of the underlying configuration space [4,15,18]. Such structural flexibility potentially embodies wide opportunities to equip the representation with the desired properties and behavior [19–21]. The analysis of this resource with the specific application to rotational motion of extended bodies, a fascinating problem with many applications [22–26]), constitutes the subject of the present paper.

A variety of ways to extend the original Wigner quantization ansatz to the case of rotational dynamics were suggested and analyzed [27–39], but only a few of them are applicable to unrestricted rotations of three-dimensional bodies. The early solutions of Refs. [35,36] reduce the problem to the canonical case at the cost of extending the phase space by two artificial dimensions. A variant suggested in Ref. [37] allows one to directly extract the most useful partial distributions but involves rather complicated quantum Liouville equations. Conversely, in the Nasyrov proposal [38], the equations for free symmetric and linear tops coincide with the classical ones at the expense of a complicated integro-differential form for the dynamical equations and common observables in the general case. Similar drawbacks also limit the utility of schemes [39] based on direct extension of the Stratonovich-Weyl correspondence for spin [40].

In this paper we suggest that the roots of many of the dynamical drawbacks are hidden in the employed phase-space quantization procedure. The latter usually follows closely the original Wigner reasoning [10,41], grounded on axiomatizing certain static properties of desired quasiprobability distribution [10,41] (a notable exception is the Nasyrov scheme [38]). This approach, however, lacks the tools to explicitly control the mathematical structure and complexity of the resulting dynamical equations.

Here we show that this issue can be resolved by replacing certain traditional axioms of phase-space quantization with the postulates imposed on the properties of evolution equations for the Wigner function. The basics of the resulting hybrid static-dynamical phase-space quantization scheme are detailed in Sec. II. In Secs. III and IV we apply this scheme to address the problem of developing a numerically efficient phase-space quantization of rotation motions. We explore two design routes by departing from the classical Euler equations and from the Liouville equations written in terms of the components of angular momenta and quaternion parameters. Correspondingly, we arrive at two alternative representations. In both cases we resolve many of the mentioned drawbacks of the existing phase-space quantizations but also gain a better understanding of quantum rotations. For example, the second representation uncovers the deep physical relation between quaternions and the raising and lowering operators of the Schwinger oscillator model and also complements the dynamical picture in the Nasyrov quantization approach [38]. These findings clarify the origins of the remarkable possibility to exactly reduce the quantum Liouville equation of the free symmetric top to the classical form. We encourage readers to check Sec. V for a brief summary of the key features and the expected advantages of the these representations in numerical simulations. We defer to five Appendixes mathematical details that we expect to interest the reader but are not necessary for conveying our message.

II. FUNDAMENTALS OF GENERALIZED WIGNER REPRESENTATIONS

Despite being essentially different, the quantum and classical statistical mechanics operate with the same set of objects: the set of all elementary physical events (the probability space) Σ, the algebra $\mathscr{B}$ of these events, and the probability measure $\mathscr{P}$ for any measurable subset in $\mathscr{B}$ [42]. Fortunately, the Hilbert space framework is fully compatible with both classical and quantum-mechanical objects [43,44]. The Wigner

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representation exploits this fundamental fact. It is constructed by equipping the classical phase space Σ with such an additional scalar product $(\ ,\ )_W$ in which the resulted Hilbert space Σ_W can simultaneously host both classical and quantum algebras. This change formally converts both classical and quantum quantities into operators acting in Σ_W. To distinguish between them we will denote the latter by a curved right overarrow (e.g. $\overset{\frown}{F}$), preserving the caret notation for operators in the ordinary configuration Hilbert space.

Compatibility with classical mechanics requires consistency of the definitions of $(\ ,\ )_W$ and (scalar-valued) classical averaging of any physical quantity F over classical canonical coordinates and momenta q_i and p_i ($i = 1, \ldots, N$):

$$\langle F \rangle = (F, \overset{\frown}{\rho})_W = \int \cdots \int_{\Sigma_W} F \overset{\frown}{\rho}\, d\Omega, \tag{1}$$

where $d\Omega = dp_1 \cdots dp_N dq_1 \cdots dq_N$ and $\overset{\frown}{\rho}$ denotes the generalized probability distribution in phase space called the Wigner function (or the Weyl symbol of the density matrix). This relation should be viewed as a classical analog of the quantum equality $\langle \hat{F} \rangle = \mathrm{Tr}[\hat{F}\hat{\rho}]$ if the classical quantity F is substituted by its quantum counterpart $\overset{\frown}{F}$,

$$\langle \hat{F} \rangle = (\overset{\frown}{F}, \overset{\frown}{\rho})_W. \tag{2}$$

Since in quantum mechanics the observables and states are treated on the same footing, it is worth requiring the following traciality relation for any two states $\hat{\rho}_1$ and $\hat{\rho}_2$:

$$(\overset{\frown}{\rho}_1, \overset{\frown}{\rho}_2)_W = C\, \mathrm{Tr}[\hat{\rho}_1 \hat{\rho}_2], \quad C = \mathrm{const}. \tag{3}$$

It is also natural to impose the constraint that the images $\overset{\frown}{F}$ of quantum observables $\hat{F}$ remain Hermitian in Σ_W:

$$\overset{\frown}{F} = \overset{\frown}{F}^\dagger. \tag{4}$$

Equations (3) and (4) imply that

$$\overset{\frown}{\rho} = \overset{\frown}{\rho}^\dagger = \overset{\frown}{\rho}^* \tag{5}$$

is a real-valued symmetric function of phase variables.

With this, the explicit form of images $\widehat{x}_i$ and $\widehat{p}_i$ of the quantum coordinate and momentum operators (termed Bopp operators [11,45]) is uniquely defined by (i) the fundamental property of Galilean invariance of nonrelativistic phase space Σ_W (which requires $\widehat{x}_i$ and $\widehat{p}_i$ to be linear in both p_i, q_i and $\frac{\partial}{\partial p_i}, \frac{\partial}{\partial q_i}$), (ii) the canonical commutation relation $[\widehat{x}_i, \widehat{p}_i] = i\hbar$, and (iii) the requirement of proper classical limit $\widehat{p}_i|_{\hbar \to 0} = p_i$ and $\widehat{q}_i|_{\hbar \to 0} = q_i$:

$$\widehat{x}_i = x_i + \frac{i\hbar}{2}\frac{\partial}{\partial p_i}, \quad \widehat{p}_i = p_i - \frac{i\hbar}{2}\frac{\partial}{\partial x_i}. \tag{6}$$

Note that these operators when applied to $\overset{\frown}{\rho}$ produce images of the left multiplications $\hat{p}_i \hat{\rho}$ and $\hat{q}_i \hat{\rho}$. It is convenient to introduce the operators $\overset{\frown}{p}_i$ and $\overset{\frown}{q}_i$ whose effect on $\overset{\frown}{\rho}$ is associated with the right multiplications. The associativity relations of form $\forall \hat{\rho} : \hat{q}_i(\hat{\rho}\hat{q}_j) = (\hat{q}_i \hat{\rho})\hat{q}_j$ and the equality $[\hat{q}_i, \hat{p}_i]\hat{\rho} = -(\hat{\rho}^\dagger[\hat{q}_i, \hat{p}_i])^\dagger$ imply that the right operators should

satisfy the commutation relations

$$[\overset{\frown}{p}_i, \overset{\frown}{p}_j] = [\overset{\frown}{p}_i, \overset{\frown}{q}_j] = [\overset{\frown}{q}_i, \overset{\frown}{p}_j] = [\overset{\frown}{q}_i, \overset{\frown}{q}_j] = 0,$$
$$[\overset{\frown}{q}_i, \overset{\frown}{p}_j] = -i\delta_{i,j}\hbar. \tag{7}$$

Combining Eq. (7) with the requirements of Galilean invariance and proper classical limit, one can conclude that

$$\overset{\frown}{p}_i = x_i - \frac{i\hbar}{2}\frac{\partial}{\partial p_i} = \widehat{p}_i^{\,*}, \quad \overset{\frown}{q}_i = p_i + \frac{i\hbar}{2}\frac{\partial}{\partial x_i} = \widehat{q}_i^{\,*}. \tag{8}$$

The equality $\mathrm{Tr}[\hat{q}_i^n \hat{\rho}] = \frac{1}{2^n}\sum_{m=0}^n C_n^m \mathrm{Tr}[\hat{q}_i^m \hat{\rho}\hat{q}_i^{n-m}]$, where C_n^m are binomial coefficients, and the similar expression for $\hat{p}_i$ lead to the conclusion that

$$(\widehat{q}_i^{\,n}, \overset{\frown}{\rho})_W = \frac{1}{2^n}((\widehat{q}_i + \overset{\frown}{q}_i)^n, \overset{\frown}{\rho})_W = (q_i^n, \overset{\frown}{\rho})_W,$$
$$(\widehat{p}_i^{\,n}, \overset{\frown}{\rho})_W = (p_i^n, \overset{\frown}{\rho})_W. \tag{9}$$

In particular, Eqs. (9) mean that the partial integration on the right-hand side of (1) over coordinates (momenta) with $F = 1$ returning the correct marginal probability distributions for values of momenta (coordinates).

Equations (6) and (8) completely specify the quantum algebra. Indeed, they establish a one-to-one correspondence between an arbitrary quantum operator $\hat{F} = F(\hat{p}, \hat{q})$, its Wigner image $\overset{\frown}{F} = F(\overset{\frown}{p}, \overset{\frown}{q})$, and the Weyl symbol $F_W(p, q) = F(\overset{\frown}{p}, \overset{\frown}{q})1$ (see [11,12]) for details), and define the image of the master equation $\frac{\partial}{\partial t}\hat{\rho} = \frac{-i}{\hbar}[\hat{H}, \hat{\rho}]$ with the Hamiltonian $\hat{H} = H(\hat{p}, \hat{q})$:

$$\frac{\partial}{\partial t}\overset{\frown}{\rho} = \overset{\frown}{\mathscr{L}}\overset{\frown}{\rho}, \tag{10}$$

where the quantum Liouville operator $\overset{\frown}{\mathscr{L}}$ is given by the real operator

$$\overset{\frown}{\mathscr{L}} = \frac{-i}{\hbar}[H(\overset{\frown}{p}, \overset{\frown}{q}) - H(\widehat{p}, \widehat{q})]. \tag{11}$$

However, in the general case of noncanonical phase spaces, Eqs. (2)–(10) are not self-consistent and hence some of them must be relaxed. For example:

(I) one can impose the desired static properties of the quasiprobablilty distribution like (2), (3), (5), and (9) and then deduce from them the expressions for Weyl symbols, Moyal products, and evolution equations or, alternatively,

(II) one can depart from the desired algebraic and dynamic properties of images of quantum operators and/or generators of motion [e.g., Eqs. (4), (6), and (7)].

Algorithm I is rigorously axiomatized [41]; its abstracted generalization in group-theoretic terms (termed Stratonovich-Weyl correspondence [40]) can be applied to arbitrary phase spaces with complex symmetries (see, e.g., [46,47]). One practical and formally justified [48,49] axiomatic basis for algorithm II postulates the equations of motion for averages of certain physical quantities [38,50]. Another possible starting point is Feynman's path-integral representations of the time evolution [12]. One of the fundamental origins of this diversity of possible definitions is the wide freedom in choosing either the Weyl symbols of density matrices $\overset{\frown}{\rho}$ or quantum observables $\overset{\frown}{F}$ to be a main carriers of nonclassicality (see [4,51] for details).

The quantization method presented in this paper uses this diversity to construct Wigner quantizers tailored to specific dynamical problems. First, we identify the desired dynamical characteristics of the quantizer that would enhance its applied value. In our examples we consider goals such as computational simplicity of the quantum equations of motion and preferable forms of certain Bopp operators. At the next step we introduce these preferences into the standard set of postulates of Wigner quantization. In doing this, we have to relax some of these canonical postulates (considered as the least important ones in the context of the anticipated applications) in order to obtain a consistent axiomatic basis. The modified postulates no longer uniquely specify the representation but are accompanied by additional loose dynamical criteria (such as simplicity of certain operators) defined in physical rather than mathematical terms. This makes the construction algorithm II more suitable: One starts with deducing the forms of dynamical Bopp operators that best account for these additional criteria and then completes the definition of the Wigner function and its static properties accordingly. In the following sections we demonstrate the efficacy of this general scheme by obtaining different computationally efficient phase-space representations of the rotational dynamics of rigid tops.

III. QUANTIZATION OF THE EULER EQUATIONS

Euler's celebrated equations

$$\frac{d}{dt}L_i = \sum_{j,k=1}^{3} \epsilon_{i,j,k}\left(\frac{1}{I_k} - \frac{1}{I_j}\right)\frac{L_j L_k}{2} \quad (i=1,2,3), \quad (12)$$

where $\epsilon_{i,j,k}$ is the Levi-Città symbol, I_k are moments of inertia about the principal axes $\vec{e}_k$ of the rigid body, and L_k are the projections of the angular momentum on $\vec{e}_k$, describe the free dynamics of rigid bodies in the moving frame S in terms of the phase space $\Sigma_W^L = \{L_1, L_2, L_3\}$. We will require the quantum generalization of Eq. (12) to obey:

(E:1) the condition (5) of reality of the quasiprobability distributions $\widehat{\rho} = \widehat{\rho}(L_1, L_2, L_3)$, i.e., any real-valued Weyl symbol $\widehat{\rho}'$ should correspond to a unique Hermitian (but not necessarily positive) matrix $\hat{\rho}'$;

(E:2) the traciality relation (3) in Σ_W^L where $(\widehat{\odot}_1, \widehat{\odot}_2)_W \overset{\text{def}}{=} \int_{\Sigma_W^L} \widehat{\odot}_1^* \widehat{\odot}_2 \, dL_1 dL_2 dL_3$ and $C=1$;

(E:3) the consistency of the classical limit for $\widehat{L}_i$ and the evolution equation (10) with the Euler equations (12)

$$\widehat{L}_i|_{\hbar \to 0} = L_i, \quad \widehat{\mathscr{L}}|_{\hbar \to 0} = \mathscr{L} = \sum_{i,j,k=1}^{3} \epsilon_{i,j,k}\left(\frac{L_j L_k}{I_k}\right)\frac{\partial}{\partial L_i}. \tag{13}$$

Postulates (E:1) and (E:2) match the conventional Wigner reasoning [41]. They also guarantee that the Hermiticity condition (4) holds (i.e., $\widehat{F}^*|_{\partial/\partial L_j \to -\partial/\partial L_j} = \widehat{F}$ for the image $\widehat{F}$ of any observable). Hence, these postulates preserve the essential subset of the properties of the conventional Wigner function. However, in defining (E:3) we apply algorithm II and

substitute the static condition (9) on the marginal distributions with a dynamical restriction. The postulates (E:1)–(E:3) do not uniquely determine the Wigner representation. This allows us to introduce the condition of mathematical simplicity of the quantum Liouville operator $\widehat{\mathscr{L}}$ as the additional loose dynamical figure of merit.

It is worth stressing that the postulates (E:1)–(E:3) do not allow us to immediately identify (i.e., in a way similar to the standard quantization scheme [41]) the explicit form of the isomorphism between the density-matrix formulation and the Wigner function formulation. Instead, we have to start by identifying the structure of the key dynamical Bopp operators $\widehat{L}_k$ and $\widehat{\mathscr{L}}$ consistent with the given postulates and then go back and complete the identification of the function $\widehat{\rho}$ that enters Eq. (10).

The images of the associativity and commutation relations $\forall \rho : \hat{L}_k(\rho \hat{L}_l) = (\hat{L}_k \rho)\hat{L}_l$; $[\hat{L}_l, \hat{L}_k] = i\hbar \sum_m \epsilon_{k,l,m} \hat{L}_m$ satisfying (E:1)–(E:3) read

$$\forall k,l : [\widehat{L}_k, \widehat{L}_l] = 0, \quad [\widehat{L}_k, \widehat{L}_l] = -i\hbar \sum_{m=1}^{3} \epsilon_{k,l,m} \widehat{L}_m. \tag{14}$$

Postulate (E:1) uniquely defines the form of any right operator $\widehat{F}$. Indeed, the images of the expressions $i[\hat{F}, \hat{\rho}']$ and $[\hat{F}, \hat{\rho}']_+$ must be real for any Hermitian $\hat{\rho}'$. Thus, the operators $i(\widehat{F} - \widehat{F})$ and $(\widehat{F} + \widehat{F})$ have to be real, i.e.,

$$\widehat{F} = \widehat{F}^* \tag{15}$$

[cf. (6) and (8)]. Relation (15) allows us to define $\widehat{L}_k = \vec{L}_{re,k} + i\vec{L}_{im,k}$ and $\widehat{L}_k = \vec{L}_{re,k} - i\vec{L}_{im,k}$ and rewrite Eqs. (14) in the equivalent form

$$\forall k,l : [\vec{L}_{re,k}, \vec{L}_{im,k}] = 0; \tag{16a}$$

$$[\vec{L}_{im,l}, \vec{L}_{im,k}] = [\vec{L}_{re,k}, \vec{L}_{re,l}] = \frac{\hbar}{2}\sum_{m=1}^{3} \epsilon_{k,l,m}\vec{L}_{im,m}, \tag{16b}$$

$$[\vec{L}_{re,k}, \vec{L}_{im,l}] = \frac{\hbar}{2}\sum_{m=1}^{3} \epsilon_{k,l,m}\vec{L}_{re,m}. \tag{16c}$$

Using Eq. (11) and the free rigid top Hamiltonian

$$\hat{H} = \sum_{k=1}^{3} \frac{\hat{L}_k^2}{2I_k}, \tag{17}$$

one obtains

$$\widehat{\mathscr{L}} = 2\sum_{k=1}^{3} \frac{\vec{L}_{re,k}\vec{L}_{im,k}}{\hbar I_k}. \tag{18}$$

Applying (E:3) to (18) leads to the following expression for $\vec{L}_{im,k}$:

$$\vec{L}_{im,k} = \frac{1}{2}\hbar \sum_{i,j=1}^{3} \epsilon_{i,j,k} L_i \frac{\partial}{\partial L_j} \tag{19}$$

up to terms of order $\hbar^2$ that should be chosen equal to zero in order to best satisfy our requirement of simplicity of $\widetilde{\mathscr{L}}$. The system of differential equations (19) and (16) can be solved for $\vec{L}_{\mathrm{re},k}$:

$$\vec{L}_{\mathrm{re},k} = L_k + \frac{\hbar^2}{16}\left(-2\sum_{i=1}^{3} L_i \frac{\partial}{\partial L_i}\frac{\partial}{\partial L_k} \right.$$
$$\left. + L_k \sum_{i=1}^{3} \frac{\partial^2}{\partial L_i^2} + c_1 \frac{\partial}{\partial L_k} + \xi^2 \frac{L_k}{L^2} \right), \quad (20)$$

where we define $L = \sqrt{L_1^2 + L_2^2 + L_3^2}$. The angular momentum components defined by (19) and (20) satisfy the relations (14) and (15) for any real values of c_1 and ξ, but only the choice $c_1 = -3$ is consistent with the Hermiticity condition (4). The value of ξ can be selected to simplify the expression for averages originating from (E:2):

$$\langle \hat{F} \rangle = (\widehat{\mathfrak{i}}, \widetilde{F}\widehat{\rho})_{\mathrm{W}} \quad (21)$$

[cf. Eq. (2)], where $\widehat{\mathfrak{i}}$ is the Weyl symbol of the reduced identity matrix $\hat{\mathfrak{i}} = \sum_l \hat{\mathfrak{i}}_l$. Here $\hat{\mathfrak{i}}_l = \sum_{m,k=-l}^{l} \hat{\rho}_{l,k;l,k}$, where $\hat{\rho}_{l,k_1;l,k_2} = \frac{1}{2l+1}\mathrm{Tr}_m[|l,m,k_1\rangle\langle l,m,k_2|]$ are reduced projectors that satisfy the standard relations $\hat{L}^2 \hat{\rho}_{l,k_1;l,k_2} = \hbar l(l+1)\hat{\rho}_{l,k_1;l,k_2}$, $\hat{L}_3 \hat{\rho}_{l,k_1;l,k_2} = \hbar k_1 \hat{\rho}_{l,k_1;l,k_2}$, and $\mathrm{Tr}[\hat{\rho}_{l,k_1;l,k_2}\hat{\rho}_{l,k_3;l,k_4}] = \delta_{k_1,k_4}\delta_{k_2,k_3}$. The trace $\mathrm{Tr}_m[\hat{\mathfrak{O}}]$ is taken only over the quantum number m of the projection of the angular momentum on an arbitrary laboratory-fixed axis. The results of Appendix A indicate that the following isomorphism holds between the Weyl symbols $\widehat{\rho}$ and the reduced density matrices $\hat{\rho}_{\mathrm{red}} = \mathrm{Tr}_m[\hat{\rho}]$ such that $\mathrm{Tr}[\hat{\rho}_{\mathrm{red}}\hat{\rho}_{l,k_1;l_2,k_2}] = 0$ for any $l_1 \neq l_2$ (the origin of the latter limitation will be clarified below):

$$\widehat{\rho} = \sum_{l,k_1,k_2} \mathrm{Tr}[\hat{\rho}_{l,k_2;l,k_1}\hat{\rho}_{\mathrm{red}}]\widehat{\rho}_{l,k_1;l,k_2}, \quad (22a)$$

$$\hat{\rho}_{\mathrm{red}} = \sum_{l,k_1,k_2} (\widehat{\rho}_{l,k_1;l,k_2},\widehat{\rho})_{\mathrm{W}}\hat{\rho}_{l,k_1;l,k_2}. \quad (22b)$$

The general expressions for basis functions $\widehat{\rho}_{l,k_1;l,k_2}$ valid for arbitrary values of ξ are given in Appendix A. They take the most convenient nonsingular form when $\xi = \frac{1}{2}$. For this case Eqs. (A5) and (A6) give $\widehat{\mathfrak{i}}|_{\xi=1/2} = \frac{1}{\hbar\sqrt{\pi L}}$ and

$$\widehat{\rho}_{l,l;l,l}|_{\xi=1/2} = \frac{4}{\hbar\sqrt{\pi L}}(-1)^{2l}e^{-4L/\hbar}\mathcal{L}_{2l}\left[\frac{4(L+L_3)}{\hbar}\right], \quad (23)$$

$$\widehat{\rho}_{l,k_1;l,k_2} = c(\widehat{L}_1 + i\widehat{L}_2)^{l-k_1}(\widehat{L}_1 - i\widehat{L}_2)^{l-k_2}\widehat{\rho}_{l,l;l,l}, \quad (24)$$

where $c = \frac{\hbar^{k_1+k_2-2l}}{(2l)!}\sqrt{\frac{(l+k_1)!(l+k_2)!}{(l-k_1)!(l-k_2)!}}$ and the notation $\mathcal{L}_{2l}$ stands for Laguerre polynomials.

It is useful to highlight several peculiarities of the obtained representation.

1. In the angular momentum case, Eqs. (2) and (3) cannot be simultaneously satisfied because of nonuniform density of quantum states in Σ_{W}^L: $\widehat{\mathfrak{i}} \neq 1$. However, one may set $c_1 = -4$, $\xi = 0$ to satisfy (2) instead of (3), which is equivalent to the

nonunitary transformation

$$\widehat{\rho}' = (\hbar^2 \pi L)^\eta \widehat{\rho}|_{\xi=1/2,c_1=-3}, \quad \widetilde{F}' = L^\eta \widetilde{F}|_{\xi=1/2,c_1=-3}L^{-\eta}, \quad (25)$$

with $\eta = -1/2$.

2. The conceptual drawback of the Wigner representations corresponding to the choices $c_1 = -3$, $\xi = 1/2$ and $c_1 = -4$, $\xi = 0$ is that the associated Weyl symbols $\widehat{\mathfrak{i}} = \frac{1}{\hbar\sqrt{\pi L}}$ and $\widehat{\mathfrak{i}}' = \frac{1}{\hbar^2\pi L}$ are not equivalent to the Bopp operator $\widehat{\mathfrak{i}} = 1$ of the identity matrix. This drawback can be eliminated by the choice $c_1 = -2$, $\xi = 0$, which corresponds to $\eta = \frac{1}{2}$ in (25). The resulting formalism will be referred to as the $\Sigma_{\mathrm{W}}^{L\star}$ representation. It is straightforward to check that $\widehat{\mathfrak{i}}^\star = \widehat{\mathfrak{i}}^\star = 1$ so that the associated Bopp operators $\widehat{O}^\star$ and Weyl symbols $\widehat{O}^\star$ and $\widehat{\rho}_{\mathrm{red}}^\star$ of any reduced density matrix $\hat{\rho}_{\mathrm{red}}$ and operator $\hat{O}$ obey the following simple correspondence rules:

$$\widehat{O}^\star = \widehat{O}^\star\widehat{\mathfrak{i}}^\star = \widehat{O}^\star 1 = \widehat{\mathscr{W}}_{\mathrm{dir}}(\hat{O}), \quad \widehat{\rho}^\star = \widehat{\mathscr{W}}_{\mathrm{dir}}(\hat{\rho}_{\mathrm{red}}); \quad (26a)$$

$$\hat{\rho}_{\mathrm{red}} = \mathscr{W}_{\mathrm{rev}}(\widehat{\rho}^\star), \quad \hat{O} = \mathscr{W}_{\mathrm{rev}}(\widehat{O}^\star), \quad (26b)$$

where the direct and reverse transforms are defined as

$$\widehat{\mathscr{W}}_{\mathrm{dir}}(\hat{\mathfrak{O}}) = \mathrm{Tr}[\hat{\mathfrak{O}}\widehat{\hat{\Delta}}], \quad \mathscr{W}_{\mathrm{rev}}(\widehat{\mathfrak{O}}) = \left(\frac{1}{\hbar^2(\pi L)},\widehat{\hat{\Delta}}\widehat{\mathfrak{O}}\right)_{\mathrm{W}}. \quad (27)$$

Here $\widehat{\hat{\Delta}}$ is the Stratonovich-Weyl (SW) kernel

$$\widehat{\hat{\Delta}} = \sum_{l,k_1,k_2} \widehat{\rho}_{l,k_1;l,k_2}^\star \hat{\rho}_{l,k_2;l,k_1} \quad (28)$$

and the basis functions $\widehat{\rho}_{l,k_1;l,k_2}$ are related to those defined by Eqs. (23) and (24) as

$$\widehat{\rho}_{l,k_1;l,k_2}^\star = \hbar(\pi L)^{1/2}\widehat{\rho}_{l,k_1;l,k_2}. \quad (29)$$

Remarkably, the mathematical structure of the resulting Wigner images $\widehat{L}_k^\star$ is identical (up to the complex conjugation) to the conventional generalized Bopp operators for spin [12,52]. This analogy makes it evident that the correspondences (26) allow us to define the familiar phase-space star product of any two Weyl symbols

$$\widehat{\mathfrak{O}}_1^\star \star \widehat{\mathfrak{O}}_2^\star = \widehat{\mathfrak{O}}_1^\star \widehat{\mathfrak{O}}_2^\star = \widehat{\mathscr{W}}_{\mathrm{dir}}[\mathscr{W}_{\mathrm{rev}}(\widehat{\mathfrak{O}}_1^\star)\mathscr{W}_{\mathrm{rev}}(\widehat{\mathfrak{O}}_2^\star)], \quad (30)$$

so that, e.g.,

$$\langle \hat{F} \rangle = \left(\frac{1}{\hbar^2(\pi L)},\widetilde{F}^\star\widehat{\rho}^\star\right)_{\mathrm{W}} = \left(\frac{1}{\hbar^2(\pi L)},\widehat{F}^\star\star\widehat{\rho}^\star\right)_{\mathrm{W}}. \quad (31)$$

In principle, one can similarly construct the star product version of any generalized Wigner quantizer. However, as we will see in Sec. IV, in the general case there is no guarantee that complete consistency with the SW formalism will be achieved. This is the price one has to pay for extended capabilities of algorithm II to give the quantizer's desirable dynamic properties.

3. It follows from Eq. (16b) that $\frac{1}{2}(\widehat{L}_i + \widehat{L}_i) = \vec{L}_{\mathrm{re},i} \neq L_i$ regardless of the particular choice of averaging and normalization. This precludes the analogs of relations (9) for angular

components $\widehat{L}_k$, so one can no longer obtain meaningful marginal distributions via partial integration over $\widehat{\rho}$. Nevertheless, other characteristic semiclassical features of the Wigner representation remain preserved. Specifically, one can still apply the recipe from [48,49] to relate the Wigner equations of motion (10) for pure states (with $c_1 = -3$, $\xi = 1/2$) to the respective classical equations in standard or Koopman–von Neumann form.

4. The truncated Euler phase space Σ_W^L is incapable of handling the orientation of top relative to the laboratory frame S'. In particular, we cannot define the $\vec{e}\,'_3$ projection of the angular momentum and the associated quantum number m. Also, it is easy to verify the equality $\widehat{L}^2 = \widehat{L}^2$, which gives rise to the relation $[\hat{L}^2, \hat{\rho}_{red}] = (\widehat{L}^2 - \widehat{L}^2)\widehat{\rho} = 0$ and many-to-one ambiguity

$$\forall \alpha : \alpha \hat{L}^2 \hat{\rho}_{red} + (1-\alpha)\hat{\rho}_{red}\hat{L}^2 \leftrightarrow \widehat{L^2\rho}. \qquad (32)$$

Consequently, the feasible density matrices $\hat{\rho}_{red}$ must obey the condition $\forall l_1 \neq l_2 : \mathrm{Tr}[\hat{\rho}_{l_1,k_1;l_2,k_2}\hat{\rho}_{red}] = 0$, which justifies the specific form of the isomorphism (22). Beyond that, the equality $\widehat{L}^2 = \widehat{L}^2$ implies that the quantum Liouville operator of the free spherical top exactly coincides with its classical counterpart $\widehat{\mathscr{L}} = \mathscr{L} = 0$.

5. By virtue of the many-to-one ambiguousness (32), the equation $\widehat{L}^2 \hat{\imath}_l = \hbar l(l+1)\hat{\imath}_l$ has bounded, isotropic, Lebesgue, and square integrable in Σ_W solutions $\hat{\imath}_l$ for any real $l > -\frac{1}{2}$ [see Eq. (A4)]. However, the coefficients $\kappa_{l,j}$ in the expansion $\hat{\imath}_l = \sum_{j=0}^{\infty} \kappa_{l,j}\hat{\imath}_{j/2}$ take negative values for noninteger values of $2l$ since $\kappa_{l,j}|_{2l \notin \mathbb{Z}, j \to \infty} \propto \frac{(-1)^j}{j}$. Consequently, $\hat{\imath}_l|_{2l \notin \mathbb{Z}}$ are Weyl symbols of nonpositive operators and do not represent valid physical states. These properties should be considered with caution in calculations because they indicate that a small numerical error can result in a dramatic physical mistake.

IV. COMPLETE PHASE-SPACE REPRESENTATION OF ROTATIONAL MOTION

Various sets of generalized coordinates enable establishing the link of the rotational dynamics with the laboratory frame missed in the Euler quantization picture. However, the evolution equations take the most elegant form in terms of the four quaternions λ_k defined as

$$\lambda_0 = \cos\frac{\Phi}{2}, \quad \lambda_k = \eta_k \sin\frac{\Phi}{2} \quad (k = 1,2,3), \qquad (33)$$

where the parameters $\vec{\eta}$ and Φ are such that rotation about the vector $\vec{\eta}$ by angle $-\Phi$ will superimpose the axes $\vec{e}_k$ and $\vec{e}\,'_k$ of the moving and laboratory frames S and S' (Fig. 1). Unlike angular variables, the quaternions are true canonical coordinates (in the sense Ref. [19]). This makes the construction of the Wigner representation in terms of λ_i and the associated canonically conjugated momenta $p_{\lambda,i}$ straightforward since the associated Bopp operators obey the canonical commutation relations identical to (6) and (8) [35,36] (see Appendix B for details and a brief review of the algebra of quaternions). However, extra dimensionality
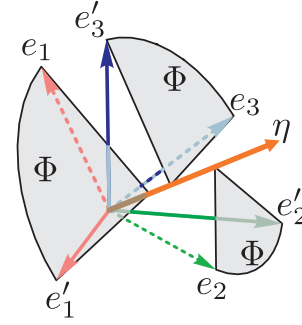

FIG. 1. (Color online) Physical meaning of the parameters entering the definition (33) of the quaternions λ_k.

of the phase space makes this approach computationally impractical.

In order to solve this problem while keeping the simple form of the dynamical equations, we will consider the noncanonical phase space $\Sigma_W^{\lambda,L'}$ composed of λ_k and the projections L'_k of the angular momentum on the laboratory axes. The corresponding classical Liouville operator reads

$$\mathscr{L} = \frac{1}{2}\sum_{k=1}^{3}\sum_{m,n=0}^{3} m_{m,n,k}^{Q}\omega_k \lambda_m \frac{\partial}{\partial \lambda_n}, \qquad (34)$$

where $m_{m,n,k}^{Q}$ are quaternion multiplication coefficients

$$m_{k,i,j}^{Q}$$
$$= \begin{cases} \epsilon_{i,j,k} & \text{if } i > 0 \wedge j > 0 \wedge k > 0 \\ \delta_{j,0}\delta_{k,i} + \delta_{i,0}\delta_{k,j} - \delta_{k,0}\delta_{i,j} & \text{otherwise} \end{cases} \qquad (35)$$

and $\omega_k = \sum_{j=1}^{3} Q_{k,j}L'_j/I_k$ are the angular frequencies about axes $\vec{e}_k$. The entries $Q_{i,j}$ of the directional cosine matrix are bilinear in terms of λ_k, $Q_{i,j} = (\vec{e}_i, \vec{e}\,'_j) = \sum_{m,n} q_{i,j,m,n}\lambda_m\lambda_n$, with coefficients

$$q_{i,j,m,n} = (1 - 2\delta_{j,m})\sum_{k=0}^{3} m_{i,j,k}^{Q} m_{k,m,n}^{Q}. \qquad (36)$$

In choosing a strategy to quantize Eqs. (33) and (34), we will follow the reasoning of the previous section and define a set of postulates similar to (E:1)–(E:3):

(C:1) the enforced reality condition similar to (E:1),

(C:2) the traciality relation (3) [with $(\widehat{\odot}_1, \widehat{\odot}_2)_W \overset{\text{def}}{=} \int_{\Sigma_W^{\lambda,L'}} \widehat{\odot}_1^* \, \widehat{\odot}_2 \, dL_1 dL_2 dL_3 d\lambda_0 d\lambda_1 d\lambda_2 d\lambda_3$],

(C:3) (C:3) the proper classical limits

$$\widehat{L}'_i|_{\hbar \to 0} = L'_i, \quad \widehat{L}_i|_{\hbar \to 0} = L_i, \quad \widehat{\mathscr{L}}|_{\hbar \to 0} = \mathscr{L}, \quad \widehat{\lambda}_j|_{\hbar \to 0} = \lambda_i, \qquad (37)$$

together with the commutation relations (14)–(16) and

$$\frac{2}{i\hbar}[\hat{L}_i, \hat{\lambda}_j] = \sum_{k=1}^{3} m_{k,j,i}^{Q}\hat{\lambda}_k. \qquad (38)$$

As in Sec. III, our intention is to make use of the variability in the framework of the above postulates in favor of the simplest form of the phase-space Liouville operator $\widehat{\mathscr{L}}$.

Reproducing the steps leading to Eqs. (19) and (20), one gets

$$\widetilde{L}_k = \frac{1}{2}\left[\frac{\sum_{s=1}^{3} Q_{k,s} L'_s}{\sum_{s=0}^{3} \lambda_s^2}, 1 - \frac{\hbar^2}{16}\sum_{s=1}^{3}\frac{\partial^2}{\partial L_s'^2}\right]_+$$
$$+ \sum_{m,n=0}^{3}\left(\frac{\hbar^2}{16}\sum_{s=1}^{3} q_{k,s,m,n}\frac{\partial}{\partial L'_s} + i\frac{\hbar}{4}m^{\mathrm{Q}}_{m,n,k}\right)\lambda_m\frac{\partial}{\partial \lambda_n},$$
(39)

$$\widetilde{\lambda}_k = \widetilde{N}^{-\frac{1}{2}}\left(\lambda_k + i\frac{\hbar}{4}\sum_{m=0}^{3}\sum_{n=1}^{3} m^{\mathrm{Q}}_{k,n,m}\lambda_m\frac{\partial}{\partial L'_n}\right),$$
(40)

$$\widetilde{N} = \left(\sum_{s=0}^{3}\lambda_s^2\right)\left(1 - \frac{\hbar^2}{16}\sum_{s=1}^{3}\frac{\partial^2}{\partial L_s'^2}\right),$$
(41)

where the operator $\widetilde{N}$ commutes with all physical observables of the form $F(\widetilde{L}'_k, \widetilde{\lambda}_n)$. The existence of such $\widetilde{N} \neq 1$ is due to overcompleteness of our seven-dimensional phase space $\Sigma_{\mathrm{W}}^{\lambda, L'}$.

The Wigner representation (39)–(41) is not convenient for exact numerical implementation. One problem is caused by the term $\widetilde{N}^{-\frac{1}{2}}$ in the expression (40) for the quaternion images, which is a differential operator of infinite order. This prefactor, however, can be ignored when choosing to work only with images of states satisfying the equation $\widetilde{N}\widehat{\rho} = \widehat{\rho}$ (which is possible owing to its commutation properties). Another possibility is to Fourier or Laplace transform the phase space $\Sigma_{\mathrm{W}}^{\lambda, L'}$ with respect to L'_1, L'_2, and L'_3. However, the resulting equations will lose the key characteristic properties of the Wigner representation. Another technical complication is introduced by the excessive dimensionality of $\Sigma_{\mathrm{W}}^{\lambda, L'}$. In addition, unlike the classical generator of free motion (34), the quantum counterpart (11) no longer evidently manifests the angular momentum conservation by preserving the values of L'_k. One would desire to retain this remarkable property of Eq. (34) in the quantum case because it would allow reduction of the seven-dimensional differential propagation equation to a series of four-dimensional ones.

We found that these issues can be resolved by relaxing the traciality requirement (C:2) in favor of more explicit specification of the desired form of the Bopp operators $\widetilde{L}_k$. The resulting modified set of postulates imposes:

(C̄:1) the reality condition (15),

(C̄:2) the requirement (37) of well-defined classical limits,

(C̄:3) the requirement of the absence of derivatives over L'_1, L'_2, and L'_3 in the expressions for the images $\widetilde{L}_k$.

The expressions for $\widetilde{L}_k$ satisfying these postulates can be compactly written in terms of the variables $\Lambda_m = \sqrt{L'}\sqrt{8/\hbar}\lambda_m$:

$$\widetilde{L}_k = \frac{\hbar}{8}\sum_{m,n=0}^{3}\left[\sum_{s=1}^{3} q_{k,s,m,n}\frac{L'_s}{L'}\left(\Lambda_m\Lambda_n - \frac{\partial^2}{\partial\Lambda_m\partial\Lambda_n}\right)\right.$$
$$\left. + 2i\,m^{\mathrm{Q}}_{m,n,k}\Lambda_m\frac{\partial}{\partial\Lambda_n}\right],$$
(42)

where $L' = \sqrt{\sum_{k=1}^{3} L_k'^2}$. It is useful to introduce the intermediate fixed frame S'' whose third axis $\vec{e}''_3$ coincides with the (conserved) direction of the angular moment. We denote by $\frac{1}{L'}\boldsymbol{q}$ the quaternion that represents the rotation connecting S' and S'' and introduce the parameters λ_m as exact analogs of the parameters Λ_m characterizing the orientation of the rotor relative to S'':

$$L'_n = \sum_{m,n=0}^{3} q_{i,j,m,n}\mathscr{q}_i\mathscr{q}_j, \quad \Lambda_k = \frac{\sum_{i,j=0}^{3} m^{\mathrm{Q}}_{k,i,j}\mathscr{q}_i\lambda_j}{\sqrt{\sum_{n=0}^{3}\mathscr{q}_n^2}}. \quad (43)$$

Note that Eqs. (43) do not fix the directions of the axes $\vec{e}''_1$ and $\vec{e}''_2$ of S'' and so do not uniquely define $\mathscr{q}_k$. Equation (42) takes simple forms in terms of the variables λ_m:

$$\widetilde{L}_1 \pm i\widetilde{L}_2 = \hbar\widehat{a}^{\dagger}_{\mp}\widehat{a}_{\pm}, \quad \widetilde{L}_3 = \frac{\hbar}{2}(\widehat{a}^{\dagger}_{+}\widehat{a}_{+} - \widehat{a}^{\dagger}_{-}\widehat{a}_{-}), \quad (44)$$

where $\widehat{a}^{\dagger}_{\pm}$ and $\widehat{a}_{\pm}$ are the conventional ladder operators

$$\widehat{a}^{\dagger}_{\pm} + \widehat{a}_{\pm} = \lambda_{2\pm1} \pm i\frac{\partial}{\partial\lambda_{1\mp1}}, \quad \widehat{a}_{\pm} - \widehat{a}^{\dagger}_{\pm} = \frac{\partial}{\partial\lambda_{2\pm1}} \pm i\lambda_{1\mp1};$$
(45)

$$[\widehat{a}_{\pm}, \widehat{a}^{\dagger}_{\pm}] = 1, \quad [\widehat{a}_{\mp}, \widehat{a}^{\dagger}_{\pm}] = [\widehat{a}_{\mp}, \widehat{a}_{\pm}] = 0. \quad (46)$$

(It is worth stressing that notation such as $\widehat{a}^{\dagger}_{\pm}$ hereafter means the Wigner image of the creation operator $\widehat{a}^{\dagger}$, not the Hermitian conjugate of the Bopp operator $\widehat{a}_{\pm}$.)

The fact that $\widehat{a}^{\dagger}_{\pm}$ and $\widehat{a}_{\pm}$ do not depend on $\mathscr{q}_k$ and the commutation relation $\forall m,n : [\widehat{L}_m, \widehat{L}'_n] = 0$ hints that the images $\widetilde{L}'_m$ should have a form similar to (44),

$$\widetilde{L}'_1 \pm i\widetilde{L}'_2 = \hbar\widehat{b}^{\dagger}_{\pm}\widehat{b}_{\mp}, \quad \widetilde{L}'_3 = \frac{\hbar}{2}(\widehat{b}^{\dagger}_{+}\widehat{b}_{+} - \widehat{b}^{\dagger}_{-}\widehat{b}_{-}), \quad (47)$$

where the operators $\widehat{b}^{\dagger}_{\pm}$ and $\widehat{b}_{\pm}$ do not depend on λ_k and satisfy commutation relations identical to (46). The validity of Eqs. (47) is proven in Appendix C, where the following explicit expressions are obtained [up to invariance transformation (52); see below]:

$$\widehat{b}^{\dagger}_{\pm} = \sqrt{2}(\mathscr{q}_{1\mp1} + i\mathscr{q}_{2\pm1}), \quad \widehat{b}_{\pm} = \frac{1}{\sqrt{8}}\left(\frac{\partial}{\partial\mathscr{q}_{1\mp1}} - i\frac{\partial}{\partial\mathscr{q}_{2\pm1}}\right);$$
(48)

$$\widetilde{L}'_k = \frac{L'_k}{L'}\frac{\widehat{\ell} + \widehat{\ell}}{2} - \frac{\hbar}{4}\sum_{r=0}^{3}\sum_{s=1}^{3} m^{\mathrm{Q}}_{k,r,s}\left((1 - \delta_{r,0})\frac{L'_r}{L'} + i\delta_{r,0}\right)$$
$$\times \sum_{m,n=0}^{3} m^{\mathrm{Q}}_{n,s,m}\left(2(1 - \delta_{m\times n,0})L'_m\frac{\partial}{\partial L'_n} + \Lambda_m\frac{\partial}{\partial\Lambda_n}\right),$$
(49)

where $\widehat{\ell}$ is the image of operator of quantum number l:

$$\widehat{\ell}(\widehat{\ell} + \hbar) = \sum_{k=1}^{3}\widetilde{L}_k^2 = \sum_{k=1}^{3}\widetilde{L}_k'^2 = \widetilde{L}^2, \quad (50)$$

$$\widehat{\ell} = \frac{\hbar}{8} \sum_{m=0}^{3} \left(\Lambda_m^2 - \frac{\partial^2}{\partial \Lambda_m^2} - 1 - 2i \right.$$
$$\left. \times \sum_{k=1}^{3} \sum_{n=0}^{3} m_{n,k,m}^{\mathrm{Q}} \frac{L_k'}{L'} \Lambda_m \frac{\partial}{\partial \Lambda_n} \right). \quad (51)$$

We emphasize several important properties of the Wigner quantizer generated by operators (42) and (49).

1. The following invariance relations hold for any operator $\mathcal{S}$ representable as a function of only $\widehat{\ell}$, $\overleftarrow{\ell}$, L', and the operator $\overleftrightarrow{Q} = \sum_{s=1}^{3} L_s' \frac{\partial}{\partial L_s'}$:

$$\widehat{L}_k = \mathcal{S} \widehat{L}_k \mathcal{S}^{-1}, \quad \widehat{L}_k' = \mathcal{S} \widehat{L}_k' \mathcal{S}^{-1}. \quad (52)$$

In particular, if $\widehat{\rho}_\epsilon$ is the solution of the eigenvalue problem $f(\widehat{L}_k, \widehat{L}_l', \widehat{L}_m, \widehat{L}_n', L') \rho_\epsilon = \epsilon \rho_\epsilon$ with arbitrary function f, then $\mathcal{S} \rho_\epsilon$ is also its solution. Furthermore, the variables replacement $L_k' \to \epsilon L_k'$ with arbitrary ϵ does not change the form of the operators (49), (42), and (51). This implies that the basis function $\widehat{\rho}_{\alpha,\beta}$ corresponding to an arbitrary projector $\hat{\rho}_{\alpha,\beta} = |l_\alpha, m_\alpha, k_\alpha\rangle \langle l_\beta, m_\beta, k_\beta|$ can be written as

$$\widehat{\rho}_{\alpha,\beta} = r_{L',l_\alpha,l_\beta} \widehat{\rho}_{\alpha,\beta}^{(0)} \left(\boldsymbol{\Lambda}, \frac{L_1'}{L'}, \frac{L_2'}{L'}, \frac{L_3'}{L'} \right), \quad (53)$$

where the variable prefactor $\overleftrightarrow{r}_l(L')$ depends on the choice of $\mathcal{S}$ in the invariance relation (52).

2. The Bopp operators $\widehat{L}_k$, $\widehat{L}^2$, and $\frac{1}{2}(\widehat{L}_3' - \widehat{L}_3)$ are Hermitian in $\Sigma_{\mathrm{W}}^{\Lambda,L'}$, but the operators $\widehat{L}_k'$ are not:

$$\widehat{L}_k'^\dagger = \widehat{L}_k^*|_{\partial/\partial\Lambda_n \to -\partial/\partial\Lambda_n, \partial/\partial L_m \to -\partial/\partial L_m} + \hbar \frac{L_k'}{L'} \neq \widehat{L}_k'. \quad (54)$$

[The extra term $\hbar(L_k'/L')$ in Eq. (54) arises from symmetrization of the operators (49).] The Bopp operators $\widehat{L}_k'^\dagger$ also fulfill relations (14) and (50) and can be used as an alternative variant of the images of the operators $\hat{L}_k'$. Nevertheless, the original isomorphism $\hat{L}_k' \leftrightarrow \widehat{L}_k'$ results in more convenient forms of the Weyl symbols. Indeed, we denote by $\widehat{\mathrm{i}}_l$ the Weyl symbols of the identity submatrices $\sum_{k,m=-l}^{l} |l,m,k\rangle \langle l,m,k|$ for subspaces with well-defined quantum number l. Each $\widehat{\mathrm{i}}_l$ should be the symmetric solution of the eigenvalue problem $\overleftrightarrow{\ell} \, \widehat{\mathrm{i}}_l = l \widehat{\mathrm{i}}_l$. If the isomorphism $\hat{L}_k' \leftrightarrow \widehat{L}_k'$ is accepted, one obtains

$$\widehat{\mathrm{i}}_l = r_l(L') \mathcal{L}_{2l}^1 \left(\sum_{m=0}^{3} \Lambda_m^2 \right) \exp\left(-\frac{1}{2} \sum_{m=0}^{3} \Lambda_m^2 \right) \quad (55)$$

and $\widehat{\mathrm{i}}_l|_{\Lambda_m \to \pm\infty} = 0$. Conversely, the choice $\hat{L}_k' \leftrightarrow \widehat{L}_k'^\dagger$ leads to the divergent solutions $\widehat{\mathrm{i}}_l \propto \mathcal{L}_{2l}^1(\sum_{m=0}^{3} \Lambda_m^2) \exp(+\frac{1}{2}\sum_{m=0}^{3} \Lambda_m^2)$. This substantially complicates the definition of normalization and the rule for calculation of averages and makes this choice inconvenient. However, even the original definition (42) leads to nonorthogonality of certain basis

functions because of the non-Hermiticity of the operators $\widehat{L}_k'$:

$$\iiint \prod_{k=1}^{3} dL_k \iiiint \prod_{m=0}^{3} d\Lambda_k \rho_{\alpha_1,\beta_1}^* \rho_{\alpha_2,\beta_2} \neq 0$$

for any pair of basis functions such that $l_{\alpha_1} = l_{\alpha_2}$, $l_{\beta_1} = l_{\beta_2}$, $k_{\alpha_1} = k_{\alpha_2}$, and $k_{\beta_1} = k_{\beta_2}$ $m_{\alpha_1} - m_{\alpha_2} = m_{\beta_1} - m_{\beta_2}$, including the cases where $m_{\alpha_1} \neq m_{\alpha_2}$. Thus, the discussed Wigner representation cannot be equipped with a traciality relation similar to (3).

Nevertheless, due to the invariance relation (52), one can define a convenient rule for the calculation of averages by selecting the following L'-independent prefactor r_{L',l_α,l_β} in Eq. (53):

$$r_{l_\alpha,l_\beta} = \frac{(-1)^{l_\alpha + l_\beta}}{16\pi^3} \sqrt{2l_\alpha + 1} \sqrt{2l_\beta + 1}. \quad (56)$$

In this case only six out of the seven arguments of the Wigner function $\widehat{\rho} = \widehat{\rho}(\boldsymbol{\Lambda}, \frac{L_1'}{L'}, \frac{L_2'}{L'}, \frac{L_3'}{L'})$ are independent, so the effective size of the phase space is equal to 6. Furthermore, the averaging rule is defined through the scalar product (1), where

$$(\odot_1^*, \odot_2)_{\mathrm{W}} = \iiiint_{-\infty}^{\infty} d\Lambda_0 d\Lambda_1 d\Lambda_2 d\Lambda_3 \int_\Omega d\boldsymbol{\omega} \frac{\odot_1 \odot_2}{\hbar^2 \kappa^2}. \quad (57)$$

The inner integral $\int_\Omega d\boldsymbol{\omega} \cdots$ in Eq. (57) is taken over the surface of a sphere $L' = \kappa \hbar$ with an arbitrary radius κ.

3. Lack of a traciality relation like (C:2) makes elucidation of the correspondence rule $\hat{\rho} \leftrightarrow \widehat{\rho}$ less straightforward than in the canonical case. Formally, the Weyl symbol $\widehat{\rho}_{\alpha,\beta}$ of any basis function $\hat{\rho}_{\alpha,\beta}$ can be obtained by sequential application of the Schwinger ladder operators (45) and (48) to the ground state $\rho = \widehat{\mathrm{i}}_0$. However, the direct analogs of these operators cannot exist in $\Sigma_{\mathrm{W}}^{\Lambda,L'}$ space because they would result in two independent and conflicting definitions for the operator $\widehat{L}^2$. Nevertheless, the Wigner images $a_\pm \widehat{b}_\pm$ and $a_\pm^\dagger \widehat{b}_\pm^\dagger$ of the compound Schwinger operators $\hat{a}_\pm \hat{b}_\pm$ and $\hat{a}_\pm^\dagger \hat{b}_\pm^\dagger$ are well defined and can be directly deduced by the technique used in Appendix C. The derivation and resulting rather cumbersome expressions are deferred to Appendix D.

The compound ladder operators allow us to explicitly calculate any basis function (53):

$$\widehat{\rho}_{\alpha,\beta} = \overleftrightarrow{\mathscr{R}}_\uparrow(\alpha,\beta) \widehat{\mathrm{i}}_0, \quad \widehat{\mathrm{i}}_0 = \overleftrightarrow{\mathscr{R}}_\downarrow(\alpha,\beta) \widehat{\rho}_{\alpha,\beta}, \quad (58)$$

where

$$\overleftrightarrow{\mathscr{R}}_\uparrow(\alpha,\beta) = \prod_{\kappa,\mu=\pm 1} \frac{(a_\kappa^\dagger \widehat{b}_\mu^\dagger)^{p_{\kappa,\mu}(\alpha)} (a_\kappa^\dagger \widehat{b}_\mu^\dagger)^{p_{\kappa,\mu}(\beta)}}{\prod_{\lambda=m,k} \prod_{\xi=\alpha,\beta} \sqrt{(l_\xi - \lambda_\xi)!(l_\xi + \lambda_\xi)!}}, \quad (59)$$

$$\overleftrightarrow{\mathscr{R}}_\downarrow(\alpha,\beta) = \prod_{\kappa,\mu=\pm 1} \frac{(a_\kappa \widehat{b}_\mu)^{p_{\kappa,\mu}(\alpha)} (a_\kappa \widehat{b}_\mu)^{p_{\kappa,\mu}(\beta)}}{\prod_{\lambda=m,k} \prod_{\xi=\alpha,\beta} \sqrt{(l_\xi - \lambda_\xi)!(l_\xi + \lambda_\xi)!}}, \quad (60)$$

and the factors $p_{\kappa,\mu}$ can be any set of non-negative numbers satisfying the relations $\sum_{\kappa,\mu=\pm 1} p_{\kappa,\mu}(\xi) = 2l_\xi$,

$\sum_{\kappa,\mu=\pm 1} \kappa p_{\kappa,\mu}(\xi) = 2k_\xi$, and $\sum_{\kappa,\mu=\pm 1} \mu p_{\kappa,\mu}(\xi) = 2m_\xi$. Using the operator $\vec{\mathcal{R}}_\downarrow$ and the orthogonality relation $\forall \widehat{\rho}_{\alpha,\beta} \neq \widehat{i}_0 : (\widehat{i}_0, \widehat{\rho}_{\alpha,\beta})_W = 0$ (the latter follows from the Hermiticity of the images $\widehat{L}_k$), one can establish the desired correspondences $\widehat{\rho} = \widetilde{\mathcal{W}}_{\text{dir}}(\widehat{\rho})$ and $\widehat{\rho} = \mathcal{W}_{\text{rev}}(\widehat{\rho})$:

$$\widetilde{\mathcal{W}}_{\text{dir}}(\widehat{\rho}) = \sum_{\alpha,\beta} \vec{\mathcal{R}}_\uparrow(\alpha,\beta) \widehat{i}_0 \, \text{Tr}[\widehat{\rho}_{\alpha,\beta}^\dagger \widehat{\rho}], \qquad (61\text{a})$$

$$\mathcal{W}_{\text{rev}}(\widehat{\rho}) = \sum_{\alpha,\beta} (\widehat{i}_0, \vec{\mathcal{R}}_\downarrow(\alpha,\beta) \widehat{\rho})_W \rho_{\alpha,\beta}$$

$$= \sum_{\alpha,\beta} (\vec{\mathcal{R}}_\downarrow^\dagger(\alpha,\beta) \widehat{i}_0, \widehat{\rho})_W \widehat{\rho}_{\alpha,\beta}. \qquad (61\text{b})$$

It is worth stressing that the notation $\vec{\mathcal{R}}_\downarrow^\dagger(\alpha,\beta)$ means the Hermitian conjugate of the phase-space Bopp operator $\vec{\mathcal{R}}_\downarrow(\alpha,\beta)$ in $\Sigma_W^{\Lambda,L'}$ rather than the Wigner image of the Hermitian conjugate $\widehat{\mathcal{R}}_\downarrow^\dagger$ of the associated operator $\widehat{\mathcal{R}}_\downarrow$.

4. Unlike the Wigner quantization of the Euler equations, the traciality-deficient $\{\Lambda, L'\}$ representation cannot be tuned to become fully consistent with the standard Stratonovich-Weyl quantization scheme. Specifically, it is still possible to achieve the identity $\widehat{i}^\star = \widehat{i}^\star = 1$ by applying the suitable invariance transform (52) with $\mathcal{S} = \mathcal{S}^\star = \frac{256\pi^3}{\sum_{m=0}^3 \Lambda_m^2}$,

$$\widehat{\rho}^\star = \mathcal{S}^\star \widehat{\rho}, \quad \widetilde{F}^\star = S^\star \widetilde{F}(\mathcal{S}^\star)^{-1}, \qquad (62)$$

and introduce the Weyl symbols of operators $\widehat{F}^\star = \widetilde{F}^\star \widehat{i}^\star$. However, the inequality $\vec{\mathcal{R}}_\uparrow^\star(\alpha,\beta) \neq \vec{\mathcal{R}}_\downarrow^{\star\dagger}(\alpha,\beta)$ makes it impossible to define the Stratonovich-Weyl kernel similar to (28). Instead, one has to introduce the direct and reverse transforms $\widetilde{\mathcal{W}}_{\text{dir}}^\star(\widehat{\odot}) = S^\star \widetilde{\mathcal{W}}_{\text{dir}}(\widehat{\odot})$ and $\mathcal{W}_{\text{rev}}^\star(\widehat{\odot}) = \mathcal{W}_{\text{rev}}[(S^\star)^{-1} \widehat{\odot}]$ as independent operations. These transforms nevertheless allow us to define the analog of the star product algebra similar to (30) so that, e.g.,

$$\langle \widehat{F} \rangle = (\mathcal{S}^{-1}, \widetilde{F}\widehat{\rho})_W = (\mathcal{S}^{-1}, \widetilde{F} \star \widehat{\rho})_W. \qquad (63)$$

5. None of the expressions $\frac{1}{2}(\widehat{L}'_k + \widehat{L}'_k)$ and $\frac{1}{2}(\widehat{\lambda}'_n + \widehat{\lambda}'_n)$ coincides with its classical analog L_k, L'_k, and λ_n, hence the marginal distributions associated with the Wigner function $\widehat{\rho}$ have no exact physical meaning. Instead, any of the representations $\{\Lambda, L'\}$, $\{\Lambda, q\}$, and $\{\lambda, \mathbf{q}\}$ allow us to easily cast the generators of motion $\widehat{\mathcal{L}}$ for free linear, spherical, or symmetric tops with principal moments of inertia $I_1 = I_2 \neq I_3$ in the familiar classical-like form

$$\widehat{\mathcal{L}} = -A\vec{\mathfrak{J}} \frac{\partial}{\partial \alpha} + (A-B)\vec{\mathfrak{K}} \frac{\partial}{\partial \gamma}, \qquad (64)$$

where $A = \frac{1}{I_1} = \frac{1}{I_2}$, $B = \frac{1}{I_3}$ ($B = \vec{\mathfrak{K}} = 0$ for linear tops), and

$$\vec{\mathfrak{J}} = \frac{\widehat{\ell} + \widehat{\ell} + \hbar}{2}, \quad \vec{\mathfrak{K}} = \frac{\widehat{L}_3 + \widehat{L}_3}{2}, \qquad (65)$$

by expressing the parameters λ_m in terms of the Euler angles α, β, and γ relating the frames S' and S'' [53]:

$$\lambda_0 \pm i\lambda_3 = \sqrt{\frac{8L'}{\hbar}} \cos\left[\frac{\beta}{2}\right] \exp\left[\pm i\frac{\alpha+\gamma}{2}\right],$$

$$\lambda_1 \pm i\lambda_2 = \sqrt{\frac{8L'}{\hbar}} \sin\left[\frac{\beta}{2}\right] \exp\left[\pm i\frac{\alpha-\gamma}{2}\right]. \qquad (66)$$

(Note that in the definitions of Euler angles we adopted the conventions of Edmonds [54] and Zare [55].)

The only difference between the quantum Liouville operator (64) and its classical counterpart is that the classical variables L' and L'_3 are replaced with the quantum Bopp operators $\vec{\mathfrak{J}}$ and $\vec{\mathfrak{K}}$ with discrete spectra. That is, the values of axial and precession frequencies of the quantum top can take only a discrete set of equidistant values $(A-B)\frac{\hbar}{2}k$ and $A\frac{\hbar}{2}(|j|+1)$ $(k, j \in \mathbb{Z})$. which gives rise to the familiar phenomenon of quantum rotational revivals.

The analogy to the classical case can be pushed even further by forcing the operators $\widehat{\ell}$ and $\widehat{L}_3$ to take the mathematical structure of canonical Bopp operators $\widehat{x}_j$ and $\widehat{p}_j$ [Eq. (6)] via an appropriate variable transformation. By comparing the form (65) of the operators $\vec{\mathfrak{J}}$ and $\vec{\mathfrak{K}}$ with the fact that $\frac{\widehat{x}_j + \widehat{x}_j}{2} = x_j$, we can expect that such a transformation will lead us to a Nasyrov-type Wigner quantizer [38], in which the quantum generator of motion (64) is identical to the classical one: $\widehat{\mathcal{L}} = \mathcal{L}$. The derivation and the properties of this representation are detailed in Appendix E. Its existence leads to the remarkable and intriguing conclusion that the free symmetric top shares with the free particle and harmonic oscillator the exceptional property of having identical classical and quantum dynamics.

V. CONCLUSION

In this work we introduced a semiphenomenological quantization scheme on nonstandard phase spaces and validated its utility by finding useful generalized Wigner quantizers of rotation motion. Specifically, the truncated quantizer derived in Sec. III allows one to perform the density-matrix-type calculations within the wave-function-sized rotational phase space of three parameters and to fully account for any rotational effects in isotropic environments on the intramolecular dynamics. Thus, it may be useful, for instance, in calculations of emission spectra or dissociation rates resulting from pulsed laser excitation. We have also shown that there exists a large family of quantizers (parametrized by c_1 and ξ), including the two variants that are especially convenient for calculation of averages or the normalization of quasiprobability distributions, and a version fully consistent with the Stratonovich-Weyl formalism. One can easily switch between these representations via the simple nonunitary transformation developed. Along with its practical potential, this quantizer also has the pedagogical value of establishing the bridge between the formal quantization of the spin degrees of freedom [12] and the classical Euler equations.

The second proposed $\{\Lambda, L'\}$ quantizer has the important feature of translating the angular momentum conservation laws into conservation of the parameters L'_1, L'_2, and L'_3 in the course of free rotations. This feature allows for natural parallelization of the code via splitting the initial six-dimensional problem into a series of independent four-dimensional ones for evolution of the parameters Λ_m ($m = 0, \ldots, 3$). Note that several known representations (see, e.g., [37,38]) allow a similar trick. However, their generalized parametric spaces are not singularity-free and suffer from the gimbal lock problem. The latter problem can be resolved in the framework of the standard Wigner quantization procedure only by introducing artificial degrees of freedom [35,36]. In contrast, both of the proposed quantizers resolve the gimbal lock issue without paying this price [we recall that the $\{\Lambda, L'\}$ quantizer with the scalar product (57) is effectively six dimensional]. That is, they allow convenient and low-dimensional grid discretizations in numerical dynamical simulations. In addition, they benefit from expressing the generators of free motion as low-order differential operators of continuous arguments. This should facilitate relatively inexpensive propagation of the evolution equations and is an important prerequisite for effective application of the initial value approximations.

On the conceptual level, our findings uncover the direct connection [Eq. (44)] between the quaternion parameters and the raising and lowering operators entering the Schwinger oscillator model. This connection clarifies the physical meaning and the nature of the mathematical beauty of this model.

We also established the relationship between the $\{\Lambda, L'\}$ quantizer and the Nasyrov representation [38]. The latter formally allows one to reduce the quantum Liouville equation for free linear and symmetric tops to the form identical to the classical Liouville equation and propagate it using the familiar method of characteristics. In addition, we presented exact differential expressions for the key Bopp operators in this representation (Appendix E).

We hope that all the mentioned advantages will make the proposed representations useful for analysis of future experiments in quantum physics and quantum chemistry involving the complex semiclassical rotational dynamics of polyatomic molecules.

Finally, it is worth noting that the $\{\Lambda, L'\}$ quantizer cannot be derived within the standard Stratonovich-Weyl formalism. This fact indicates that the proposed generalized Wigner quantization method nontrivially extends the conventional procedure and may be useful to better balance the numerical utility and physical transparency. At the same time, the proposed method is semiphenomenological and lacks the mathematical rigor at the stage of the choice of the underlying set of postulates. Hence, additional research is needed to determine if it can be systematically applied to other nonstandard phase spaces.

APPENDIX A: FINDING THE IMAGES OF ANGULAR EIGENSTATES IN Σ_W^L FOR THE CASE $c_1 = -3$ IN (20)

Consider the orthogonality relation (3) for the set of Weyl symbols of operators $\hat{i}_l$:

$$\left(\widehat{\hat{i}}_{l_1}, \widehat{\hat{i}}_{l_2}\right)_W \propto \delta_{l_1, l_2}. \tag{A1}$$

The Weyl symbols $\widehat{\hat{i}}_l$ can only depend on the scalar argument $L = \sqrt{L_1^2 + L_2^2 + L_3^2}$ due to isotropy of the operators $\hat{i}_l$ and must be the solutions of the eigenvalue problem

$$\widehat{\hat{L}^2}\,\widehat{\hat{i}}_l(L) = \hbar^2 l(l+1)\widehat{\hat{i}}_l(L). \tag{A2}$$

The general solution of the fourth-order differential equation (A2) depends on four free parameters $c_{\alpha,\beta}$ ($\alpha, \beta = \pm 1$):

$$\widehat{\hat{i}}_l(L) = \sum_{\alpha,\beta=\pm 1} c_{\alpha,\beta} \frac{e^{-4L/\hbar}}{(L/\hbar)^{1-\xi}} \mathcal{L}^{(2\alpha\xi)}_{(2l+1)\beta - \alpha\xi - 1/2}\left[\frac{8L}{\hbar}\right], \tag{A3}$$

where $\mathcal{L}^{(j)}_i$ denotes the associated Laguerre polynomial. The particular solution of interest satisfies the conditions (A1) and $\widehat{\hat{i}}_l(L)|_{L\to\infty} \to 0$:

$$\widehat{\hat{i}}_l(L) = \sqrt{\frac{(2l+1)2^{6\xi+1}}{\Gamma(2l-\xi+3/2)\Gamma(2l+\xi+3/2)}} \frac{e^{-4L/\hbar}}{\sqrt{\pi}\hbar^{3/2}}\left(\frac{L}{\hbar}\right)^{\xi-1}$$
$$\times \Bigg(U(-2l+\xi-1/2, 2\xi+1, 8L/\hbar)$$
$$- \frac{\Gamma(2l+\xi+3/2)}{\Gamma(-2l+\xi-1/2)} U(2l+\xi+3/2, 2\xi+1, 8L/\hbar)\Bigg), \tag{A4}$$

where the $U(a,b,z)$ are the confluent hypergeometric functions of the second kind and $l > -\frac{1}{2}$. Equation (A4) allows us to find the basis functions $\widehat{\rho}_{l,l;l,l}$:

$$\widehat{\rho}_{l,l;l,l} = \frac{1}{[\hbar^{2l}(2l)!]^2}(\widetilde{L}_1 - i\widetilde{L}_2)^{2l}(\widetilde{L}_1 + i\widetilde{L}_2)^{2l}\widehat{\hat{i}}_l. \tag{A5}$$

In the special case of $\xi = 1/2$ only one of $c_{\alpha,\beta}$ is nonzero:

$$\widehat{\hat{i}}_l(L)|_{\xi=1/2} = \frac{4}{\hbar\sqrt{\pi L}}\frac{(-1)^{2l}e^{-4L/\hbar}\mathcal{L}^{(1)}_{2l}(8L/\hbar)}{\sqrt{L/\hbar}},$$
$$l = 0, \frac{1}{2}, 1, \frac{3}{2}, \ldots. \tag{A6}$$

Using (A6) and the relation

$$e^{-\gamma x} = \sum_{i=0}^{\infty} \frac{\gamma^i}{(1+\gamma)^{i+\alpha+1}} \mathcal{L}^{(\alpha)}_i(x), \tag{A7}$$

one can show that $\widehat{\hat{i}}|_{\xi=1/2} = \frac{1}{\hbar\sqrt{\pi L}}$.

APPENDIX B: CLASSICAL AND QUANTUM DESCRIPTION OF THE RIGID BODY DYNAMICS IN TERMS OF QUATERNIONS

For the sake of completeness of the presentation, in this appendix we review the key formulas of the quaternion algebra and briefly outline the standard representation of the rotational

motion in terms of quaternions (33) (for further details see, e.g., [53]). For clarity, we use bold symbols $\boldsymbol{x} = (x_0, x_1, x_2, x_3)$ for quaternion parameters and the symbol $*$ to denote the standard quaternion product

$$(\boldsymbol{y} * \boldsymbol{x})_k = \sum_{i,j=0}^{3} m^Q_{k,i,j} y_i x_j, \tag{B1}$$

where the coefficients $m^Q_{k,i,j}$ are defined by Eq. (35). From the physical point of view, the product (B1) represents the result of two successive rotations $\boldsymbol{x}$ and $\boldsymbol{y}$. For this reason, the product (B1) is not commutative. If the norm $\|\boldsymbol{x}\| = \sqrt{\sum_{k=0}^{3} x_i^2}$ of quaternion is not equal to one then each rotation $\boldsymbol{x}$ is also accompanied by uniform scaling by the factor $\|\boldsymbol{x}\|$. The transformation $\boldsymbol{x}^{-1}$ reciprocal to $\boldsymbol{x}$ [i.e., one that restores the initial geometry $\boldsymbol{x}\boldsymbol{x}^{-1} = \boldsymbol{x}^{-1}\boldsymbol{x} = \boldsymbol{1}$, where $\boldsymbol{1} = (1,0,0,0)$] is given by $\boldsymbol{x}^{-1} = \frac{\boldsymbol{x}^*}{\|\boldsymbol{x}\|^2}$, where the quaternion $\boldsymbol{x}^*$ is the conjugate of $\boldsymbol{x}$ defined as $\boldsymbol{x}^* = (x_0, -x_1, -x_2, -x_3)$. The components ω_k of angular frequency in this notation read

$$\omega_k(\boldsymbol{\lambda}, \dot{\boldsymbol{\lambda}}) = 2(\boldsymbol{\lambda}^* * \dot{\boldsymbol{\lambda}})_k, \tag{B2}$$

where $\dot{\boldsymbol{\lambda}} = \frac{d\boldsymbol{\lambda}}{dt}$. Equation (B2) allows us to determine the generalized momenta $p_{\lambda,k}$ canonically conjugate to λ_k:

$$\boldsymbol{p}_\lambda = \frac{\partial \mathrm{Lg}}{\partial \dot{\boldsymbol{\lambda}}} = \boldsymbol{\lambda} * \tilde{\boldsymbol{L}}, \tag{B3}$$

where Lg is the classical Lagrangian of the rigid rotor $\mathrm{Lg} = \frac{1}{2}\sum_{k=1}^{3} I_k \omega_k^2(\boldsymbol{\lambda}, \frac{d\boldsymbol{\lambda}}{dt})$ and $\tilde{\boldsymbol{L}} = (0, L_1, L_2, L_3)$. The canonical expressions for components L_k and L'_k of angular momenta relative to the moving and laboratory frames can be determined by applying to (B3) a reciprocal transform and expressions (36) for the direction cosines:

$$L_k = \frac{1}{2}(\boldsymbol{\lambda}^* * \boldsymbol{p}_\lambda)_k, \quad L'_k = -\frac{1}{2}(\boldsymbol{\lambda} * \boldsymbol{p}_\lambda^*)_k. \tag{B4}$$

The quaternions allow one to eliminate the singularities inherent to integration of the dynamical equations in terms of Euler angles. This makes them convenient for a variety of the scientific, engineering, technical, and graphics applications [56] including molecular-dynamics simulations [57,58].

It can be shown [35,36] that the passage to the Schrödinger quantum description of rotations can be done in the ordinary way by replacing the $p_{\lambda,k}$ with $-i\hbar\frac{\partial}{\partial\lambda_k}$ in Eq. (B4). Strictly speaking, the variables λ_k in this picture represent the angular quaternions up to scaling factors. For this reason, one has to explicitly enforce the correct normalization in the potential part of the Hamiltonian by replacing the λ_k with $\hat{\lambda}_k = \frac{\lambda_k}{\|\boldsymbol{\lambda}\|}$. The associated phase-space representation can be trivially obtained using the original Wigner recipe or via the substitutions (6) and (8) [35,36].

It is worth mentioning that although the operators $\hat{\lambda}_k$ are Hermitian, they cannot be associated with the quantum-mechanical observable since they include the matrix elements corresponding to fractional changes of angular momentum quantum number l that have never been observed in experiments. The fundamental reason for that is that the corresponding set of operators can be introduced only in an overcomplete configuration space.

APPENDIX C: DERIVATION OF EQS. (48) and (49)

It follows from the definition of the quantum-mechanical angular momentum operators that

$$\exp\left(-\frac{2}{\hbar}\Phi\vec{L}'_{\mathrm{im},k}\right)\bigg|_{\hbar\to 0}\hat{\rho} = \mathscr{R}_k(\Phi)\hat{\rho}, \tag{C1}$$

where $\vec{L}'_{\mathrm{im},k} = -i\frac{1}{2}(\hat{L}'_k - \hat{L}'_k)$ and $\mathscr{R}_k(\Phi)$ is the classical operator of rotation about the axis $\vec{e}'_k$ by the angle Φ, i.e., for $\mathscr{R}_k(\Phi)$,

$$L'_s \to \cos[(1-\delta_{k,s})\Phi]L'_s + \sum_{n=1}^{3}\epsilon_{s,k,n}\sin(\Phi)L'_n,$$

$$\lambda_s \to \sum_{m,n=0}^{3} m^Q_{s,m,n} r_{k,m}\lambda_n, \tag{C2}$$

$$\mathscr{q}_s \to \sum_{m,n=0}^{3} m^Q_{s,m,n} r_{k,m}\mathscr{q}_n,$$

where the quaternions $r_{k,m} = \delta_{0,m}\cos\frac{\Phi}{2} + \delta_{k,m}\sin\frac{\Phi}{2}$ generate rotations about each axis $\vec{e}'_k$ [see Eq. (33)]. Equations (C1) and (C2) allow us to determine expressions for the imaginary parts of $\hat{L}'_k$ in different phase spaces $\Sigma_W^{\Lambda,L'}$, $\Sigma_W^{\Lambda,\mathscr{q}}$, and $\Sigma_W^{\lambda,\mathscr{q}}$:

$$\vec{L}'_{\mathrm{im},k} = \begin{cases} -\frac{\hbar}{4}\left(\sum_{m,n=0}^{3} m^Q_{n,k,m}\Lambda_m\frac{\partial}{\partial\Lambda_n} \right. \\ \left. \quad + 2\sum_{m,n=1}^{3} m^Q_{n,k,m}L'_m\frac{\partial}{\partial L'_n}\right), \quad \Sigma_W^{\Lambda,L'} \\ \frac{\hbar}{4}\left(\vec{\mu}_k - \sum_{m,n=0}^{3} m^Q_{n,k,m}\Lambda_m\frac{\partial}{\partial\Lambda_n}\right), \quad \Sigma_W^{\Lambda,\mathscr{q}} \\ \frac{\hbar}{4}\vec{\mu}_k, \quad\quad\quad\quad\quad\quad\quad\quad\quad \Sigma_W^{\lambda,\mathscr{q}}, \end{cases} \tag{C3}$$

where $\vec{\mu}_k = -\sum_{m,n=0}^{3} m^Q_{n,k,m}\mathscr{q}_m\frac{\partial}{\partial\mathscr{q}_n}$. Here the transformation (43) was applied to obtain the last line in Eq. (C3). The latter relation together with Eqs. (47) and (15) and commutation relations identical to (46) specifies the possible forms of the ladder operators $\hat{b}_\pm$. The two simplest solutions are given by the operators (48) and

$$\hat{b}_\pm = i\mathscr{q}_{2\pm1} - \mathscr{q}_{1\mp1}, \quad \hat{b}^\dagger_\pm = \frac{1}{2}\left(\frac{\partial}{\partial\mathscr{q}_{1\mp1}} + i\frac{\partial}{\partial\mathscr{q}_{2\pm1}}\right) \tag{C4}$$

[the latter choice leads to the transpose of (49) with unbounded right eigenstates and hence should be rejected]. The specific choice of constant prefactors in Eq. (48) is made with the goal to simplify the expressions for quaternion operators (see Appendix D). Applying the transformation (43) to (47) and (48), one obtains the following formula for the angular momentum operators in the space $\Sigma_W^{\Lambda,\mathscr{q}}$:

$$\hat{L}'_k = \frac{L'_k(\mathscr{q})}{L'(\mathscr{q})}[\{\vec{\ell}'_{\mathrm{re}}(\mathscr{q}) - \vec{\ell}_{\mathrm{re}}(\Lambda)\} + \vec{\ell}_{\mathrm{re}}(\Lambda)]$$

$$+ \frac{\hbar}{4}\sum_{r=0}^{3}\sum_{s=1}^{3} m^Q_{k,r,s}\left((1-\delta_{r,0})\frac{L'_r(\mathscr{q})}{L'(\mathscr{q})} + i\delta_{r,0}\right)$$

$$\times\left(\vec{\mu}_s - \sum_{m,n=0}^{3} m^Q_{n,s,m}\Lambda_m\frac{\partial}{\partial\Lambda_n}\right), \tag{C5}$$

where $L'_n(\boldsymbol{q})$ is defined by the first of Eqs. (43) and the operators $\overleftrightarrow{\widetilde{\ell}}_{\mathrm{re}}(\boldsymbol{\Lambda}) = \frac{\hbar}{8}[\sum_{n=0}^{3}(\Lambda_n^2 - \frac{\partial^2}{\partial \Lambda_n^2}) - 4]$ and $\overleftrightarrow{\widetilde{\ell}}'_{\mathrm{re}}(\boldsymbol{q}) = \frac{\hbar}{4}\sum_{n=0}^{3}\boldsymbol{q}_n\frac{\partial}{\partial \boldsymbol{q}_n}$ are the real parts of the operators $\overleftrightarrow{\widetilde{\ell}}'(\boldsymbol{q})$ and $\overleftrightarrow{\widetilde{\ell}}(\boldsymbol{\Lambda})$ such that

$$\widetilde{\ell}'(\boldsymbol{q})[\widetilde{\ell}\,'(\boldsymbol{q}) + \hbar] = \widehat{L}'^2, \quad \widetilde{\ell}(\boldsymbol{\Lambda})[\widetilde{\ell}(\boldsymbol{\Lambda}) + \hbar] = \widehat{L}^2. \quad \text{(C6)}$$

Thus, the operators $\widetilde{\ell}'(\boldsymbol{q})$ and $\widetilde{\ell}(\boldsymbol{\Lambda})$ are physically equivalent (i.e., they must produce the same action when applied to any valid physical state $\widehat{\rho}$) and are mathematically distinct only due to redundant dimensionality of the phase spaces $\Sigma_{\mathrm{W}}^{\widetilde{\lambda},\boldsymbol{q}}$ and $\Sigma_{\mathrm{W}}^{\Lambda,\boldsymbol{q}}$. This fact allows one to omit the term in the curly brackets in Eq. (C5). Together with Eqs. (43) and (C6) it leads to the following set of correspondence relations between phase spaces $\Sigma_{\mathrm{W}}^{\Lambda,\boldsymbol{q}}$ and $\Sigma_{\mathrm{W}}^{\Lambda,L}$:

$$L'_n(\boldsymbol{q}) \leftrightarrow L'_n, \quad \vec{\mu}_k \leftrightarrow -2\sum_{m,n=1}^{3} m_{n,k,m}^{\mathrm{Q}} L'_m \frac{\partial}{\partial L'_n},$$

$$\overleftrightarrow{\ell}'_{\mathrm{re}}(\boldsymbol{q}) - \overleftrightarrow{\ell}_{\mathrm{re}}(\boldsymbol{\Lambda}) \leftrightarrow 0. \quad \text{(C7)}$$

Their substitution into Eq. (C5) leads to Eq. (49). One can directly check that Eqs. (49) and (42) are consistent with the condition $\sum_{k=1}^{3}(\widehat{L}_k'^2 - \widehat{L}_k^2) = 0$.

APPENDIX D: EXPLICIT EXPRESSIONS FOR THE WIGNER IMAGES OF THE LADDER AND QUATERNION OPERATORS IN $\Sigma_{\mathrm{W}}^{\Lambda,L}$ PHASE SPACE

The aim of this Appendix is to complete the construction of the quantum algebra of the phase space $\Sigma_{\mathrm{W}}^{\Lambda,L'}$ by finding the images of the quaternion operators $\widehat{\lambda}_k$. For convenience of readers who are not interested in the technical details of the derivation we start by providing the final result.

Consider the $\Sigma_{\mathrm{W}}^{\Lambda,L'}$ Wigner representation with the normalization (56) in Eq. (53), the scalar product (57), and the images of components of angular momentum defined by Eqs. (42) and (49). The corresponding images of the quaternion operators $\widehat{\lambda}_k$ are given by Eq. (D12), where the compound operators $\widetilde{a_\xi b}_\chi$ and $\widetilde{a_\xi^\dagger b}_\chi^\dagger$ ($\xi, \chi = \pm 1$) are specified by Eqs. (D5)–(D8).

To prove this result, we note that the images $\widehat{\lambda}_k$ can be readily defined in terms of the ladder operators (45) and (48) (see below). However, the ladder operators $\widetilde{a}_\pm^\dagger, \widetilde{a}_\pm$ and $\widetilde{b}_\pm^\dagger, \widetilde{b}_\pm$ themselves can exist only in the overcomplete phase space $\{\widetilde{\lambda}, \boldsymbol{q}\}$ since their separate application leads to unphysical states with mismatched values of the total angular momentum measured in the laboratory and moving frames. Nevertheless, the phase space $\Sigma_{\mathrm{W}}^{\Lambda,L'}$ can host the compound operators $\widetilde{a_\xi b}_\chi$ and $\widetilde{a_\xi^\dagger b}_\chi^\dagger$, which are free of this problem.

The explicit expressions for the compound operators can be found by applying replacements and substitutions (43) and (C7) to their counterparts $e^{i(\pi/4)}\widehat{a}_\xi\widetilde{b}_\chi$ and $e^{-i(\pi/4)}\widehat{a}_\xi^\dagger\widetilde{b}_\chi^\dagger$ in the $\{\widetilde{\lambda}, \boldsymbol{q}\}$ representation [see Eqs. (45) and (48)]. The result is the following operators $\widetilde{g}_{\xi,\chi}^-$ and $\widetilde{g}_{\xi,\chi}^+$:

$$\widetilde{g}_{\xi,\chi}^+ = e^{-i(\pi/4)}\frac{1}{2\sqrt{2\hbar L'}}\left\{ (\delta_{\chi,-\xi} + \chi\delta_{\chi,\xi})\left(\frac{\xi+\chi}{2}(\xi L' + L_3') + \frac{\xi-\chi}{2}(L_1' - i\xi L_2')\right)\left[\Lambda_3 - \frac{\partial}{\partial\Lambda_3} - i\xi\left(\Lambda_0 - \frac{\partial}{\partial\Lambda_0}\right)\right] \right.$$

$$\left. + \left(\frac{\chi-\xi}{2}(L_3' - \xi L') + \frac{\xi\chi+1}{2}(L_1' + i\xi L_2')\right)\times\left[\Lambda_1 - \frac{\partial}{\partial\Lambda_1} - i\xi\left(\Lambda_2 - \frac{\partial}{\partial\Lambda_2}\right)\right] \right\}, \quad \text{(D1)}$$

$$\widetilde{g}_{\xi,\chi}^- = -e^{i(\pi/4)}\sqrt{\hbar}\frac{(\chi\delta_{\xi,\chi} + \delta_{\xi,-\chi})}{8\sqrt{2}(L')^{3/2}}\left(\sum_{n=+1,-1}\frac{(L_3' - \xi n L')(\xi\chi - n) + (n\xi\chi + 1)(L_1' - in\xi L_2')}{2}\right.$$

$$\times\left[n\left(\Lambda_{2-n}\frac{\partial}{\partial\Lambda_{n+1}} - \Lambda_{n+1}\frac{\partial}{\partial\Lambda_{2-n}}\right) - i\chi + 2iL'\frac{\partial}{\partial L_3'}\right]\left[\Lambda_{n+1} + \frac{\partial}{\partial\Lambda_{n+1}} - i\xi\left(\Lambda_{2-n} + \frac{\partial}{\partial\Lambda_{2-n}}\right)\right]$$

$$+ \sum_{n=0}^{3}i^{\xi n}\left\{\sum_{k=0}^{3}\sum_{s=1}^{3}\sum_{j=0}^{3}\left[L_s'\left(\frac{\xi+\chi(1-(3-n)n)}{2}\right)^2\{1 - [1-(3-s)s][2(\delta_{j,3-n} + \delta_{k,3-n}) - 1]\} + \xi i^{(1-s\chi)}f_{\xi,\chi,n}\right.\right.$$

$$\left.\times(1-\delta_{3,s})(\delta_{j,n} + \delta_{k,n})\right]m_{k,s,j}^{\mathrm{Q}}\Lambda_j\frac{\partial}{\partial\Lambda_k} + 2L'\xi f_{\xi,\chi,n}\left(\frac{\partial}{\partial L_2'} + i\chi\frac{\partial}{\partial L_1'}\right)$$

$$\left.\left. + 4iL'\left(\frac{\xi+\chi(1-(3-n)n)}{2}\right)^2\right\}\left(\Lambda_n + \frac{\partial}{\partial\Lambda_n}\right)\right), \quad \text{(D2)}$$

where

$$f_{\xi,\chi,n} = \tfrac{1}{2}(\{L'\xi[1-(3-n)n] + L'_3\}\{1-\xi\chi[1-(3-n)n]\}$$
$$+ \{\xi\chi + [1-(3-n)n]\}\{L'_1 + iL'_2\xi[1-(3-n)n]\}) \quad (D3)$$

(the phase factors $e^{\pm i(\pi/4)}$ are included for consistency with the generally accepted normalization of the rotational eigenstates $|l,m,k\rangle$). The simplest way to study the effect of the operators (D1) and (D2) on the Weyl symbols is to apply them to the isotropic states (55) and (56):

$$\sum_{\xi,\chi=-1,1} \widehat{g}^{+}_{\xi,\chi}\widehat{g}^{+}_{\xi,\chi}\widehat{\mathfrak{i}}_l = \mu_l(L')4\left(l+\frac{1}{2}\right)^2\widehat{\mathfrak{i}}_{l+1/2}, \quad (D4)$$

where $\mu_l(L') = \frac{1}{2(l+1/2)+1}\frac{2L'}{\hbar}$ is an additional factor compared to the expected effect of the compound ladder operator. The correct form of the compound operator can be found by applying to (D1) and (D2) the following transformation, which eliminates this factor:

$$a_\xi\widehat{b}_\chi = \mathcal{S}_0\widehat{g}^{-}_{\xi,\chi}\mathcal{S}_0^{-1} = \left(\sqrt{\frac{2L'}{\hbar}}\widehat{g}^{-}_{\xi,\chi} + \delta\widehat{g}^{-}_{\xi,\chi}\right)\sqrt{\frac{1}{2\widehat{\ell}/\hbar+1}}, \quad (D5)$$

$$a_\xi^\dagger\widehat{b}_\chi^\dagger = \mathcal{S}_0\widehat{g}^{+}_{\xi,\chi}\mathcal{S}_0^{-1} = \sqrt{\frac{2\widehat{\ell}}{\hbar}+1}\sqrt{\frac{\hbar}{2L'}}\widehat{g}^{+}_{\xi,\chi}, \quad (D6)$$

where $\mathcal{S}_0$ is the invariance operator [see Eq. (52)]

$$\mathcal{S}_0 = \sqrt{\Gamma\left(\frac{2\widehat{\ell}}{\hbar}+2\right)\Gamma\left(\frac{2\widehat{\ell}}{\hbar}+2\right)}\left(\frac{2L'}{\hbar}\right)^{-(1/\hbar)(\widehat{\ell}+\widehat{\ell})} \quad (D7)$$

[here $\Gamma(z)$ is the Euler Gamma function] and

$$\delta\widehat{g}^{-}_{\xi,\chi} = \sum_{n=0}^{1}\frac{(L'+\chi_3)\delta_{\xi,-(-1)^n\chi} + (L_1-i\chi L_2)\delta_{\xi,(-1)^n\chi}}{i^{-n}e^{-i\pi/4}4L'}$$
$$\times\left[\Lambda_{1-n} + \frac{\partial}{\partial\Lambda_{1-n}} + i\xi(-1)^n\left(\Lambda_{n+2} + \frac{\partial}{\partial\Lambda_{n+2}}\right)\right]$$
$$\times\frac{\widehat{\ell}+\widehat{\ell}}{\hbar}. \quad (D8)$$

To deduce the Wigner images $\widehat{\lambda}_k$ of the quaternion operators we apply the known relations

$$\hat{\lambda}_n = \frac{(-i)^n}{2}D^{1/2}_{1/2,(1-(3-n)n)/2} + \frac{e^{i(\pi/4)[(-1)^n-1]}}{2}D^{1/2}_{-1/2,((3-n)n-1)/2}; \quad (D9)$$

$$D^{1/2}_{\delta m,\delta k}|l,m,k\rangle$$
$$= \sum_{j=l-1/2}^{l+1/2}i^{l-j}\sqrt{(2l+1)(2j+1)}$$
$$\times(-1)^{\delta m+m-(\delta k+k)}\begin{pmatrix} j & \frac{1}{2} & l \\ -(\delta k+k) & \delta k & k \end{pmatrix}$$
$$\times\begin{pmatrix} j & \frac{1}{2} & l \\ -(\delta m+m) & \delta m & m \end{pmatrix}|j,m+\delta m,k+\delta k\rangle, \quad (D10)$$

where $D^l_{m,k}$ are Wigner D functions. Since

$$\left|l+\frac{1}{2},m+\frac{\mu}{2},k+\frac{\kappa}{2}\right\rangle = \frac{\hat{a}_\kappa^\dagger\hat{b}_\mu^\dagger}{\sqrt{l+\kappa k+1}\sqrt{l+\mu m+1}}|l,m,k\rangle,$$
$$\left|l-\frac{1}{2},m+\frac{\mu}{2},k+\frac{\kappa}{2}\right\rangle = \frac{\hat{a}_{-\kappa}\hat{b}_{-\mu}}{\sqrt{l-\kappa k}\sqrt{l-\mu m}}|l,m,k\rangle \quad (D11)$$

($\kappa,\mu=\pm 1$), the action of the operators (D9) can be represented as a bilinear combination of the ladder operators $\hat{\lambda}|l,m,k\rangle = \sum_{\mu,\kappa}[\hat{a}_\kappa^\dagger\hat{b}_\mu^\dagger c_1(l,m,k) + \hat{a}_\kappa\hat{b}_\mu c_2(l,m,k)]|l,m,k\rangle$. Since the operators $\hat{L}^2$, $\hat{L}_3$, and $\hat{L}'_3$ commute, the coefficients $c_n(l,m,k)$ may be replaced by the operators $\hat{c}_n = c_n(\frac{\hat{L}}{\hbar},\frac{\hat{L}'_3}{\hbar},\frac{\hat{L}_3}{\hbar})$. After converting the resulting operator into the Wigner representation using Eqs. (D5), (D6), and (51), one finally gets

$$\widehat{\lambda}_n = \frac{1}{2\sqrt{\widehat{\ell}/\hbar+1/2}}[e^{i\pi/4}(-i)^n a_{-1}^\dagger\widehat{b}^\dagger_{(3-n)n-1}$$
$$+ e^{(i\pi/4)(-1)^n}a_1^\dagger\widehat{b}^\dagger_{1-(3-n)n} - e^{-(i\pi/4)(-1)^n}a_1\widehat{b}_{1-(3-n)n}$$
$$- e^{-i\pi/4}i^n a_{-1}\widehat{b}_{(3-n)n-1}]\frac{1}{2\sqrt{\widehat{\ell}/\hbar+1/2}}. \quad (D12)$$

One can readily check that the requested consistency of classical limits (C̄:2) holds: $\lim_{\hbar\to 0}\widehat{\lambda}_m = \lambda_m$. Thus, the Wigner $\{\boldsymbol{\Lambda},\boldsymbol{L}\}$ representation defined by Eqs. (42), (49), (57), (D5), (D6), and (D12) is self-consistent and complete.

APPENDIX E: NASYROV-TYPE PHASE REPRESENTATION OF THE ROTATIONAL MOTION

The goal of this Appendix is to derive a Wigner representation in which the quantum generator of motion (64) for the symmetric top coincides with the corresponding classical Liouville operator $\mathscr{L}$. We will depart from the $\{\lambda,\boldsymbol{q}\}$ representation and convert it into the desired form via a series of transformations. The procedure (and the final expressions for $\widehat{\ell}$ and $\widehat{L}_k$) in the case of variables $\{\boldsymbol{\Lambda},\boldsymbol{L}'\}$ remains the same. However, in this case the expressions of the Bopp operators $\widehat{L}'_k$ and $\widehat{\lambda}_m$ are rather cumbersome due to the complicated form of relations $\Lambda_m(\lambda,\boldsymbol{L}')$ and will not be presented here.

We start with the fractional Laplace transform of the variables λ_m:

$$\hat{\rho}(\boldsymbol{r},\boldsymbol{L}) = \iiiint_{-\infty}^{\infty} \left(\prod_{m=0}^{3} d\lambda_m\right)$$
$$\times \exp\left[\sum_{m=0}^{3}\left(\sqrt{2}\lambda_m r_m - \frac{\lambda_m^2}{2} - \frac{r_m^2}{2}\right)\right]\hat{\rho}(\lambda,\boldsymbol{L}). \tag{E1}$$

The operators in the spaces $\Sigma_W^{r,\boldsymbol{\varphi}}$ and $\Sigma_W^{\lambda,\boldsymbol{\varphi}}$ are connected via the correspondence:

$$\lambda_m \to \frac{r_m + \partial/\partial r_m}{\sqrt{2}}, \quad \frac{\partial}{\partial\lambda_m} \to \frac{\partial/\partial r_m - r_m}{\sqrt{2}}. \tag{E2}$$

Thus, the effect of r_m and $\frac{d}{dr_m}$ on λ_m is identical to the effect of the ladder operators on the canonical coordinate. Treating the variables r_m as proportional to the components of a quaternion, we can formally express them in terms of the associated Euler angles α and γ:

$$r_0 = R_1\cos\left(\frac{\alpha+\gamma}{2}\right), \quad r_1 = R_2\cos\left(\frac{\alpha-\gamma}{2}\right),$$
$$r_2 = R_2\sin\left(\frac{\alpha-\gamma}{2}\right), \quad r_3 = R_1\sin\left(\frac{\alpha+\gamma}{2}\right). \tag{E3}$$

The quantum Liouville operator $\hat{\mathscr{L}}$ for the symmetric top in the variables $\{R_1,R_2,\alpha,\beta,L_1,L_1,L_3\}$ takes the form (64) with

$$\vec{\mathfrak{J}} = \frac{\hat{\ell}+\hat{\ell}+\hbar}{2} = \frac{1}{4}\hbar\left(R_1\frac{\partial}{\partial R_1}+R_2\frac{\partial}{\partial R_2}+2\right),$$
$$\vec{\mathfrak{K}} = \frac{\hat{L}_3+\hat{L}_3}{2} = \frac{1}{4}\hbar\left(R_1\frac{\partial}{\partial R_1}-R_2\frac{\partial}{\partial R_2}\right). \tag{E4}$$

These expressions allow us to further trace the analogy of R_i and $\frac{\partial}{\partial R_i}$ with the ladder operators and consider $\vec{\mathfrak{J}}$ and $\vec{\mathfrak{K}}$ as $\pm$ combinations of two harmonic-oscillator Hamiltonians. Guided by this analogy, we make the operator substitution

$$\frac{\partial}{\partial R_j} \to e^{\partial/\partial s_j}\sqrt{s_j}, \quad R_j \to \sqrt{s_j}e^{-\partial/\partial s_j} \quad (j=1,2), \tag{E5}$$

which preserves the commutation relation $[\frac{\partial}{\partial R_i},R_j]=\delta_{i,j}$. Finally, we introduce the variables

$$J = \frac{\hbar}{4}(s_1+s_2+2), \quad K = \frac{\hbar}{4}(s_1-s_2). \tag{E6}$$

The parameters $\{J,K,\alpha,\beta,\boldsymbol{\varphi}\}$ constitute the required set of variables in which the Bopp operators of the quantum Liouville

operator $\hat{\mathscr{L}}$, the components $\hat{L}_k$, the ladder operators (45), and the operator $\hat{\ell}$ defined by (50) take the form

$$\hat{\mathscr{L}} = -AJ\frac{\partial}{\partial\alpha}+(A-B)K\frac{\partial}{\partial\gamma}, \tag{E7}$$

$$\hat{L}_1 \pm i\hat{L}_2 = e^{\mp i\gamma}\sqrt{\frac{J\mp K-\hbar/2}{J\pm K-\hbar/2}}$$
$$\times\frac{\hbar(\partial/\partial\gamma\pm\partial/\partial\alpha)\pm 2i(J\pm K)}{2}e^{\pm(\hbar/2)(\partial/\partial K)}, \tag{E8}$$

$$\hat{L}_3 = K - \frac{1}{2}i\hbar\frac{\partial}{\partial\gamma}, \tag{E9}$$

$$\hat{\ell} = J - \frac{\hbar}{2} - i\frac{\hbar}{2}\frac{\partial}{\partial\alpha}, \tag{E10}$$

$$\hat{a}_{\pm} = \frac{e^{i(\alpha\pm\gamma)/2}\{\partial/\partial\gamma\pm\partial/\partial\alpha-i[2(J\pm K)/\hbar]\}}{e^{i(\pi/4)(\pm 1-1)}\sqrt{2(J\pm K)/\hbar-1}}$$
$$\times e^{(\hbar/4)(\partial/\partial J\pm\partial/\partial K)}, \tag{E11}$$

$$\hat{a}_{\pm}^{\dagger} = \frac{e^{-i(\gamma\pm\alpha)/2}}{e^{-i\pi(\pm 1+1)/4}}\sqrt{2(J\pm K)/\hbar-1}\,e^{-(\hbar/4)(\partial/\partial J\pm\partial/\partial K)}, \tag{E12}$$

whereas the operators $\hat{b}_{\pm}$ and $\hat{b}_{\pm}^{\dagger}$ are still defined by Eq. (48). It is readily verified that Eqs. (E7)–(E10) have the correct classical limits [recall that the ladder operators (E11), (E12), and (48) are specified up to the invariance transform (52)].

One can see that the dynamic master equation (E7) exactly coincides with its classical analog. Similarly, the expressions for $\hat{\ell}$ and $\hat{K}$ resemble the canonical Bopp operators (6) and, in particular, obey the relations

$$(\hat{\ell}^n,\rho)_W = ((J-\hbar/2)^n,\rho)_W, \quad (\hat{L}_3,\rho)_W = (K^n,\rho)_W, \tag{E13}$$

so that the associated marginal distributions for the Wigner functions represent the probability distributions for quantities ℓ and K [cf. Eq. (9) and the subsequent discussion].

The action of operators $e^{\pm(\hbar/4)(\partial/\partial K)}$ and $e^{\pm(\hbar/4)(\partial/\partial J)}$ contained in Eqs. (E8), (E11), and (E12) on any function of variables J and K consist of discrete replacements $J\to J\pm\frac{\hbar}{4}$ and $K\to K\pm\frac{\hbar}{4}$. For this reason, the parameters J and K take a discrete set of values so that the Wigner images $\hat{\rho}_{1,2}$ of the basis functions $\hat{\rho}_{1,2}=|l_1,m_1,k_1\rangle\langle l_2,m_3,k_2|$ read

$$\hat{\rho}_{1,2} \propto \delta_{2J/\hbar,l_1+l_2+1}\delta_{2K/\hbar,k_1+k_2} \tag{E14}$$

(for more details about the explicit form and properties of such a semidiscrete Wigner functions see [38]).

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