Wigner representation of the rotational dynamics of rigid tops

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We propose the general methodology to design the Wigner representations with the desired dynamical and semiclassical properties in the phase spaces with nontrivial topology. As an illustration, two representations of molecular rotations are developed to suit the computational demands of contemporary applications of laser alignment, diagnostics of reaction dynamics, studies of scattering, and dissipative processes.

I. INTRODUCTION

The dynamics of complex quantum systems on the border between classical and quantum mechanics is of high interest for various applications in quantum optics and information, structural analysis, studies of matter waves and mechanisms of chemical reactions (see e.g. [1, 2]). Advances in the quantum state preparation and transient probing make it possible to track the details of such a dynamics with up to attosecond resolution [3, 4]. On the theory side, the approximate models are needed for the computationally feasible treatment of this dynamical regime. Most of them are based on the semiclassical propagation of the Wigner function [10–13] including phase integral approaches [14] and the huge family of initial value representations and related techniques [15, 16].

Conceptually, these approximations improve the numerical efficacy by optimizing the choice of the leading terms in the perturbation series expansions for the generators of motion [16]. This strategy, however, overlooks an additional opportunity to improve the properties of the exact phase space representations themselves [17–19] owing to an essential ambiguity and variability in their definition and construction (especially in the case of nontrivial topology) [4, 20]. The analysis of this resource with the specific application to rotational motion of extended bodies (the practically important problem for which a little was done on the theoretical side in semiclassical context [21, 22]) constitutes the subject of the present paper.

A variety of ways to extend the original Wigner’s proposal to the case of rotational dynamics were suggested and analyzed [20, 23], but only few of them are applicable to unrestricted rotations of 3-dimensional bodies. The early solutions [34, 35] reduced the problem to the canonical case at the cost of extending the phase space by two fake dimensions. A variant suggested in [37] allows to directly extract the most useful marginal distributions but involves rather complicated quantum Liouville equations. Conversely, in the Nasyrov’s proposal [38] these equations for free symmetric and linear tops coincide with the classical ones at the expense of very complicated integro-differential form for dynamical equations and common observables in general case. Similar drawbacks also limit the utility of the schemes [39] based on the direct extension of the original Stratonovich-Weyl correspondence for spin [40].

In all of these extensions the certain properties of the canonical Wigner prototype have to be sacrificed. Except for the Nasyrov’s scheme [38], the obligatory preserved properties are chosen to be the static restrictions imposed on the quasiprobability distribution (in agreement with the original Wigner’s reasoning [10, 11]). In this paper we construct new forms of the Wigner representation departing from dynamic postulates. This strategy (detailed in Sec. II) allows to directly outline the desired skeleton of the evolution equations. In the subsequent sections III, IV we develop several variants of Wigner representation of rotational motion originating from classical Euler equations and Liouville equation written in terms of components of angular momenta and quaternion parameters. We encourage readers to check the concluding section V for the brief summary of the key features and the expected advantages of the proposed schemes in the numerical simulations.

II. THE FUNDAMENTALS OF GENERALIZED WIGNER REPRESENTATION

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) $\Sigma$, the algebra $\mathcal{B}$ of these events and the probability measure $\mathcal{P}$ for any subset of $\Sigma$ [42]. Fortunately, the Hilbert space framework is fully compatible with both classical and quantum-mechanical objects [43, 44]. The Wigner representation exploits this fundamental fact. It is constructed by equipping the classical phase space $\Sigma$ by such additional scalar product $\langle \cdot, \cdot \rangle_W$ that the resulted Hilbert space $\Sigma_W$ is capable to simultaneously host both classical and quantum algebras. This change formally converts both classical and quantum quantities into an operators acting in $\Sigma_W$. To distinguish between them we will mark the latter by symbol $\tilde{\cdot}$ while preserving the “hat” notation $\hat{\cdot}$ for operators in the ordinary configuration Hilbert space.

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

$$
(F) = (F, \hat{\rho})_W = \int \ldots \int F \hat{\rho} d\Omega,
$$

(1)

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

$$
(\hat{F}) = (\hat{F}, \hat{\rho})_W.
$$

(2)

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

$$(\hat{\rho}_1, \hat{\rho}_2)_W = C \text{Tr}[\hat{\rho}_1 \hat{\rho}_2], \quad C = \text{const.}
$$

(3)

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

$$
\hat{F} = \hat{F}^\dagger.
$$

(4)

Eqs. (3), (4) imply that

$$
\hat{\rho} = \hat{\rho}^\dagger = \hat{\rho}^*.
$$

(5)

is the real-valued symmetric function of phase variables.

With this, the explicit form of images $\hat{x}_i$ and $\hat{p}_i$ of the quantum coordinate and momentum operators (known as Bopp operators [11, 45]) is uniquely defined by 1) fundamental property of Galilean invariance of non-relativistic phase space $\Sigma_W$ (which prescribes $\hat{x}_i$ and $\hat{p}_i$ to be linear in both $p_i$ and $q_i$ and $\frac{\partial}{\partial p_i}$, $\frac{\partial}{\partial q_i}$); 2) canonical commutation relation $[\hat{x}_i, \hat{p}_j] = i\hbar$ and 3) requirement of the proper classical limit $\hat{p}_i|_{\hbar \to 0} = p_i$, $\hat{q}_i|_{\hbar \to 0} = q_i$:

$$
\hat{x}_i = x_i + \frac{i\hbar}{2} \frac{\partial}{\partial p_i}; \quad \hat{p}_i = p_i - \frac{i\hbar}{2} \frac{\partial}{\partial x_i}.
$$

(6)

Note that these operators being applied to $\hat{\rho}$ produce an image of the left multiplications $\hat{p}_i \hat{\rho}$ and $\hat{q}_i \hat{\rho}$. It is convenient to introduce the operators $\hat{p}_i$ and $\hat{q}_i$ whose effect on $\hat{\rho}$ is associated with the right multiplications. The associativity relations of form $\forall \hat{\rho} : [\hat{q}_i, \hat{\rho}]_\hbar = (\hat{q}_i \hat{\rho} - \hat{\rho} \hat{q}_i)_{\hbar}$ and the equality $[\hat{q}_i, \hat{p}_j] = -i\hbar \delta_{ij}$ imply that the right operators should satisfy the commutation relations:

$$
[\hat{p}_i, \hat{p}_j] = [\hat{p}_i, \hat{q}_j] = [\hat{q}_i, \hat{p}_j] = [\hat{q}_i, \hat{q}_j] = 0 \quad \text{and} \quad [\hat{q}_i, \hat{p}_j] = -i\hbar \delta_{ij}.
$$

(7)

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

$$
\hat{p}_i = x_i - \frac{i\hbar}{2} \frac{\partial}{\partial p_i} = \hat{p}_i^*; \quad \hat{q}_i = p_i + \frac{i\hbar}{2} \frac{\partial}{\partial x_i} = \hat{q}_i^*.
$$

(8)

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

$$
(\hat{q}_i^n, \hat{\rho})_W = \frac{1}{2^n} \langle \hat{q}_i^n, \hat{\rho} \rangle W = (q_i^n, \hat{\rho})_W;
$$

$$
(\hat{p}_i^n, \hat{\rho})_W = (p_i^n, \hat{\rho})_W.
$$

(9)

In particular, eqs. (9) mean that the partial integration in the right-hand side of (1) over coordinates (momenta) with $\hbar = 1$ returns the correct marginal probability distributions for values of momenta (coordinates).

Eqs. (6) and (7) completely specify the quantum algebra and establish one-to-one correspondence between arbitrary quantum operator $\hat{F} = \hat{F}(\hat{\rho}, \hat{\phi})$, its Wigner image $\hat{F} = F(\hat{\rho}, \hat{\phi})$ and Weyl symbol $F_W(p, q) = F(\hat{p}, \hat{q})$ (see [11, 12] for details), as well as define the image of master equation $\frac{\partial}{\partial t} \hat{F} = -\frac{i}{\hbar} \hat{H} \hat{F}$ with Hamiltonian $\hat{H} = H(\hat{p}, \hat{q})$:

$$
\frac{\partial}{\partial t} \hat{\rho} = \hat{\mathcal{L}} \hat{\rho},
$$

(10)

where the quantum Liouvillean $\hat{\mathcal{L}}$ is given by real operator

$$
\hat{\mathcal{L}} = -\frac{i}{\hbar} (H(\hat{p}, \hat{q}) - H(\hat{\rho}, \hat{\phi})).
$$

(11)

However, in general case of non-canonic phase spaces, the eqs. (6), (10) are not self-consistent, so that some of them must be relaxed, e.g.:

(I) one can postulate the desired “static” properties of the quasiprobability distribution like (2), (3), (7) 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), (5), (7)).

The approach (I) is rigorously axiomatized [41]; its abstracted generalization in group-theoretical terms (known as 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] axiomatic basis for the approach (II) postulates the equations of motion for averages of certain physical quantities [33, 49]. Another possible starting point is the Feynman’s path integral representations of the time evolution [12].

The fundamental origin of this diversity of possible definitions is the wide freedom in choosing either the Weyl symbols of density matrices $\hat{\rho}$ or quantum observables $\hat{F}$ to be a main “carriers of nonclassicality” (see [4, 50] for details). In this paper we demonstrate that this feature allows to balance the complexities of evolution equations and the associated Wigner functions.
III. QUANTIZATION OF EULER EQUATIONS

Discovered in 1765, the Euler’s celebrated equations

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

(12)

(here \( \epsilon_{i,j,k} \) is Levi-Civita symbol, \( I_k \) are momenta of inertia for principal axes \( \hat{e}_k \) of the rigid body and \( L_k \) are the projections of the angular moment on \( \hat{e}_k \)) comprehensively describe the free dynamics of rigid bodies in moving frame \( S \) in terms of phase space \( \Sigma = \{ L_1, L_2, L_3 \} \). We will require a quantum extension of eqs. (12) to obey:

(E:1) the enforced condition \( \tilde{\rho} \) of reality of the quasiprobability distributions \( \tilde{\rho} = \tilde{\rho}(L_1, L_2, L_3) \); any real-valued Weyl symbol \( \tilde{\rho} \) should correspond to Hermitian (not necessarily positive) matrix \( \tilde{\rho} \);

(E:2) the orthogonality relation \( \tilde{\rho} \) in \( \Sigma_W \) where \( d\Sigma = dL_1dL_2dL_3 \) and \( C=1 \);

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

\[ \tilde{L}_i|_{h \to 0} = L_i; \quad \tilde{\rho}|_{h \to 0} = \rho = \sum_{i,j,k=1}^{3} \epsilon_{i,j,k} \left( \frac{L_j L_k}{I_k} \right) \frac{\partial}{\partial L_i}. \]  

(13)

These postulates match the Wigner’s definition (11) except for replacing the static condition (9) on the marginal distributions with the dynamical restriction (E:3). The postulates (E:2) and (E:3) still lead to the Hermiticity condition (14) (i.e. the image of each observable should be the symmetrized composition of \( L_i \) and \( \frac{\partial}{\partial \hat{L}_i} \)). The images of the associativity and commutation relations \( \forall \rho : L_k(\rho L_i) = (L_k \rho) L_i; \quad [\tilde{L}_i, \tilde{L}_k] = i\hbar \sum_{m=1}^{3} \epsilon_{i,k,m} \tilde{L}_m \) read as:

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

(14)

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

\[ \tilde{F} = \tilde{F}^* \quad (\text{cf. with} \; (11) \; \text{and} \; (8)) \]  

(15)

Relation (15) allows to define: \( \tilde{L}_k = \tilde{L}_{re,k} + i\tilde{L}_{im,k} \), \( \tilde{L}_k = \tilde{L}_{re,k} - i\tilde{L}_{im,k} \) and rewrite eqs. (14) in the equivalent form:

\[ \forall k, l : [\tilde{L}_{re,k}, \tilde{L}_{im,l}] = 0; \]  

(16a)

\[ [\tilde{L}_{im,i}, \tilde{L}_{im,k}] = [\tilde{L}_{re,k}, \tilde{L}_{im,l}] = \frac{\hbar}{2} \sum_{m=1}^{3} \epsilon_{k,l,m} \tilde{L}_{im,m}; \]  

(16b)

\[ [\tilde{L}_{re,k}, \tilde{L}_{im,l}] = \frac{\hbar}{2} \sum_{m=1}^{3} \epsilon_{k,l,m} \tilde{L}_{re,m}. \]  

(16c)

Using eq. (11) and the familiar expression

\[ \hat{H} = \sum_{k=1}^{3} \frac{\tilde{L}_k^2}{2I_k} \]  

(17)

for eigen Hamiltonian of free rigid top one obtains:

\[ \tilde{g}^2 = 2 \sum_{k=1}^{3} \tilde{L}_{re,k} \tilde{L}_{im,k} \]  

(18)

Applying (E:3) to (18) leads to the expressions for \( \tilde{L}_{im,k} \):

\[ \tilde{L}_{im,k} = \frac{1}{\hbar} \sum_{i,j=1}^{3} \epsilon_{i,j,k} \frac{\partial}{\partial L_i} \]  

(19)

up to optional terms of order \( O(\hbar^2) \) omitted here for reasons of simplicity. Eqs. (19) and (14) further give:

\[ \tilde{L}_{re,k} = L_k + \frac{\hbar}{16} \left( -2 \sum_{i=1}^{3} L_i \frac{\partial}{\partial L_i} \frac{\partial}{\partial L_k} + \sum_{i=1}^{3} \frac{\partial^2}{\partial L_i^2} + c_1 \frac{\partial}{\partial L_k} + \frac{\epsilon^2 L_1}{L_i^2} \right), \]  

(20)

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

\[ \langle \hat{F} \rangle = \hat{i}(\hat{F}, \hat{\rho})_W, \quad (\text{cf. eq.} \; (2)) \]  

(21)

where \( \hat{i} \) is the Weyl symbol of identity matrix: \( \hat{i} = \sum_{k=1}^{3} \hat{i} \). Here \( \hat{i} = \sum_{k=1}^{3} L_k |l,k \rangle \langle l,k| \) where the basis states \( |l,k \rangle \) satisfy the relations \( \hat{L}_k \hat{L}_k = \hbar (l+1) |l,k \rangle \), \( \hat{L}_3 |l,k \rangle = h k |l,k \rangle \). It is shown in Appendix A that the convenient non-singular variant \( \hat{i} = \sum_{k=1}^{3} \hat{i} \) corresponds to \( \xi = \frac{1}{2} \). For this case eqs. (A5), (A6) from Appendix A give:

\[ |l, l \rangle \langle l, l| = \frac{4}{\sqrt{\pi L}} \left( \frac{-1}{2} \right)^{2l} e^{-\frac{\sqrt{\hbar(L+\frac{L_3}{2})}}{\hbar}} \]  

(22)

\[ |l, k_1 \rangle \langle l, k_2| = \frac{k_1 - k_2}{L_1 - iL_2} \left( e^{-\frac{\sqrt{\hbar(L+\frac{L_3}{2})}}{\hbar}} \right) \]  

(23)

where the notation \( L_{2l} \) stands for Laguerre polynomials.

Let us highlight some peculiarities of the obtained representation.

1. In the angular momentum case the eqs. (2) and (3) can not be simultaneously satisfied because of nonuniform density of quantum states in \( \Sigma_W \): \( i \neq 1 \). However, one may set \( c_1 = -4, \xi = 0 \) to satisfy (22) instead of (9) which is equivalent to non-unitary transformation:

\[ \tilde{\rho} = \pi^{-\frac{L}{2}} L^\eta \tilde{\rho} \xi = \frac{1}{2}, c_1 = -3; \quad \tilde{F}^n = L^n \tilde{F} |\xi = \frac{1}{2}, c_1 = -3, L^{-n} \]  

(24)
with $\eta=-\frac{i}{\hbar}$. Remarkably, the mathematical structure of Wigner images for $\hat{L}_k$ given by eqs. \ref{24} is similar (up to the complex conjugation) to the generalized Bopp operators for spin [12, 5]. The general solution valid for arbitrary $c_1, \xi$ is given in Appendix A.

2. It follows from eq. \ref{10} that $\frac{1}{2}(\hat{L}_i+\hat{L}_j)=\hat{I}_{te,i}\neq \hat{L}_i$ regardless of the particular choice of the averaging and normalization. This precludes the analogs of relations \ref{9} for angular components $L_k$, so one no longer can get the meaningful marginal distributions via partial integration over $\hat{p}$. With this, the semiclassical dynamical properties of the Wigner representation remain preserved. Specifically, one still can directly relate the Wigner equations of motion for pure states \ref{10} $\hat{L}$ (with $c_1=-3, \xi=1/2$) to classical equations of motion both in standard and Koopman von Neumann form using the recipe from [48].

3. The truncated Euler phase space $\Sigma^L_{df}$ can not handle any information about the absolute orientation relative to the laboratory frame $S'$. In particular, we can not define the $e_3^z$-projection of angular moment and the associated quantum number $m$. In addition, it is easy to verify the relation $\hat{L}^2=\hat{L}^2$, and so $[\hat{L}^2, \hat{p}]=\langle \hat{L}^2, \hat{p}\rangle=0$ for any feasible density matrix $\hat{\rho}$. This fact precludes any quantum coherences between the eigenstates $|l, k\rangle$ and $|l', k'\rangle$ with $l \neq l'$. Note that such a coherence would violate the postulate (E:1) of one-to-one correspondence since they would lead to many-to-one relation:

$$\forall \alpha : \alpha \hat{L}^2\hat{\rho}+(1-\alpha)\hat{L}^2 \leftrightarrow \hat{L}^2 \hat{\rho}. \quad (25)$$

The equality $\hat{L}^2=\hat{L}^2$ also leads to zeroing of both classical and quantum Liouvillians of the free spherical tops: $\dot{\Omega}=\dot{\Omega}=0$.

4. The consequence of the ambiguity relation \ref{25} is the existence of the real, bounded, isotropic and square-integrable in $\Sigma^W$ solutions $\xi_l$ of the equation $\hat{L}^2 \xi_l=\hbar l(l+1) \xi_l$ for any real $l>\frac{1}{2}$ (see Appendix A, eq. \ref{41}). The solution $\xi_l$ of this equation is integrable over the phase space variables for any value of $l$. However, the coefficients $\kappa_{l,j} \xi_l=\sum_{j=0}^{\infty} \kappa_{l,j} \xi_j$ take negative values for non-integer values of $2l$. Moreover, in general case $\kappa_{l,j} \xi_{2l+j} \rightarrow \infty$ $\frac{(-1)^j}{j!} \neq 0$. For this reason, the Weyl symbols $\xi_l$ can not be normalized to represent a valid physical state. These properties should be considered with caution in calculations because they indicate that a small numerical error can result in dramatic physical mistake.

IV. COMPLETE DESCRIPTION OF THE ROTATIONAL MOTION

The rotational master equations reduce to the most elegant form in terms of four quaternions $\lambda_k$ defined as:

$$\lambda_0 = \cos \frac{\Phi}{2}, \quad \lambda_k = \eta_k \sin \frac{\Phi}{2} \quad (k = 1, 2, 3), \quad \text{(26)}$$

where the parameters $\eta$ and $\Phi$ are such that the rotation around 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 of work [17]). This makes the construction of the Wigner representation in terms of $\lambda_0$ and the associated canonically conjugated momenta $p_{\lambda,k}$ straightforward since the associated Bopp operators obey the canonical commutation relations identical to \ref{9}, \ref{51}, \ref{52}, \ref{53}, \ref{54} (see Appendix B for details and brief review of the algebra of quaternions). However, extra dimensionality 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 non-canonical phase space $\Sigma^L_{W}$ composed of $\lambda_k$ and the projections $L'_k$ of the angular momentum on the laboratory axes. The corresponding classical Liouvillian reads:

$$\mathcal{L}=\frac{1}{2} \sum_{k=1}^{3} \sum_{m,n=0}^{3} m^Q_{m,n,k} \omega_k \lambda_m \frac{\partial}{\partial \lambda_n}, \quad \text{(27)}$$

where $m^Q_{m,n,k}$ are quaternion multiplication coefficients:

$$m^Q_{k,i,j} = \begin{cases} \epsilon_{i,j,k} & \text{if } i>0 \wedge j>0 \wedge k>0, \\ \delta_{j,0} \delta_{k,i} + \delta_{i,0} \delta_{k,j} - \delta_{k,0} \delta_{i,j} & \text{otherwise,} \end{cases} \quad \text{(28)}$$

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

$$q_{i,j,m,n}=(1-2 \delta_{j,m}) \sum_{k=0}^{3} m^Q_{i,j,k} m^Q_{k,m,n}. \quad \text{(29)}$$

To quantize the eqs. \ref{26}, \ref{27} one can apply the postulates similar to (E:1) with $d\Omega=\prod_{i=1}^{3} dL_i \prod_{j=0}^{3} d\lambda_j$:

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

(C:2) the traciality relation [3]:

Figure 1. The physical meaning of the parameters entering the definition \ref{52} of quaternions $\lambda_k$. 
the proper classical limits:
\[ \hat{L}_1|\hbar \to 0 = L_1; \quad \hat{L}_2|\hbar \to 0 = L_2; \quad \hat{\lambda}_j|\hbar \to 0 = \lambda_j, \]
\[ \text{(C:3)} \]

\[ \frac{2}{i\hbar}[\hat{L}_i, \hat{\lambda}_j] = \sum_{k=1}^3 m_{k,i,j}^Q \hat{\lambda}_k, \]
\[ \text{(31)} \]

Repeating the steps leading to eqs. (19), (20) one gets:
\[ \hat{\lambda}_k = \frac{1}{2} \left[ \sum_{s=0}^3 q_{k,s,m,n} \frac{\partial}{\partial L_s^m} - i\frac{\hbar}{4} \sum_{m,n,k}^Q m_{k,i,j}^Q \frac{\partial}{\partial L_n^m} \right] \lambda_m \frac{\partial}{\partial \lambda_n}, \]
\[ \lambda_k = \hat{N}^{-\frac{1}{2}} \left( \lambda_k + i\frac{\hbar}{4} \sum_{m,n,k}^Q m_{k,m,n} \lambda_m \frac{\partial}{\partial L_n^m} \right), \]
\[ \hat{N} = \left( \sum_{s=0}^3 \lambda_s^2 \right)^{-\frac{1}{2}} \left( 1 - \frac{\hbar^2}{16} \sum_{s=1}^Q \frac{\partial^2}{\partial L_s^m} \right), \]
\[ \text{(32)} \]

\[ \text{(33)} \]

\[ \text{(34)} \]

where the operator \( \hat{N} \) commutes with all physical observables of form \( F(L_k, \hat{\lambda}_n) \). The existence of such \( \hat{N} \neq 1 \) is due to overcompleteness of our 7D phase space \( \Sigma_{W,L}^q \). The Wigner representation (32)-(34) is not convenient for exact numerical implementation because of the term \( \hat{N}^{-\frac{1}{2}} \) in the expression (33) for quaternion images which is the differential operator of infinite order. This pre-factor, however, can be ignored when choosing to work only with images of states satisfying equation \( \hat{N}\hat{\rho} = \hat{\rho} \) (which is possible owing to its commutation properties). Another possibility is to Fourier or Laplace transform the phase space \( \Sigma_{W,L}^q \) with respect to \( L_1^m, L_2^m \) and \( L_3^m \). However, the resulted equations will loose all key characteristic properties of the Wigner representation. More importantly, in any case one can not get rid of the residual extra dimensionality of \( \Sigma_{W,L}^q \). Another serious drawback is that unlike the classical generator of free motion (27) the quantum counterpart (11) no longer evidently manifest the angular momentum conservation via preserving the values of \( L_1^m \). One would desire to retain this remarkable property of eq. (27) in the quantum case because it would allow reduction of the 7-dimensional differential propagation equation to the series of 4-dimensional ones.

To resolve these issues, let us relax the axioms to the:
(C:1) the reality condition (15);
(C:2) the requirement (30) of the proper classical limits;
(C:3) the requirement of the absence of derivatives over \( L_1^m, L_2^m \) and \( L_3^m \) in the expressions for \( \hat{L}_k \).

The expressions for \( \hat{L}_k \) matching these axioms can be compactly written in the new variables \( \Lambda_m = \sqrt{L_1^m} \sqrt{L_2^m} \lambda_m \):
\[ \hat{L}_k = \frac{\hbar}{8} \sum_{m,n=0}^3 \left( \sum_{s=1}^3 q_{s,m,n} L_s^m L_s^n \left( \Lambda_m \Lambda_n - \frac{\partial^2}{\partial \Lambda_m \partial \Lambda_n} \right) \right) + 2i m^Q_{m,n,k} \Lambda_m \frac{\partial}{\partial \Lambda_n}, \]
\[ \text{(35)} \]

where \( L' = \sqrt{\sum_{m=1}^Q L_m^2} \). It is useful to introduce the intermediate fixed frame \( S' \) which third axis \( e_3' \) coincides with the (conserved) direction of the angular moment. Let us denote as \( \mathbf{q} \) the quaternion which represents the rotation connecting \( S' \) and \( S'' \) and introduce the parameters \( \gamma_m \) as the exact analogs of parameters \( \Lambda_m \) characterizing the orientation of the rotor relative to \( S'' \):
\[ L_n = \sum_{m,n=0}^3 q_{m,j,n} q_{j,m} \lambda_j; \quad \Lambda_k = \sum_{i,j=0}^3 m_{k,i,j}^Q q_{i,j} \hat{\lambda}_j, \]
\[ \text{(36)} \]

(Not to eqs. (36) do not fix the directions of the axes \( e_3' \) or \( e_3'' \) of \( S'' \), and so do not uniquely define \( q_{k} \).
Eqs. (36) take simple form in new variables \( \gamma_m \):
\[ \hat{L}_1 \pm \hat{L}_2 = \hbar \hat{a}_+ \hat{a}_-; \quad \hat{L}_3 = \frac{\hbar}{2} \hat{a}_+ \hat{a}_- \hat{a}_+, \]
\[ \text{(37)} \]

where \( \hat{a}_+ \) and \( \hat{a}_- \) are the conventional ladder operators:
\[ \hat{a}_+ \hat{a}_- + \hat{a}_- \hat{a}_+ = \gamma_{2\pm 1} \pm i \frac{\partial}{\partial \gamma_{1\pm 1}}; \quad \hat{a}_- \hat{a}_+ = \frac{\partial}{\partial \gamma_{0\pm 1}} \pm i \gamma_{1\pm 1}, \]
\[ \text{(38)} \]

(It is worth to stress that the notations like \( \hat{a}_\pm \) hereafter mean "the Wigner image of creation operator \( \hat{a}_\dagger \)", not the "Hermitian conjugate of the Bopp operator \( \hat{a}_\pm ^* \)."

The fact that \( \hat{a}_\dagger \) and \( \hat{a}_\pm \) do not depend on \( q_\alpha \) and the commutation relation \( \forall m,n : [\hat{L}_m, \hat{L}_n] = 0 \) hints that the images \( \hat{L}_m \) should have the form similar to (37):
\[ \hat{L}_1 \pm i \hat{L}_2 = \hbar \hat{b}_+ \hat{b}_-; \quad \hat{L}_3 = \frac{\hbar}{2} (\hat{b}_+ \hat{b}_- - \hat{b}_- \hat{b}_+), \]
\[ \text{(40)} \]

where the operators \( \hat{b}_\dagger \) and \( \hat{b}_\pm \) do not depend on \( \gamma_k \) and satisfy the commutation relations identical to (39). The validity of eqs. (11) is proven in Appendix C where the following explicit expressions are obtained (up to invariance transformation (41), see below):
\[ \hat{b}_+ = \sqrt{2} (q_{1\pm 1} + i q_{2\pm 1}); \quad \hat{b}_\pm = \frac{1}{\sqrt{2}} (\frac{\partial}{\partial q_{1\pm 1}} - i \frac{\partial}{\partial q_{2\pm 1}}), \]
\[ \text{(41)} \]

\[ \hat{L}_k = \frac{L'_k}{L'} \left( \frac{\hbar}{4} \sum_{r=0}^3 \sum_{s=1}^3 q_{k,r,s} (1 - \delta_{s,0}) \frac{L'_r}{L'} \hat{b}_+ \hat{b}_- + i \delta_{s,0} \right) \times \]
\[ \sum_{m,n=0}^3 m_{m,n,k}^Q \left( 2(1 - \delta_{m,n,0}) L'_m \frac{\partial}{\partial L'_n} + \Lambda_m \frac{\partial}{\partial \Lambda_n} \right), \]
\[ \text{(42)} \]
where the operator \( \hat{\ell} \) of quantum number \( l \) and its image are defined as:

\[
\hat{\ell} (\ell + \hbar) = \hat{L}^2;
\]

\[
\hat{L} = \hbar \sum_{m=0}^{3} (\lambda_n^2 - \frac{\partial^2}{\partial \lambda_n^2}) \frac{m Q}{L} \frac{\partial}{\partial \lambda_n}.
\]

(43)

The classical analogy can be pushed even further by forcing the operators \( \hat{L}_k \) and \( \hat{L}_3 \) to take the mathematical structure of canonical Bopp operators \( \hat{L}_3 \) and \( \hat{L}_3 \) with discrete spectra. That is, the values of axial and precession frequencies of the quantum top can only take on 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 famous phenomenon of quantum rotational revivals.

The only difference between the quantum Liouvillian (45) and its classical counterpart is that the classical variables \( L' \) and \( L_3' \) are replaced with the quantum Bopp operators \( \hat{L}_3 \) and \( \hat{L}_3 \) with discrete spectra. That is, the values of axial and precession frequencies of the quantum top can only take on 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 famous phenomenon of quantum rotational revivals.

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laser excitation etc. On the conceptual side, it establishes the bridge between the formal quantization of the spin degrees of freedom and classical Euler equations. We also have seen that in fact there exist a large family of such a quantizations (parametrized by $c_1$ and $\xi$) from which one can choose one with the preferable forms of equations for the calculation of averages and the normalization of quasiprobability distributions.

The important property of the complete phase space description proposed in Sec. IV is that it explicitly converts the angular momentum conservation laws into conservation of values of parameters $L_1^c$, $L_2^c$, $L_3^c$ in the course of free rotations. This feature allows for natural parallelization of the code via splitting the initial 6-dimensional problem into series of independent 4-dimensional ones for evolution of the parameters $L_m$ ($m=0, \ldots, 3$). Several other representations (e.g. [37, 38]) also allow for the similar splitting, but their parametric spaces are not singularity-free. In addition, the generator of free motion in our case is the low-order differential operator of continuous arguments (the same is also true for the Euler quantizer of Sec. III). This should greatly facilitate the relatively inexpensive exact propagation of the evolution equations and is the important prerequisite for effective application of the initial value approximations.

On the conceptual level, our findings uncover the direct connection (eq. [37]) between the quaternion parameters and the raising and lowering operators entering the Schwinger oscillator model which clarifies the physical meaning and the reasons underlying the exciting mathematical beauty of the latter.

We also established the relationship between the proposed representation and Nasyrov-type representation [28]. It formally allows to reduce the quantum Liouville equation for linear and symmetric tops to the form identical to the classical Liouville equation and propagate it using the familiar method of characteristics. In addition, to the best of our knowledge, we for the first time outlined the exact differential expressions for the key Bopp operators in this representation (Appendix IV).

We hope that the all mentioned advantages will make the proposed representations useful for analysis of the emerging experiments in quantum physics and quantum chemistry involving the complex semiclassical rotational dynamics of polyatomic molecules. We also believe that our critical revision of the axiomatic approach to the formal definition of the Wigner function from the dynamical perspective will be useful for constructing the Wigner representations for other dynamical systems with non-trivial structures of the underlying phase spaces.

**ACKNOWLEDGMENTS**

D. Z. is very grateful to Dr. Denys I. Bondar and Dr. Renan Cabrera for numerous stimulating discussions, important comments and valuable suggestions.

**Appendix A: Finding the images of angular eigenstates in $\Sigma_W$**

Let us consider the orthogonality relation (3) for the set of Weyl symbols of operators $i_l$:

$$\langle i_{l_1}, i_{l_2}\rangle_W = \delta_{l_1, l_2}, \quad (A1)$$

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

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

The general solution of the fourth-order differential equation (A2) depends on 4 free parameters $c_{\alpha,\beta}$ ($\alpha, \beta = \pm 1$):

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

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

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

where $U(a, b, z)$ stands for confluent hypergeometric function of the 2-nd kind and $l > -\frac{1}{2}$. Eq. (A4) allows to find the Weyl symbols for matrix elements $|l, l\rangle (l, l)$:

$$|l, l\rangle (l, l) = \frac{1}{(2l+1)!2l!} (\hat{L}_1 - i\hat{L}_2)^l (\hat{L}_1 + i\hat{L}_2)^l \hat{i}_l. \quad (A5)$$

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

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

Using (A6) and the well-known relation:

$$e^{-\gamma x} = \sum_{i=0}^{\infty} \frac{\gamma^i}{(1+\gamma)^{i+1}} L_i^{(1)}(x), \quad (A7)$$

one can show that $\hat{i}_l(\xi = \frac{1}{2}) = \frac{1}{\sqrt{\pi L}}$. 

Appendix B: Classical and quantum description of the rigid body dynamics in quaternions

For the sake of completeness of the presentation, in this Appendix we remind the key formulas of the quaternion algebra and briefly outline the standard representation of the rotational motion in quaternions \(20\) (for further details see e.g. \(52\)). For clarity, we will use the bold symbols \(\mathbf{x} = (x_0, x_1, x_2, x_3)\) for quaternion parameters and symbol \(\ast\) to denote the standard quaternion product:

\[
(y \ast x)_k = \sum_{i,j=0}^{3} m^Q_{k,i,j} y_i x_j, \tag{B1}
\]

where the coefficients \(m^Q_{k,i,j}\) are defined by eq. \(28\). From the physical point of view, the product \(B1\) represents the result of two successive rotations \(x\) and \(y\). For this reason, the product \(B1\) is not commutative. If the norm \(||x|| = \sqrt{\sum_{k=0}^{3} x_k^2}\) of quaternion is not equal to one then each rotation \(x\) is also accompanied by uniform scaling by factor \(|x||\). The transformation \(x^{-1}\) reciprocal to \(x\) (i.e. one which restores the initial geometry: \(x x^{-1} = x^* x = 1\), where \(1 = (1, 0, 0, 0)\)) is given by formula:

\[
x^{-1} = \frac{x^*}{(||x||)^2}, \tag{B2}
\]

where \(x^* = (x_0, -x_1, x_2, -x_3)\). The components \(\omega_k\) of angular frequency in these notations read as:

\[
\omega_k(\lambda, \bar{\lambda}) = 2(\lambda^* \dot{\lambda})_k, \tag{B3}
\]

where \(\dot{\lambda} = \frac{d\lambda}{dt}\). Eq. \(B2\) allows to determine the generalized momenta \(p_{\lambda,k}\) canonically conjugated to \(\lambda_k\):

\[
p_{\lambda} = \frac{\partial L_{\lambda}}{\partial \dot{\lambda}} = \lambda^* \dot{\lambda}, \tag{B3}
\]

where \(L_{\lambda}\) is the classical Lagrangian of rigid rotor: \(L_{\lambda} = \frac{1}{2} \sum_{k=1}^{3} \omega_k^2(\lambda, \frac{d\lambda}{dt})\) and \(L = (L_0, L_1, L_2, L_3)\). The canonical expressions for components \(L_k\) and \(L^\prime_k\) of angular momenta relative to moving and laboratory frames can be determined by applying to \(B3\) the reciprocal transform and the expressions \(29\) for direction cosines:

\[
L_k = \frac{1}{2}(\lambda^* \dot{p}_{\lambda})_k; \quad L^\prime_k = -\frac{1}{2}(\lambda \dot{p}_{\lambda}^\prime)_k. \tag{B4}
\]

The quaternions allow to eliminate the singularities inherent to integration of dynamical equations in Euler angles. This makes them convenient for variety of the scientific, engineering, technical and graphics applications \(53\) including the molecular dynamics simulations \(54\). It can be shown \(35, 36\) that the passage to the Schrodinger 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 \(B4\). Strictly speaking, the variables \(\lambda_k\) 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, expressions for direction cosines etc. by replacing \(\lambda_k\) with \(\hat{\lambda}_k = \frac{\lambda_k}{||\lambda||}\). The associated phase space representation can be trivially obtained using the original Wigner’s recipe or via substitutions \(5\) and \(8\) \(35, 36\).

It is worth to mention that despite the operators \(\lambda_k\) are Hermitian they can not be associated with any quantum-mechanical observable since they include the matrix elements corresponding to fractional changes of angular momentum quantum number \(l\) which are never observed in experiments. The fundamental reason for that is that the corresponding set of operators can be introduced only in overcomplete configuration space.

Appendix C: Derivation of eqs. \((41)\) and \((42)\)

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

\[
\exp\left(-\frac{2}{\hbar} \Phi \tilde{L}_{im,k}\right)_{\lambda \rightarrow 0} = \hat{R}_k(\Phi) \hat{\rho}, \tag{C1}
\]

where \(\tilde{L}_{im,k} = -\frac{1}{2}(\hat{L}_i - \hat{L}_j)\) and \(\hat{R}_k(\Phi)\) is the classical operator of the rotation about axis \(e_k\) on angle \(\Phi\), i.e.:

\[
\begin{align*}
L_s' &\rightarrow \cos((1-\delta_{k,s})\Phi)L_s' + \sum_{n=1}^{3} \delta_{s,k,n} \sin(\Phi)L_n' ; \\
\hat{R}_k(\Phi) &\rightarrow \begin{cases} 
\hat{L}_s \rightarrow \hat{L}_s + \sum_{m,n} m_{m,n} \hat{r}_{k,m} \hat{q}_{m,n} ; \\
\hat{q}_s \rightarrow \sum_{m,n} m_{m,n} \hat{r}_{k,m} \hat{q}_{m,n} .
\end{cases}
\end{align*} \tag{C2}
\]

where the quaternions \(r_{k,m} = \delta_{0,m} \cos \frac{\Phi}{2} + \delta_{1,m} \sin \frac{\Phi}{2}\) generate rotations around each of axes \(e_k\) (see eq. \(26\)). Eqs. \(C1\) and \(C2\) can be resolved relative to \(\tilde{L}_{im,k}\) in different phase spaces (indicated on the right):

\[
\tilde{L}_{im,k} \rightarrow \begin{cases} 
\frac{-4}{\hbar^2} \sum_{m,n} m_{n,k,m} \hat{m}_{m,n} \partial_q q_{m,n} + \Sigma L_{l'} ; \\
\frac{2}{\hbar^2} \sum_{m,n} m_{n,k,m} \hat{l}'_{m,n} \partial_q q_{m,n} ; \\
\frac{2}{\hbar^2} \sum_{m,n} \hat{m}_{m,n} \partial_q q_{m,n} ; \\
\frac{2}{\hbar^2} \sum_{m,n} \hat{l}'_{m,n} \partial_q q_{m,n} ; \\
\end{cases} \tag{C3}
\]

where \(\hat{m}_{k} = -\sum_{m,n=0}^{3} m_{n,k,m} q_{m,n} \partial_q q_{m,n}\). Here the transformation \(36\) was used to obtain the last line in \(C3\). The latter relation together with eqs. \(41\), \(42\) and the commutation relations identical to \(39\) specify the possible forms of ladder operators \(\hat{b}_{\pm}\). The two simplest solutions are given by operators \(41\) and

\[
\hat{b}_{\pm} = i q_{2\pm 1} - q_{1\pm 1}; \quad \hat{b}_{\pm}^\dagger = \frac{1}{2} \left( \frac{\partial}{\partial q_{1\pm 1}} + i \frac{\partial}{\partial q_{2\pm 1}} \right). \tag{C4}
\]

(the latter choice leads to transpose of \(42\) with unbounded right eigenstates and, thus, should be rejected). The specific choice of constant prefactors in \(41\) is made with aim to simplify the expressions for quaternion operators (see Appendix \(3\)). Applying the transformation
Thus, the operators \( \hat{L}'(\mathbf{q}) \) and \( \hat{\ell}(\mathbf{A}) \) are physically equivalent (i.e., they must produce the same action being applied to any valid physical state \( \rho \)) and are mathematically distinct only due to redundant dimensionality of the phase spaces \( \Sigma_W^{\alpha,q} \) and \( \Sigma_W^{\lambda,q} \). This fact allows to cancel the term in the curly brackets in (C5). Together with eqs. (C6) and \( \hat{L}'_{\text{re}}(\mathbf{q}) \), it leads to the following set of correspondence relations between phase spaces \( \Sigma_W^{\alpha,q} \) and \( \Sigma_W^{\lambda,L} \):

\[
L'_n(\mathbf{q}) \leftrightarrow L'_n; \quad \hat{\mu}_k \leftrightarrow -2 \sum_{m,n=1}^3 m_{n,k,m} L'_m \frac{\partial}{\partial L_n}; \quad \hat{L}'_{\text{re}}(\mathbf{q}) - \hat{L}_{\text{re}}(\mathbf{A}) \leftrightarrow 0.
\]

Their substitution into eq. (C5) leads to eq. (D1). One can directly check that the eqs. (D2) and (D5) are consistent with the condition \( \sum_{k=1}^{\ell}(\hat{L}'_k - \hat{L}_k) = 0 \).

Appendix D: Proof of the orthogonality relation (46)

By definition, the symbols \( \tilde{\rho}_{\alpha,\beta} \) are the eigenstates of operators \( \hat{L}_1, \hat{L}_3, \hat{L}_2, \hat{L}'_1, \hat{L}'_2, \hat{L}'_3 \) and \( \frac{1}{2}(\hat{L}'_1 - \hat{L}'_3) \) given by eqs. (30) and (42) and \( \tilde{\rho}_{\alpha,\beta} = \tilde{\rho}_{\alpha,\beta} \). This is violated. Thus, to prove the orthogonality of projectors it remains to justify eq. (46) in the case when all of the equalities (42) are satisfied but \( m_{\alpha_1} + m_{\beta_2} \neq m_{\alpha_2} + m_{\beta_2} \).

In order to simplify the notations, let us fix the arbitrary values of quantum numbers \( l \) and \( k \) and denote as \( \tilde{\rho}_{m_1,m_2} \) the Weyl symbol of projector \( [l, m_1, k] \{l, m_2 \} \).

The Weyl symbols \( \tilde{\rho}_{m_1,m_2} \) of projector \( \{l, m_1 \} \{l, m_2 \} \) for the subspaces with well-defined quantum number \( l \) are the isotropic solutions of the eigenvalue problem \( \hat{\ell}_1 \tilde{\rho}_{m_1,m_2} \) with \( \tilde{\rho}_{m_1,m_2} \) given by (44):

\[
\hat{\ell}_1 \tilde{\rho}_{m_1,m_2} = \sum_{m=-\ell}^{\ell} \Lambda_m \tilde{\rho}_{m_1,m_2} \quad \text{with} \quad \sum_{m=0}^\infty \Lambda_m^2 = 0.
\]

where the variable prefactor \( \tilde{\rho}_{m_1,m_2} \) depends on the choice of \( S \) in the invariance relation (45). The explicit expressions for any symbol \( \tilde{\rho}_{m_1,m_2} \) can be found by application of the operators \( \tilde{\rho}_{m_1,m_2} \) and their right counterparts to (D3). The result has the following general form:

\[
\tilde{\rho}_{m_1,m_2} = \frac{1}{2^{l+3}} e^{-\frac{3}{8}l(l+1)} \tilde{\rho}_{m_1,m_2} \quad \text{with} \quad \sum_{m=0}^\infty \Lambda_m^2 = 0.
\]

Appendix E: The explicit expressions for Wigner images of the ladder and quaternion operators in \( \Sigma_W^{\lambda,L} \) phase space

The results of Appendix D indicate that the orthogonality of two states \( \rho_{\alpha_1,\beta_1} \) and \( \rho_{\alpha_2,\beta_2} \) relative to the scalar product (40) is preserved under their multiplication on arbitrary bounded functions \( f_{l_1,l_2}(\mathbf{L}) \). The invariance relation (45) is the reflection of this property in the operator space. The specific form of the factors \( f_{l_1,l_2}(\mathbf{L}) \) is prescribed by eq. (44). More specifically, we require the Weyl symbol of any proper physical state to be of form \( \rho(\mathbf{L}, \hat{L}'_1, \hat{L}'_2, \hat{L}'_3) \) which effectively reduces the dimension of the phase space to six since the last 3 arguments are not independent. The goal of this Appendix is to find

\[

\to \quad \text{and (11)} \quad \text{one obtains the following formula for the angular momentum operators in the space} \quad \Sigma_W^{l,q}:

\[
\hat{L}'_k = \frac{L'_k(\mathbf{q})}{L'(\mathbf{q})} \left( \left( \hat{L}'_{\text{re,0}}(\mathbf{q}) - \hat{L}_{\text{re,0}}(\mathbf{A}) \right) + \hat{L}_{\text{re}}(\mathbf{A}) \right) + \frac{\hbar}{4} \sum_{r=1}^3 \sum_{s=1}^3 m_{r,k,s} L'_m \frac{\partial}{\partial L_n} ;
\]

\[
\hat{L}'_{\text{re}}(\mathbf{q}) - \hat{L}_{\text{re}}(\mathbf{A}) \equiv 0.
\]
the equivalents of the operators (38), (41) in the phase space $\Sigma^\Lambda L$ which are consistent with this choice.

The ladder operators $\hat{a}^\dagger_\pm, \hat{a}_\pm$ and $\hat{b}^\dagger_\pm, \hat{b}_\pm$ can be defined only in the overcomplete phase space since their separate application leads to the unphysical states with the mismatched values of the total angular momentum measured in the laboratory and moving frames. For this reason, we will further deal with the set of compound operators $\hat{a}^\dagger_\pm, \hat{b}^\dagger_\pm$ which is free of this problem and sufficient to describe any physical process.

We begin with application of the replacements and substitutions (36), (37) to the compound operators $e^{i\frac{\pi}{2} \hat{a}^\dagger_\pm \hat{b}^\dagger_\pm}$ in $\{\gamma, q\}$-representation (eqs. (38), (41)). The result is the following new operators $\hat{g}^+_\xi, \hat{g}^-_\xi$:

$$
\hat{g}^+_\xi \chi = e^{-i\frac{\pi}{2}} \frac{1}{2\sqrt{2}hL} \left( \left( \chi + \xi \delta_\xi \chi \right) \left( \xi + \frac{\chi}{2} (L' + L_2') \right) + \left( \frac{\chi - \xi}{2} (L_3' - \xi L') \right) \right) \left( \Lambda - \frac{\partial}{\partial \Lambda} - i \xi \left( \Lambda_0 - \frac{\partial}{\partial \Lambda_0} \right) \right)
\hat{g}^-_\xi \chi = e^{i\frac{\pi}{2}} \frac{1}{2\sqrt{2}hL} \left( \left( \chi + \xi \delta_\xi \chi \right) \left( \xi + \frac{\chi}{2} (L' + L_2') \right) \right) \left( \Lambda + \frac{\partial}{\partial \Lambda} - i \xi \left( \Lambda_0 - \frac{\partial}{\partial \Lambda_0} \right) \right).
$$

(E1)

where

$$
f_{\xi, \chi, n} = \frac{1}{2} \left( (L' \chi (1 - (3 - n)n) + L_3') (1 - \chi (1 - (3 - n)n)) + (\chi + (1 - (3 - n)n)) (L_1' + iL_2 \chi (1 - (3 - n)n)) \right)
$$

(E3)

(the phase factors $e^{i\frac{\pi}{2}}$ 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 (E1) and (E2) on the Weyl symbols is to apply them to isotropic states (D3) (their normalization consistent with (E3) is $r_i(L') = (\frac{(-1)^2}{16\pi})(2l + 1)$):

$$
\sum_{\xi, \chi = -1, 1} \hat{g}^+_\xi \chi \hat{g}^+_\chi \chi = \mu_i(L') 4(l + \frac{1}{2} \hat{\xi} + \hat{\chi}).
$$

(E4)

where $\mu_i(L') = \frac{1}{2l(l + \frac{1}{2} + 1)} \frac{2L'}{h}$ is the additional factor compared to the expected effect of compound ladder operator. The correct form of the compound operator can be found by applying to (E1) and (E2) the following transformation which wipes out this factor:

$$
a_{\xi} \hat{b}_\chi = \delta_0 \hat{g}^-_\xi \chi \delta_0^{-1} = (\sqrt{\frac{2L'}{h}} \hat{g}^-_\xi \chi + \delta_0 \hat{g}^+_\xi \chi) \sqrt{\frac{h}{2L' + 1}}
$$

(E5)

$$
a_{\xi} \hat{b}^\dagger_\chi = \delta_0 \hat{g}^+_\xi \chi \delta_0^{-1} = \sqrt{\frac{2L' + 1}{h}} \sqrt{\frac{h}{2L' \delta_0 \hat{g}^+_\chi \chi}}
$$

(E6)

where $\delta_0$ is the invariance operator (see eq. (E6)):

$$
\delta_0 = \sqrt{\Gamma \left( \frac{2L'}{h} + 2 \right) \Gamma \left( \frac{2L'}{h} + 2 \right) \left( \frac{2L'}{h} \right)}
$$

(E7)

(here $\Gamma(z)$ is the Euler gamma-function) and

$$
\hat{g}^-_\xi \chi = \sum_{n = 0} \left( (L' + \chi L_3) \delta_\xi (-1)^n + (L_{1} - i \chi L_2) \delta_\xi (-1)^n \chi \times i \cdot n \cdot e^{i\xi \chi \Lambda L'} \right) \frac{1}{\Lambda_0 - \frac{\partial}{\partial \Lambda_0} - n \xi \chi \Lambda_0 + \frac{\partial}{\partial \Lambda_0} + i \xi (-1)^n (\Lambda_0 + 2) \frac{\partial}{\partial \Lambda_0} + i \xi \chi L_1 - \frac{\partial}{\partial \Lambda_0} + i \xi \chi L_2 - \frac{\partial}{\partial \Lambda_0}}
$$

(E8)

To deduce the Wigner images $\hat{\lambda}_k$ of the quaternion
operators we need the following well-known relations:
\[ \hat{\lambda}_n = \frac{(-i)^n}{2} D_{\frac{1}{2}, \frac{1}{2}}^{\delta_m, \delta_k} \left\{ \begin{array}{c} l \\delta_m \\delta_k \\ m \end{array} \right\} \left( -1 \right)^{l+m+n} \frac{1}{\sqrt{2(l+1)(2j+1)}} \times \]
\[ \times \left( \begin{array}{c} j \\delta_m \\delta_k \\ k \end{array} \right) \times \left( \begin{array}{c} j \\delta_m+n \\delta_k \\ m \end{array} \right) \right) \] (E9)

\[ D_{\frac{1}{2}, \frac{1}{2}}^{l, m} |l, m, k\rangle = \sum_{j=1}^{l} i^{-j} \sqrt{(2l+1)(2j+1)} \times \left( \begin{array}{c} j \\delta_m \\delta_k \\ k \end{array} \right) \times \left( \begin{array}{c} j \\delta_m+n \\delta_k \\ m \end{array} \right) \] (E10)

where \( D_{l, m} \) are Wigner D-functions. Since

\[ |l+1, m+\mu, k+\kappa\rangle = \frac{\hat{a}_l^\dagger \hat{a}_{-\frac{1}{2}}}{\sqrt{l+\kappa+1}} |l, m, k\rangle, \]

\[ |l-1, m+\mu, k+\kappa\rangle = \frac{\hat{a}_l \hat{a}_{-\frac{1}{2}}}{\sqrt{l-\kappa+1}} |l, m, k\rangle, \] (E11)

\((\kappa, \mu = \pm 1)\) the effect of operators \( \hat{a}_l \) can be represented as a bilinear combination of the ladder operators:

\[ \hat{\lambda}_n = \sum_{l=1}^{\infty} \sum_{m=0}^{\infty} \sum_{k=0}^{\infty} e^{i\pi m/2} a_l \hat{a}_{-\frac{1}{2}}^\dagger a_{l+\frac{1}{2}} b_{l+\frac{1}{2}}^\dagger b_{l-\frac{1}{2}} \left\{ \begin{array}{c} l \\delta_m \\delta_k \\ m \end{array} \right\} \] (E12)

The quantum Liouvillian \( \hat{L} \) for symmetric top in variables \( \{R_1, R_2, \alpha, \beta, L_1, L_2, L_3\} \) takes the form [48] with

\[ \frac{\partial}{\partial R_3} \left( \frac{\partial}{\partial R_3} + \frac{\partial}{\partial R_2} + \frac{\partial}{\partial R_1} \right), \]

\[ \left( \frac{3}{2} \right) \hat{L}_3^2 \hat{L}_3' + \frac{1}{2} \right) \] (F1)

The operators in the spaces \( \Sigma_{W, q}^m \) and \( \Sigma_{W, q}^{\lambda, q} \) are connected via correspondence:

\[ \lambda_m \rightarrow r_m + \frac{\partial}{\partial r_m} = \frac{\partial}{\partial r_m} - \frac{r_m}{\sqrt{2}} \] (F2)

Thus, the effect of \( r_m \) and \( \frac{\partial}{\partial r_m} \) on \( \lambda_m \) is identical to the effect of the ladder operators on the canonical coordinate. Treating the new variables \( r_m \) as being proportional to components of some quaternion we formally can express them in terms of Euler angles \( \alpha \) and \( \gamma \):

\[ r_0 = R_1 \cos \left( \frac{\alpha + \gamma}{2} \right); \]

\[ r_1 = R_2 \cos \left( \frac{\alpha - \gamma}{2} \right); \]

\[ r_2 = R_2 \sin \left( \frac{\alpha - \gamma}{2} \right); \] (F3)

The goal of this Appendix is to derive the Wigner representation in which the quantum generator of motion [48] for the symmetric tops coincides with the corresponding classical Liouvillian \( \mathcal{L} \). We will depart from the \( \{\lambda, q\} \)-representation and convert it into the desired form via series of transformations. The procedure (and the final expressions for \( \hat{\ell} \) and \( \hat{\ell}_k \)) in the case of variables \( \{A, L\} \) remains the same. However, in this case the explicit formulas for Bopp operators \( \hat{L}_k^l \) and \( \hat{\lambda}_m \) are rather cumbersome due to the complicated form of relations \( \Lambda_m(\alpha, L') \) and will not be presented here.

We start with the fractional Laplace transform of variables \( \lambda_m \):

\[ \tilde{\rho}(r, L) = \int_{-\infty}^{\infty} \prod_{m=0}^{\infty} \frac{d\lambda_m}{\lambda_m} \times \]

\[ e^{\sum_{m=0}^{\infty} (\sqrt{\pi} \lambda_m r - \frac{\lambda_m^2}{2} - \frac{\lambda_m^4}{4})} \tilde{\rho}(\lambda, L) \] (F1)

The Appendix F: Nasyrov-type phase representation of the rotational motion

It is easily to check that \( \lim_{n \rightarrow \infty} \hat{\lambda}_m = \lambda_m \), so that all requirements of the postulate (C2) are satisfied. Thus, the Wigner \( \{A, L\} \)-representation defined by eqs. (E5), (E6), (E7) and (E12) is self-consistent and complete.
the operator $\hat{\ell}$ defined by (13) take the form:

$$\hat{P} = -AJ \frac{\partial}{\partial \alpha} + (A-B)K \frac{\partial}{\partial \gamma},$$

(F7)

$$\hat{L}_1 \pm i\hat{L}_2 = e^{i\gamma} \sqrt{\frac{J + K - \frac{h}{2}}{J + K - \frac{h}{2}} h \frac{\partial}{\partial \gamma} \pm \frac{2i(J + K)}{2}} e^{\pm \frac{i}{2} \frac{\partial}{\partial \alpha}};$$

(F8)

$$\hat{L}_3 = K - \frac{1}{2} h \frac{\partial}{\partial \gamma};$$

(F9)

$$\hat{\ell} = J - \frac{1}{2} h \frac{\partial}{\partial \alpha};$$

(F10)

$$\hat{a}_\pm = \frac{e^{\frac{i}{2} \gamma (\pm \alpha)}}{e^{\frac{1}{2} \gamma (\pm 1)} \sqrt{\frac{2(J + K)}{h} - 1}} e^{\frac{i}{2} \gamma (\pm \alpha)};$$

(F11)

$$\hat{a}_\pm^\dagger = \frac{e^{-\frac{i}{2} \gamma (\pm \alpha)}}{e^{-\frac{i}{2} \gamma (\pm 1)} \sqrt{\frac{2(J + K)}{h} - 1}} e^{-\frac{i}{2} \gamma (\pm \alpha)};$$

(F12)

whereas the operators $\hat{b}_\pm$ and $\hat{b}_\pm^\dagger$ are still defined by eqs. (11). It is easy to check that the eqs. (F7) - (F10) have the correct classical limits (recall that the ladder operators (11), (F11) and (F12) are specified up to the invariance transform (15)).

One can see that the dynamic master equation (F7) exactly coincide 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:

$$\langle \hat{\ell}^n, \rho \rangle_W = (\langle J - h/2 \rangle^n, \rho \rangle_W; \langle \hat{L}_3, \rho \rangle_W = (K^n, \rho \rangle_W.$$  

(F13)

so that the associated marginals of the Wigner functions will represent the probability distributions for quantities $\ell$ and $K$ (cf. eq. (9) and subsequent discussion).

The effect of operators $e^{\pm \frac{i}{2} \frac{\partial}{\partial \alpha}}$ and $e^{\pm \frac{i}{2} \frac{\partial}{\partial \gamma}}$ contained in eqs. (F8) - (F11), (F12) on any function of variables $J$ and $K$ consist of discrete replacements: $J \to J \pm \frac{h}{2}$, $K \to K \pm \frac{h}{2}$. For this reason, the parameters $J$ and $K$ take the discrete set of values, so that the Wigner images $\hat{\rho}_{1,2}$ of the projectors $\hat{P}_{1,2} = \{1, 1, k_1 \} \{l_2, m_3, k_2 \}$ read:

$$\hat{\rho}_{1,2} \propto \delta_{l_1, l_1+j_2+1} \delta_{k_1, k_2}$$

(F14)

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

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