Principal-axis hyperspherical description of N-particle systems: Classical treatment

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Principal-axis hyperspherical coordinates (made up of one hyperradius and $3N-7$ angles as internal coordinates) and three Eulerian angles as external (rotational) coordinates are defined from the Eckart coordinate system [Phys. Rev. **46**, 383 (1934)]. They can describe any N-particle system. The exact classical Hamiltonian of the system in terms of these coordinates is derived, and it is remarkably simple. The quantization of this Hamiltonian is deferred for future study.

I. INTRODUCTION

Owing to the fact that the concept of size of an N-particle system refers to a unique length, it is desirable to describe all the possible deformations of the system for a given size by using only one length-type coordinate, the $3N-7$ other internal coordinates being dimensionless (i.e., angles). The hyperspherical (or related) coordinates have been, for a long time, studied for this purpose.[1–23]

A coordinate system is said to be "principal-axis (PA) hyperspherical" if, and only if, the $3N-6$ *internal* coordinates split into one coordinate having the dimension of length (possibly mass weighted) ρ, the hyperradius, the $3N-7$ other coordinates being angles, the so-called hyperangles (which are actually generalized Euler angles), so that the following applies:

(i)

$$\sum_{k=1}^{N}\sum_{g}|r_{kg}|^2=\rho^2 ,$$

where r_{kg} denotes the gth component ($g=x,y,z$) of the *mass-weighted* position vector $\mathbf{r}_k$ of the kth particle viewed in the moving-axis system (MS).

(ii) The MS is a principal-axis system (PAS), i.e., the axes of the MS coincide with the instantaneous principal inertia axes of the system.

(iii) The overall rotation of the MS with respect to the laboratory-axis system (LS) is described by the three usual Euler angles.

Thus, leaving out the center-of-mass motion, the system is described by means of one length and $3N-4$ angles.

It should be emphasized that the PA-hyperspherical coordinates are basically different from the usual hyperspherical coordinates. These coordinates parametrize the components of $N-1$ mass-weighted relative position vectors of the system, $\mathbf{R}_l$ ($l=1,\dots,N-1$), directly in the LS by means of $3N-4$ angles, according to

$$R_{1,X}=\rho\sin\theta_{3N-4}\cdots\sin\theta_3\sin\theta_2\sin\theta_1 ,$$
$$R_{1,Y}=\rho\sin\theta_{3N-4}\cdots\sin\theta_3\sin\theta_2\cos\theta_1 ,$$
$$R_{1,Z}=\rho\sin\theta_{3N-4}\cdots\sin\theta_3\cos\theta_2 ,$$
$$\vdots$$
$$R_{N-1,X}=\rho\sin\theta_{3N-4}\sin\theta_{3N-5}\cos\theta_{3N-6} ,$$
$$R_{N-1,Y}=\rho\sin\theta_{3N-4}\cos\theta_{3N-5} ,$$
$$R_{N-1,Z}=\rho\cos\theta_{3N-4} ,$$

with $0\le\rho<\infty$, $0\le\theta_1\le2\pi$, and $0\le\theta_i\le\pi$ ($i=2,\dots,3N-4$).[24]

Although the hyperradius ρ is the same as noted above,

$$\rho^2=\sum_{l=1}^{N-1}\sum_{G}|R_{lG}|^2 \quad (G=X,Y,Z) ,$$

the role of internal coordinates for N particles (which are $3N-6$ in number) clearly does not suit the usual hyperspherical coordinates. This is disadvantageous, because the hyperspherical coordinates are orthogonal, whereas the PA-hyperspherical coordinates are not, and therefore they cause many mathematical complications. This reflects the price to be paid for preserving the notions of internal coordinates and the rotating system fixed frame, which are so useful in molecular and nuclear physics.

The dynamics of N interacting particles, described by means of curvilinear coordinates, rests on the explicit relations between the $3N$ moving-frame mass-weighted Cartesian coordinates of the particles [collectively denoted by $\mathbf{r}=(r_{1x},r_{1y},r_{1z},r_{2x},\dots,r_{NZ})$] and the $3N-6$ curvilinear internal coordinates [$\mathbf{q}=(q_1,q_2,\dots,q_{3N-6})$]. First, it is possible to start from the $3N$ relationships: $\mathbf{r}=\mathbf{r}(\mathbf{q})$; the MS is thus *implicitly* defined. The alternative possibility is to start from the $3N-6$ relationships: $\mathbf{q}=\mathbf{q}(\mathbf{r})$, by definition invariant under translation and rotation; in this case, supposing that the center of mass is at rest at the origin of both the LS and the MS, the orientation of the latter must be *explicitly* determined by three additional relationships: $0=C_g(\mathbf{r})$, $g=x,y,z$, often called "axial constraints" (for reviews of these topics, see Refs. 25–27).

Here, the former attitude will be adopted throughout. A summary of what its application requires in actual practice is presented in Sec. II. In Sec. III, after recalling that the use of Jacobi vector components instead of position vector components greatly simplifies the problem, the PA-hyperspherical coordinates are given a mathematical definition, with the help of a formal analogy between Jacobi vector components and orthogonal matrix elements. This approach was first used by Eckart[28] long ago and has been rediscovered recently by Robert and Baudon.[29,30] The basic algebra for using, in practice, the PA-hyperspherical coordinates is presented in Sec. IV, including the derivation of the exact classical mechanical Hamiltonian. There is *no limitation* of the number of particles.

The quantization of this Hamiltonian, and in particular the study of the commutation relations of the operators associated with the quasimomenta introduced below, is planned as the subject of a subsequent article.[31] Finally, the physical interpretation of the hyperspherical description of an N-particle system will be thoroughly discussed, along the line of the arguments of both Aquilanti and coworkers[21–23] and Robert and Baudon,[29,30] in a third paper.

The authors are aware of the tedium of some passages below; in particular, in Sec. IV. They, however, beg the reader to realize that the reported calculations are just a small part of the calculations actually undertaken. Presenting fewer of these would mean preventing one from checking. But calculation is nothing but a means to an end. Most worthy of consideration is the concise—and hopefully important—result in Sec. IV D below.

II. CLASSICAL MECHANICAL EXPRESSION OF THE KINETIC ENERGY DERIVED FROM $\mathbf{r}=\mathbf{r}(\mathbf{q})$

A. Classical kinetic energy

The exact classical Hamiltonian expression of the kinetic energy T of the N particles decomposes (after separation of the center-of-mass motion) as follows:[27]

$$2T=(\mathbf{P}_q \quad \mathbf{J})\cdot\mathbf{g}(\mathbf{q})\cdot\begin{bmatrix}\mathbf{P}_q^T\\ \mathbf{J}^T\end{bmatrix}, \tag{1}$$

where

$$\mathbf{J}=\mathbf{P}_E\cdot\boldsymbol{\omega}^{*-1\,T}(\mathbf{E}) \tag{2a}$$

and

$$\mathbf{g}(\mathbf{q})=\begin{bmatrix}\mathbf{S}(\mathbf{q}) & \mathbf{C}^T(\mathbf{q})\\ \mathbf{C}(\mathbf{q}) & \mathbf{I}(\mathbf{q})\end{bmatrix}^{-1}=\begin{bmatrix}1 & -\mathbf{S}^{-1}\cdot\mathbf{C}^T\\ 0 & 1\end{bmatrix}\cdot\begin{bmatrix}\mathbf{S}^{-1}(\mathbf{q}) & 0\\ 0 & \boldsymbol{\mu}(\mathbf{q})\end{bmatrix}\cdot\begin{bmatrix}1 & 0\\ -\mathbf{C}\cdot\mathbf{S}^{-1} & 1\end{bmatrix}, \tag{2b}$$

$\mathbf{P}_q$ and $\mathbf{P}_E$ are row-vector shorthand notations for the momenta conjugate to, respectively, the $3N-6$ internal coordinates $\mathbf{q}$ (considered curvilinear) and the three Euler angles α, β, and γ, collectively denoted by $\mathbf{E}$. Matrix $\boldsymbol{\omega}^{*-1}(\mathbf{E})$ is that of the transformation[32]

$$\mathbf{J}^T=\begin{bmatrix}J_x\\ J_y\\ J_z\end{bmatrix}=\begin{bmatrix}\cot\beta\cos\gamma & \sin\gamma & -\cos\gamma/\sin\beta\\ -\cot\beta\sin\gamma & \cos\gamma & \sin\gamma/\sin\beta\\ 1 & 0 & 0\end{bmatrix}\cdot\begin{bmatrix}P_\gamma\\ P_\beta\\ P_\alpha\end{bmatrix} \tag{3}$$

from $\mathbf{P}_E$ to $\mathbf{J}$, the angular momentum vector, whose *components along the MS axes* are, respectively, J_x, J_y, and J_z. $\mathbf{S}(\mathbf{q})$ is the $(3N-6)$-dimensional symmetric matrix of elements:

$$S_{ij}(\mathbf{q})=\sum_{k=1}^{N}\left[\frac{\partial r_{kx}}{\partial q_i}\frac{\partial r_{kx}}{\partial q_j}+\frac{\partial r_{ky}}{\partial q_i}\frac{\partial r_{ky}}{\partial q_j}+\frac{\partial r_{kz}}{\partial q_i}\frac{\partial r_{kz}}{\partial q_j}\right] \quad (i,j=1,\ldots,3N-6)\ , \tag{4}$$

i.e., the contravariant components of the metric tensor of the transformation $\mathbf{r}=\mathbf{r}(\mathbf{q})$, $\mathbf{C}(\mathbf{q})$ is the $3\times(3N-6)$ Coriolis matrix of elements:

$$C_{gi}(\mathbf{q})=\sum_{k=1}^{N}\left[r_{kg'}\frac{\partial r_{kg''}}{\partial q_i}-r_{kg''}\frac{\partial r_{kg'}}{\partial q_i}\right] \quad (i=1,\ldots,3N-6;\ \ g\,g'\,g''\text{ is an even permutation of }x\,y\,z)\ , \tag{5}$$

and

$$\boldsymbol{\mu}(\mathbf{q})=[\mathbf{I}^*(\mathbf{q})]^{-1}\ , \tag{6}$$

where

$$\mathbf{I}^*(\mathbf{q})=\mathbf{I}(\mathbf{q})-\mathbf{C}(\mathbf{q})\cdot\mathbf{S}^{-1}(\mathbf{q})\cdot\mathbf{C}^T(\mathbf{q}) \tag{7}$$

is the effective inertia tensor, $\mathbf{I}(\mathbf{q})$ being the usual inertia tensor represented by the three-dimensional symmetric matrix of elements:

$$I_{gg}(\mathbf{q})=\sum_{k=1}^{N}(r_{kg'}^2+r_{kg''}^2) \quad (g\,g'\,g''\text{ is a permutation of }x\,y\,z)\ , \tag{8a}$$

$$I_{gg'}(q)=-\sum_{k=1}^{N}r_{kg}r_{kg'} \quad (g,g'=x,y,z)\ . \tag{8b}$$

B. Quasivelocities and quasimomenta

An important point should be discussed now. Equation (1) is nothing but the Hamiltonian counterpart of the Lagrangian expression of the kinetic energy:

$$2T=(\dot{\mathbf{q}}\ \ \boldsymbol{\omega})\cdot\begin{bmatrix}\mathbf{S}(\mathbf{q}) & \mathbf{C}^T(\mathbf{q})\\ \mathbf{C}(\mathbf{q}) & \mathbf{I}(\mathbf{q})\end{bmatrix}\cdot\begin{bmatrix}\dot{\mathbf{q}}^T\\ \boldsymbol{\omega}^T\end{bmatrix}. \tag{9}$$

The transformation has been achieved with the help of the usual definition of conjugate momenta, $\mathbf{P}=\partial T/\partial\mathbf{V}$, where $\mathbf{V}=(\dot{\mathbf{q}}\ \ \boldsymbol{\omega})$ is the $(3N-3)$-dimensional velocity vector, whose components are, respectively, the internal and angular velocities, so that

$$\mathbf{P}=(\mathbf{P}_q\ \ \mathbf{J})=(\dot{\mathbf{q}}\ \ \boldsymbol{\omega})\cdot\begin{bmatrix}\mathbf{S}(\mathbf{q}) & \mathbf{C}^T(\mathbf{q})\\ \mathbf{C}(\mathbf{q}) & \mathbf{I}(\mathbf{q})\end{bmatrix}$$

and $2T$ takes the same form as in Eq. (1), $2T=\mathbf{P}\cdot\mathbf{g}(\mathbf{q})\cdot\mathbf{P}^T$.

Now, one is always at liberty to use a *nonsingular* $(3N-3)$-dimensional square matrix $\mathbf{B}(\mathbf{q})$ to transform the velocities into quasivelocities, namely,

$$\mathbf{V}'=\mathbf{V}\cdot\mathbf{B}(\mathbf{q}), \tag{10a}$$

so that

$$2T=\mathbf{V}'\cdot\mathbf{B}^{-1}\cdot\begin{bmatrix}\mathbf{S} & \mathbf{C}^T\\ \mathbf{C} & \mathbf{I}\end{bmatrix}\cdot\mathbf{B}^{-1\,T}\cdot\mathbf{V}'^T$$

whence

$$\mathbf{P}'=\frac{\partial T}{\partial\mathbf{V}'}=\mathbf{V}'\cdot\mathbf{B}^{-1}\cdot\begin{bmatrix}\mathbf{S} & \mathbf{C}^{\mathrm{T}}\\ \mathbf{C} & \mathbf{I}\end{bmatrix}\cdot\mathbf{B}^{-1\,T} \tag{10b}$$

and

$$2T=\mathbf{P}'\cdot\mathbf{B}^T(\mathbf{q})\cdot\mathbf{g}(\mathbf{q})\cdot\mathbf{B}(\mathbf{q})\cdot\mathbf{P}'^T. \tag{10c}$$

In classical mechanics, this is just a canonical transformation, generally invoked if it simplifies the matrix inversion problem, namely, if there exists a matrix $\boldsymbol{\Gamma}(\mathbf{q})$, easy to invert, $\boldsymbol{\Gamma}^{-1}(\mathbf{q})=\mathbf{g}'(\mathbf{q})$, which allows to write the Lagrangian expression of T in terms of quasivelocities, $2T=\mathbf{V}'\cdot\boldsymbol{\Gamma}(\mathbf{q})\cdot\mathbf{V}'^T$, and thus the Hamiltonian expression in terms of quasimomenta, $2T=\mathbf{P}'\cdot\mathbf{g}'(\mathbf{q})\cdot\mathbf{P}'^T$. A simple example of this is the generalized Wilson-Howard[33] and Darling-Dennison[34] expression of T:

$$2T=(\mathbf{P}_q\ \ \mathbf{J}')\cdot\mathbf{g}'(\mathbf{q})\cdot\begin{bmatrix}\mathbf{P}_q^T\\ \mathbf{J}'^T\end{bmatrix},$$

where it is obvious from (1) and (2) that

$$\mathbf{g}'(\mathbf{q})=\begin{bmatrix}\mathbf{S}^{-1}(\mathbf{q}) & \mathbf{0}\\ \mathbf{0} & \boldsymbol{\mu}(\mathbf{q})\end{bmatrix}$$

and $\mathbf{J}'=\mathbf{J}-\mathbf{P}_q\cdot\mathbf{S}^{-1}(\mathbf{q})\cdot\mathbf{C}^T(\mathbf{q})$ is the generalized angular momentum vector (in the original form of Darling and Dennison, the coordinates are normal, i.e., $\mathbf{S}=\mathbf{1}$, and $\mathbf{J}'=\mathbf{J}-\mathbf{P}_q\cdot\mathbf{C}^T$, where $\mathbf{P}_q\cdot\mathbf{C}^T$ is the so-called *vibrational angular momentum*).

In quantum mechanics, it is not immaterial whether one uses momenta or quasimomenta. Indeed, the action of momentum operators on wave functions consists either in partial differentiations with respect to coordinates, or preferably in creation and annihilation operators if standard bases can be derived for these operators. In all cases, this is possible if, and only if, particular commutation relationships of the quasimomenta have been established beforehand. The most celebrated example of quasivelocities and quasimomenta is the one of the MS components of the angular velocity and angular momentum vectors, e.g., $[J_x,J_y]=-i\hbar J_z$, and so on, and the $\mathcal{D}^J_{M\Omega}(\mathbf{E})$ Wigner functions as basis functions.[35]

Both reasons (simplification of the analytical matrix inversion problem in the present article, and particular commutation relations resulting in efficient standard representations for the wave function in the planned future work) will justify the use of the quasivelocities and quasimomenta initially suggested by Eckart.[28]

III. MATHEMATICAL DEFINITION OF THE PA-HYPERSPHERICAL COORDINATES: THE ECKART SUBGROUP OF SO(n)

It is well known that a system of N particles can always be replaced by $n=N-1$ *relative particles* and the center of mass. More precisely, the relation between, on the one hand, N mass-weighted (by $\sqrt{m_k}$) particle position vectors $\mathbf{r}_k$ $(k=1,N)$ and, on the other hand, $n=N-1$ mass-weighted (by $\sqrt{\mu_\lambda}$) relative Jacobi vectors $\boldsymbol{\rho}_\lambda$ $(\lambda=1,n)$ and the mass-weighted [by $\sqrt{M}=(\sum_k m_k)^{1/2}$] position vector of the center of mass $\boldsymbol{\rho}_N$, is given by

$$\mathbf{r}_k=\sum_{k'=1}^{N}\sigma_{kk'}(m_1,\ldots,m_N)\boldsymbol{\rho}_{k'}, \tag{11}$$

where $\sigma_{kk'}(m_1,\ldots,m_N)$ $(k,k'=1,\ldots,N)$ are the elements (depending only on the masses of the particles) of an orthogonal matrix $\boldsymbol{\sigma}$, such that

$$\sum_{k''=1}^{N}\sigma_{kk''}\sigma_{k'k''}=\delta_{kk'}.$$

The orthogonality property of $\boldsymbol{\sigma}$ can be directly inferred from the conservation of the arc length that is required for the kinetic energy to be invariant, namely,

$$2T=\sum_{k=1}^{N}m_k(\dot{x}_k^2+\dot{y}_k^2+\dot{z}_k^2)=\sum_{k=1}^{N}\dot{\mathbf{r}}_k^2=\dot{\boldsymbol{\rho}}_N^2+\sum_{\lambda=1}^{n}\dot{\boldsymbol{\rho}}_\lambda^2$$

implies that

$$\sum_{k=1}^{N}d\mathbf{r}_k^2=\sum_{\lambda=1}^{n}d\boldsymbol{\rho}_\lambda^2+d\boldsymbol{\rho}_N^2. \tag{12}$$

Owing to the orthogonality of matrix $\boldsymbol{\sigma}$, and from Eqs. (4), (5), and (8), it comes out

$$S_{ii'}(q_1,\ldots,q_{3N-6})=\sum_{k=1}^{N}\sum_{g}\frac{\partial r_{kg}}{\partial q_i}\frac{\partial r_{kg}}{\partial q_{i'}}=\sum_{\lambda=1}^{n}\sum_{g}\frac{\partial\rho_{\lambda g}}{\partial q_i}\frac{\partial\rho_{\lambda g}}{\partial q_{i'}}\qquad(i,i'=1,\ldots,3N-6), \tag{13}$$

$$C_{gi}(q_1,\ldots,q_{3N-6})=\sum_{k=1}^{N}\sum_{g',g''} r_{kg'}\frac{\partial r_{kg''}}{\partial q_i}\varepsilon_{gg'g''}=\sum_{\lambda=1}^{n}\sum_{g',g''}\rho_{\lambda g'}\frac{\partial \rho_{\lambda g''}}{\partial q_i}\varepsilon_{gg'g''}\quad (i=1,\ldots,3N-6;\ g=x,y,z)\ , \tag{14}$$

where $\varepsilon_{gg'g''}$ is the signature of the permutation $gg'g''$ of xyz, and

$$I_{gg'}(q_1,\ldots,q_{3N-6})=\sum_{k=1}^{N}\left[\sum_{g''} r_{kg''}^2\delta_{gg'}-r_{kg}r_{kg'}\right]=\sum_{\lambda=1}^{n}\left[\sum_{g''}\rho_{\lambda g''}^2\delta_{gg'}-\rho_{\lambda g}\rho_{\lambda g'}\right]\quad (g,g'=x,y,z)\ . \tag{15}$$

A. Eckart's formulation

More than 55 years ago, Eckart proposed a solution to the problem of N-particle classical dynamics[28] that perfectly suits the PA-hyperspherical coordinate parametrization. The present work is directly descended from Eckart's approach. We reformulate it as follows.

First, let us recall that the Whitten and Smith initial hyperspherical coordinates ρ', Θ, and Φ for a three-body system were defined as[4]

$$r'_{1x}=d\rho'\cos\Theta\cos\Phi,\quad r'_{2x}=d^{-1}\rho'\cos\Theta\sin\Phi\ ,$$
$$r'_{1y}=-d\rho'\sin\Theta\sin\Phi,\quad r'_{2y}=d^{-1}\rho'\sin\Theta\cos\Phi\ , \tag{16}$$

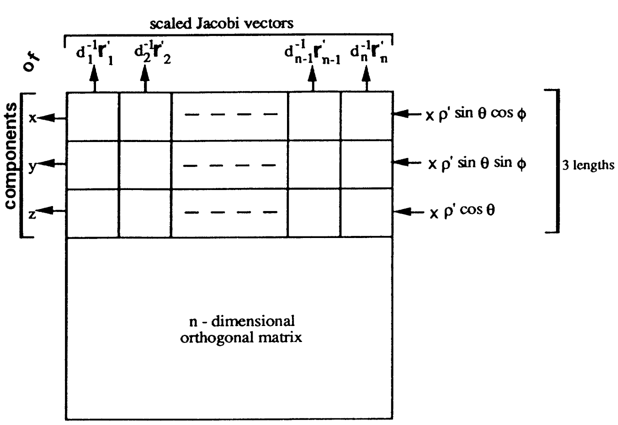

FIG. 1. The MS components of the scaled Jacobi vectors $d_\lambda^{-1}\mathbf{r}'_\lambda$ are by construction equal to the product of an element of the three first lines of an n-dimensional orthogonal matrix depending on $3n-6$ angles, by one out of three length-type quantities, for the x,y, and z-components, respectively. These quantities are actually the three instantaneous gyration radii of the system and, for the PA-hyperspherical coordinates, they are parametrized by means of spherical coordinates:

$$d_\lambda=\sqrt{\mu/\mu_\lambda},\quad \mu=\frac{\sqrt{m_1\times m_2\times\cdots\times m_N}}{(m_1+m_2+\cdots+m_N)^{(N/2)-1}}\ ,$$

and μ_λ is the reduced mass corresponding to the Jacobi vector $\mathbf{r}'_\lambda$ $(\lambda=1,n)$.

i.e., a direct parametrization of Jacobi-vector MS components, with $d=(\mu_{12,3}/\mu)^{1/2}$, a scaling factor, such that

$$\mu\rho'^2=\mu_{12}(\mathbf{r}'_1)^2+\mu_{12,3}(\mathbf{r}'_2)^2\ , \tag{17}$$

$\mu=(m_1m_2m_3/M)^{1/2}$ is a totally symmetrized characteristic mass (M is the total mass), and $\mu_{12,3}=(m_1+m_2)m_3/M$ and $\mu_{12}=m_1m_2/(m_1+m_2)$ are the usual reduced masses.

Formally speaking, since the system reduces to *two* Jacobi vectors, which determine a *plane* of $\mathbb{R}^3$, the four vector components in Eq. (16) can be identified with those of a *planar* rotation matrix (angle Φ), weighted by either $\rho'\cos\Theta$ or $\rho'\sin\Theta$, i.e., any two lengths suitably parametrized, and finally rescaled ($d_1=d$ and $d_2=d^{-1}$).

More recently, by formal analogy with the three-particle case, the nine components of the *three* Jacobi vectors of a four-particle system have been identified with those of a *three-dimensional* rotation matrix (Euler matrix with angles Φ, Θ, and X), suitably weighted by any three lengths parametrized by spherical coordinates (ρ', Ω, and Λ) and rescaled.[36] The result is

$$\begin{aligned}
r'_{1x}&=d_1\rho'\sin\Omega\cos\Lambda(\cos\Theta\cos\Phi\cos X-\sin\Phi\sin X)\ ,\\
r'_{1y}&=-d_1\rho'\sin\Omega\sin\Lambda(\cos\Theta\cos\Phi\sin X+\sin\Phi\cos X)\ ,\\
r'_{1z}&=d_1\rho'\cos\Omega\sin\Theta\cos\Phi\ ,\\
r'_{2x}&=d_2\rho'\sin\Omega\cos\Lambda(\cos\Theta\sin\Phi\cos X+\cos\Phi\sin X)\ ,\\
r'_{2y}&=-d_2\rho'\sin\Omega\sin\Lambda(\cos\Theta\sin\Phi\sin X-\cos\Phi\cos X)\ ,\\
r'_{2z}&=d_2\rho'\cos\Omega\sin\Theta\sin\Phi\ ,\\
r'_{3x}&=-d_3^{-1}\rho'\sin\Omega\cos\Lambda\sin\Theta\cos X\ ,\\
r'_{3y}&=d_3^{-1}\rho'\sin\Omega\sin\Lambda\sin\Theta\sin X\ ,\\
r'_{3z}&=d_3^{-1}\rho'\cos\Omega\cos\Theta\ ,
\end{aligned} \tag{18}$$

where $d_1=(\mu/\mu_{12})^{1/2}$, $d_2=(\mu/\mu_{12,3})^{1/2}$, and $d_3=(\mu_{123,4}/\mu)^{1/2}$ are scaling factors such that

$$\mu\rho'^2=\mu_{12}(\mathbf{r}'_1)^2+\mu_{12,3}(\mathbf{r}'_2)^2+\mu_{123,4}(\mathbf{r}'_3)^2\ , \tag{19}$$

$\mu=(m_1m_2m_3m_4)^{1/2}/M$ (M is the total mass) is the totally symmetric characteristic mass, and

$$\mu_{12}=m_1m_2/(m_1+m_2)\ ,$$

$\mu_{12,3}=(m_1+m_2)m_3/(m_1+m_2+m_3)$ and $\mu_{123,4}=(m_1+m_2+m_3)m_4/M$ are the usual reduced masses associated with the three Jacobi vectors.

Now, in the general N-particle case, the problem reduces to $n=N-1$ Jacobi vectors and appropriate reduced masses. The formal analogy between the Jacobi vector MS components and the weighted elements of an n-dimensional orthogonal matrix can be extended. The procedure to do this is schematically illustrated in Fig. 1, where, now, $d_\lambda=(\mu/\mu_\lambda)^{1/2}$,

$$\mu=(m_1\times m_2\times\cdots\times m_N)^{1/2}/M^{(N/2)-1}\ ,$$

$M=m_1+m_2+\cdots+m_N$, and μ_λ is the appropriate reduced mass associated with the Jacobi vector $\mathbf{r}'_\lambda$ $(\lambda=1,n)$.

Eckart elegantly identified the three quantities a_g ($g=x,y,z$) with the three usual gyration radii of the system and the mass-weighted Jacobi vectors $\mathbf{r}_\lambda=\sqrt{\mu_\lambda}\mathbf{r}'_\lambda$ with $\sqrt{\mu}d_\lambda^{-1}\mathbf{r}'_\lambda$, so that the following can be readily established.[28–30]

(i)

$$\sum_{\lambda=1}^{n}\mu_\lambda(\mathbf{r}'_\lambda)^2=\sum_{\lambda=1}^{n}(\mathbf{r}_\lambda)^2=\mu\rho'^2=\rho^2\ , \tag{20a}$$

ρ being the mass-weighted (by $\sqrt{\mu}$) hyperradius.

(ii)

$$2T=\sum_{\lambda=1}^{n}\mu_\lambda(d\mathbf{r}'_\lambda/dt)^2=\sum_{\lambda=1}^{n}(d\mathbf{r}_\lambda/dt)^2\ , \tag{20b}$$

T being the classical kinetic energy.

(iii)

$$\begin{aligned}&I_{xx}=\mu(a_y^2+a_z^2)=\rho^2(1-\sin^2\theta\cos^2\phi)\ ,\\ &I_{yy}=\mu(a_z^2+a_x^2)=\rho^2(1-\sin^2\theta\sin^2\phi)\ ,\\ &I_{zz}=\mu(a_x^2+a_y^2)=\rho^2\sin^2\theta,\quad I_{xy}=I_{yz}=I_{zx}=0\end{aligned} \tag{20c}$$

I being the inertia tensor, i.e., the MS is implicitly a PAS.

All conditions required are thus put together to identify PA-hyperspherical coordinates. These are, namely, (i) ρ, the hyperradius; θ and ϕ, the spherical coordinates to parametrize the mass-weighted gyration radii (θ and $\phi\in[0,\pi/2]$); (ii) α, β, and γ, three usual Eulerian angles to orientate the PAS-MS with respect to the LS; and (iii) $3n-6=3N-9$ angles appearing in the three first lines of an n-dimensional orthogonal matrix.

Here lies the only difficulty with Eckart's original idea. Eckart shows that the orthogonal n-dimensional matrix can be considered the eigenvector matrix of the symmetric Gram matrix of the Jacobi vectors $\mathbf{r}'_\lambda$ (i.e., the matrix of element $\mathbf{r}'_\lambda\cdot\mathbf{r}'_{\lambda'}$).[28,30] As such, due to the fact that only three of the n Jacobi vectors are linearly independent in $\mathbb{R}^3$, whence only three eigenvalues of the Gram matrix are nonidentically zero (namely, a_x^2, a_y^2, and a_z^2) the orthogonal matrix should depend on $3n-6$ angles only. Although this observation is true, it is of little use for practical applications. The inadequacy is removed as follows. In practice, we do not use the Jacobi vector components to define the elements of the orthogonal matrix (via the diagonalization of the Gram matrix), but we do the reverse. Namely, the elements of the three first lines of an orthogonal matrix (suitably parametrized) help to parametrize the Jacobi vector components.

However, Eckart's basic idea is preserved, providing that an n-dimensional orthogonal matrix depending only on $3n-6$ angles is properly constructed. This is achieved in Sec. III B.

B. The Eckart subgroup E_{3n-6} of the group of rotations of $\mathbb{R}^n$, SO(n)

A celebrated theorem states that[24] any rotation g of the SO(n) group of rotations of $\mathbb{R}^n$ $(x_1,x_2,\ldots,x_n)$ can be written

$$g=g_{(1)}g_{(2)}\cdots g_{(n-1)}\ , \tag{21}$$

where

$$g_{(k)}=g_k(\theta_k^{n-1})g_{k-1}(\theta_{k-1}^{n-2})\cdots g_1(\theta_1^{n-k}) \tag{22}$$

and $g_i(\alpha)$ denotes the elementary rotation of angle α inside the (x_i,x_{i+1}) plane of $\mathbb{R}^n$.

Apart from the poles where one of the angles θ_k^k $(2\le k\le n-1)$ is 0 or π, the above expressions define g uniquely. The angles θ_j^k $(1\le k\le n-1,\ 1\le j\le k)$ are the generalized Eulerian angles for rotation g. They are $n(n-1)/2$ in number. By construction, the range of variation of the Eulerian angles is

$$0\le\theta_1^k<2\pi,\quad 0\le\theta_j^k<\pi\quad(j>1)\ .$$

Now, an n-dimensional orthogonal matrix $\mathbf{g}$ is a representation of g in the natural basis of $\mathbb{R}^n$. In particular,

$$\mathbf{g}_i(\alpha)=\begin{pmatrix}1&&&&&&&\\&\ddots&&&&&0&\\&&1&&&&&\\&&&\cos\alpha&\sin\alpha&&&\\&&&-\sin\alpha&\cos\alpha&&&\\&&&&&1&&\\&&0&&&&\ddots&\\&&&&&&&1\end{pmatrix}\begin{matrix}1\\ \vdots\\ \vdots\\ i\\ i+1\\ \vdots\\ \vdots\\ n\end{matrix}\ . \tag{23}$$
$$\qquad 1\ \cdots\ \cdots\ i\quad i+1\ \cdots\ \cdots\ n$$

If matrix **g** is written down,

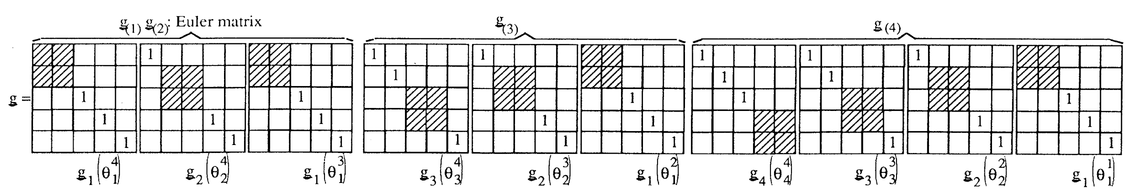

FIG. 2. Explicit construction of matrix **g**, in the case where $n=5$. Each shaded 2×2 matrix represents a planar rotation, by the angle indicated in the last line.

$$\mathbf{g}=\mathbf{g}_1(\theta_1^{n-1})\mathbf{g}_2(\theta_2^{n-1})\mathbf{g}_1(\theta_1^{n-2})\mathbf{g}_3(\theta_3^{n-1})\mathbf{g}_2(\theta_2^{n-2}) \times \mathbf{g}_1(\theta_1^{n-3})\mathbf{g}_4(\theta_4^{n-1})\mathbf{g}_3(\theta_3^{n-2})\mathbf{g}_2(\theta_2^{n-3})\mathbf{g}_1(\theta_1^{n-4})\cdots = \mathbf{g}_{(1)}\mathbf{g}_{(2)}\mathbf{g}_{(3)}\mathbf{g}_{(4)}\cdots , \quad (24)$$

or, in a more illustrative way in Fig. 2 (for $n=5$), where each shaded 2×2 matrix depends on one angle, two important properties emerge.

(i) If $n>2$, a usual three-dimensional Euler matrix (according to Goldstein's convention[37]) is factorized in the upper left corner of $\mathbf{g}$. This will be shown later on to have an important physical consequence.

(ii) If $n>3$, the three last columns of $\mathbf{g}$ depend on $3n-6$ angles only, namely,

$$\theta_1^{n-1},\theta_2^{n-1},\theta_1^{n-2},(\theta_3^{n-1},\theta_2^{n-2},\theta_1^{n-3}),\ldots,(\theta_{n-1}^{n-1},\theta_{n-2}^{n-2},\theta_{n-3}^{n-3}) .$$

The latter property is recursively demonstrated. It is obviously true for $n=4$ [then $3n-6=n(n-1)/2$]. It is also true for $n=5$ (see the developed expression in Fig. 2 above, taking into account that the last matrix $\mathbf{g}_1(\theta_1^1)$ does not modify the last three columns of $\mathbf{g}$). Let us suppose it to be true for $n-1$. The situation for n is illustrated in Fig. 3. Clearly, starting from θ_{n-4}^{n-4} up to θ_1^1, all the Eulerian angles appearing in 2×2 shaded matrices that do not overlap the three last columns of, respectively, $\mathbf{g}_{n-4}$ to $\mathbf{g}_1$, do not appear in the three last columns of the product matrix $\mathbf{g}$. Q.E.D.

As a matter of fact, the three last columns of $\mathbf{g}$, which actually depend on $3n-6$ angles only, could be used as well as the three first lines to define Jacobi-vector components; for example, by transposition of $\mathbf{g}$ with respect to the secondary diagonal. But then property (i) would be lost, which we do not want. We therefore construct an *ad hoc* subgroup of SO(n), which we call the Eckart subgroup, and note E_{3n-6}, whose elements g fulfill the two following requirements.

(i) If $n>2$, a three-dimensional Euler rotation appears on the left in the developed expression of $\bar{g}$.

(ii) If $n>2$, $\bar{g}$ depends on $3n-6$ Eulerian angles only.

The construction of E_{3n-6} is illustrated in the case $n=7$, but it can straightforwardly be recursively generalized to any value of n. Indeed, for $n=7$

$$g=g_1(\theta_1^6)g_2(\theta_2^6)g_1(\theta_1^5)g_3(\theta_3^6)g_2(\theta_2^5)g_1(\theta_1^4)g_4(\theta_4^6) \times g_3(\theta_3^5)g_2(\theta_2^4)g_1(\theta_1^3)g_5(\theta_5^6)g_4(\theta_4^5)g_3(\theta_3^4)g_2(\theta_2^3) \times g_1(\theta_1^2)g_6(\theta_6^6)g_5(\theta_5^5)g_4(\theta_4^4)g_3(\theta_3^3)g_2(\theta_2^2)g_1(\theta_1^1) . \quad (25)$$

Now, owing to the fact that two planar rotations g_i and g_j commute if $|i-j|>1$ (this property is immediately observed on block-diagonal matrices $\mathbf{g}_i$ and $\mathbf{g}_j$), g can be rewritten in the form

$$g=g_1(\theta_1^6)g_2(\theta_2^6)g_1(\theta_1^5)g_3(\theta_3^6)g_2(\theta_2^5)g_1(\theta_1^4)g_4(\theta_4^6)g_3(\theta_3^5) \times g_2(\theta_2^4)g_5(\theta_5^6)g_4(\theta_4^5)g_3(\theta_3^4)g_6(\theta_6^6)g_5(\theta_5^5)g_4(\theta_4^4) \times g_1(\theta_1^3)g_2(\theta_2^3)g_1(\theta_1^2)g_3(\theta_3^3)g_2(\theta_2^2)g_1(\theta_1^1) . \quad (26)$$

$\bar{g}\in E_{3n-6}$ is defined by setting all the angles of the planar rotations of the last line in the product of Eq. (26) equal to zero.

As far as orthogonal matrices are concerned, the following theorem holds. An n-dimensional orthogonal matrix depending on $3n-6$ angles can be built in all generality as follows:

$$\bar{\mathbf{g}}=\mathbf{g}_{(1)}\mathbf{g}_{(2)}\bar{\mathbf{g}}_{(3)}\bar{\mathbf{g}}_{(4)}\cdots\bar{\mathbf{g}}_{(n-1)} , \quad (27)$$

where

$$\mathbf{g}_{(1)}=\mathbf{g}_1(\theta_1^{n-1}) ,$$
$$\mathbf{g}_2=\mathbf{g}_2(\theta_2^{n-1})\mathbf{g}_1(\theta_1^{n-2}) ,$$
$$\bar{\mathbf{g}}_{(k)}=\mathbf{g}_k(\theta_k^{n-1})\mathbf{g}_{k-1}(\theta_{k-1}^{n-2})\mathbf{g}_{k-2}(\theta_{k-2}^{n-3}) \quad (k\geq 3) ,$$

and $\theta_1^i\in[0,2\pi[$, $\theta_j^i\in[0,\pi[$ $(i\geq j>1)$. It is worthwhile to note that the $3n-6$ angles appearing in $\bar{\mathbf{g}}$ are precisely those on which the three last columns of the general rotation matrix $\mathbf{g}$ depend.

Finally, just for convenience, the Euler angles in $\bar{\mathbf{g}}$ are renumbered as follows:

$$\mathbf{g}_{(1)}=\mathbf{g}_1(\theta_1) ,$$
$$\mathbf{g}_{(2)}=\mathbf{g}_2(\theta_2)\mathbf{g}_1(\theta_3) , \quad (28)$$
$$\bar{\mathbf{g}}_{(k)}=\mathbf{g}_k(\theta_{3k-5})\mathbf{g}_{k-1}(\theta_{3k-4})\mathbf{g}_{k-2}(\theta_{3k-3}) \quad (k\geq 3) ,$$

with $(\theta_1,\theta_3,\theta_6)\in[0,2\pi[$ and $(\theta_2,\theta_4,\theta_5,\theta_{i\geq 7})\in[0,\pi[$.

The following convention will be used hereafter for the Euler matrix $\mathbf{\Phi}(\theta_1,\theta_2,\theta_3)=\mathbf{g}_1(\theta_1)\mathbf{g}_2(\theta_2)\mathbf{g}_1(\theta_3)$, written as a 3×3 orthogonal matrix:

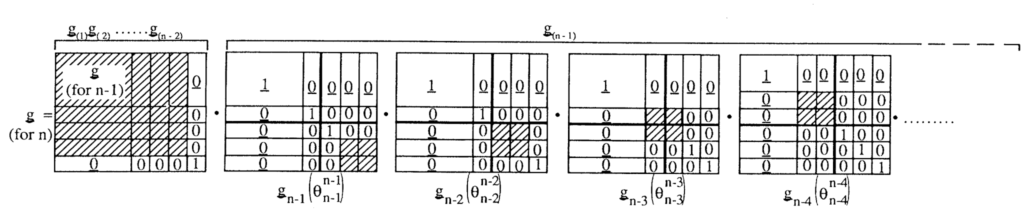

FIG. 3. Schematic illustration of the fact that, according to the mathematical construction of matrix $\mathbf{g}$ (for n), all the angles θ_{n-4}^{n-4} to θ_1^1 do not appear in the last three columns of $\mathbf{g}$. Each shaded 2×2 matrix represents a planar rotation.

$$\begin{bmatrix} \Phi_{xx}\Phi_{xy}\Phi_{xz} \\ \Phi_{yx}\Phi_{yy}\Phi_{yz} \\ \Phi_{zx}\Phi_{zy}\Phi_{zz} \end{bmatrix} = \begin{bmatrix} \cos\theta_1\cos\theta_3-\sin\theta_1\cos\theta_2\sin\theta_3 & \cos\theta_1\sin\theta_3+\sin\theta_1\cos\theta_2\cos\theta_3 & \sin\theta_1\sin\theta_2 \\ -\sin\theta_1\cos\theta_3-\cos\theta_1\cos\theta_2\sin\theta_3 & -\sin\theta_1\sin\theta_3+\cos\theta_1\cos\theta_2\cos\theta_3 & \cos\theta_1\sin\theta_2 \\ \sin\theta_2\sin\theta_3 & -\sin\theta_2\cos\theta_3 & \cos\theta_2 \end{bmatrix} . \tag{29}$$

IV. CLASSICAL HAMILTONIAN EXPRESSION OF THE KINETIC ENERGY IN TERMS OF PA-HYPERSPHERICAL COORDINATES

The problem is now to particularize the results of Sec. II, in light of the results of Sec. III.

A. Construction of matrices S and C

The starting point consists of the relations ($\lambda=1,\ldots,n=N-1$; $g=x,y,z$):

$$\rho_{\lambda g}(q_1,\ldots,q_{3N-6})=a_g(\rho,\theta,\phi)G_{g\lambda}(\theta_1,\ldots,\theta_{3n-6}) , \tag{30}$$

where

$$a_x=\rho\sin\theta\cos\phi,\quad a_y=\rho\sin\theta\sin\phi,\quad a_z=\rho\cos\theta \tag{31}$$

is a spherical representation of the three gyration radii of the N-particle system, and $G_{g\lambda}$ is an element of the three first lines of a rotation matrix $\bar{\mathbf{g}}$ in $\mathbb{R}^n$ that depends only on $3n-6$ angles, so that

$$\sum_{\lambda=1}^{n} G_{g\lambda}G_{g'\lambda}=\delta_{gg'} . \tag{32}$$

The set of the PA-hyperspherical coordinates is therefore

$$(q_1,\ldots,q_i,\ldots,q_{3N-6}) =(\rho,\theta,\phi,\theta_1,\ldots,\theta_\mu,\ldots,\theta_{3n-6}) .$$

Moreover, by construction, we have [cf. Eqs. (27) and (28)]

$$G_{g\lambda}(\theta_1,\ldots,\theta_{3N-6}) =\sum_{g'}\Phi_{gg'}(\theta_1,\theta_2,\theta_3)b_{g'\lambda}(\theta_4,\ldots,\theta_{3N-6}) , \tag{33a}$$

where $\Phi_{gg'}$ is given in Eq. (29) and $b_{g'\lambda}$ is an element of the three first lines of the orthogonal matrix

$$\mathbf{b}=\mathbf{g}_{(3)}\bar{\mathbf{g}}_{(4)}\cdots\bar{\mathbf{g}}_{(n-1)} , \tag{33b}$$

so that

$$\sum_{g''}\Phi_{gg''}\Phi_{g'g''}=\delta_{gg'},\quad \sum_{\lambda=1}^{n} b_{g\lambda}b_{g'\lambda}=\delta_{gg'} . \tag{34}$$

θ_1, θ_2, and θ_3 are *internal coordinates* and must definitely not be confused with α, β, and γ, the overall rotation angles of the system. Let us define ($i=4,\ldots,3N-6$):

$$\omega_{gg',i}(\theta_1,\ldots,\theta_{3n-6}) =\sum_{\lambda=1}^{n}\frac{\partial G_{g\lambda}(\theta_1,\ldots,\theta_{3n-6})}{\partial\theta_{i-3}}G_{g'\lambda}(\theta_1,\ldots,\theta_{3n-6}) , \tag{35a}$$

and, by virtue of Eq. (32),

$$\omega_{gg',i}=-\omega_{g'g,i} . \tag{35b}$$

Let us also put $\omega^*_{g,i}=\omega_{g'g'',i}$, where $(gg'g'')$ is a circular permutation of (xyz). Two cases must be distinguished. For $i=4,5,6$,

$$\omega_{gg',i}=\sum_{g''}\frac{\partial\Phi_{gg''}(\theta_1,\theta_2,\theta_3)}{\partial\theta_{i-3}}\Phi_{g'g''}(\theta_1,\theta_2,\theta_3) \tag{36a}$$

or, in matrix form

$$\begin{bmatrix} \omega^*_{x,4}\omega^*_{x,5}\omega^*_{x,6} \\ \omega^*_{y,4}\omega^*_{y,5}\omega^*_{y,6} \\ \omega^*_{z,4}\omega^*_{z,5}\omega^*_{z,6} \end{bmatrix} = \begin{bmatrix} 0 & \cos\theta_1 & \sin\theta_1\sin\theta_2 \\ 0 & -\sin\theta_1 & \cos\theta_1\sin\theta_2 \\ 1 & 0 & \cos\theta_2 \end{bmatrix} =\boldsymbol{\omega}^{*T}(\theta_1,\theta_2) . \tag{36b}$$

For $i=7,\ldots,3N-6$, if, by analogy with $\omega_{gg',i}$, we define

$$\gamma_{gg',i}(\theta_4,\ldots,\theta_{3n-6}) =\sum_{\lambda=1}^{n}\frac{\partial b_{g\lambda}(\theta_4,\ldots,\theta_{3n-6})}{\partial\theta_{i-3}}b_{g'\lambda}(\theta_4,\ldots,\theta_{3n-6}) ,$$
$$\gamma_{gg',i}=-\gamma_{g'g,i},\quad \gamma_{g,i}=\gamma_{g'g'',i} , \tag{37}$$

then one obtains

$$\omega^*_{g,i}(\theta_1,\ldots,\theta_{3n-6}) =\sum_{g'}\Phi_{gg'}(\theta_1,\theta_2,\theta_3)\gamma_{g'i}(\theta_4,\ldots,\theta_{3n-6}) , \tag{38a}$$

or, in matrix form,

$$\boldsymbol{\Phi}(\theta_1,\theta_2,\theta_3)\cdot\boldsymbol{\gamma}(\theta_4,\ldots,\theta_{3n-6}) = \begin{bmatrix} \omega^*_{x,7} & \omega^*_{x,8} & \cdots & \omega^*_{x,3N-6} \\ \omega^*_{y,7} & \omega^*_{y,8} & \cdots & \omega^*_{y,3N-6} \\ \omega^*_{z,7} & \omega^*_{z,8} & \cdots & \omega^*_{z,3N-6} \end{bmatrix} , \tag{38b}$$

where $\boldsymbol{\Phi}$ is the 3×3 Euler matrix in Eq. (29) and γ is the $3\times(3n-9)$ matrix whose elements are defined in Eq. (37).

We are now in a position to make **S** and **C** explicit. Let us first recall that [cf. Eq. (20c)]

$$\mathbf{I}=\begin{bmatrix} a_y^2+a_z^2 & 0 & 0 \\ 0 & a_z^2+a_x^2 & 0 \\ 0 & 0 & a_x^2+a_y^2 \end{bmatrix}=\begin{bmatrix} \rho^2(1-\sin^2\theta\cos^2\phi) & 0 & 0 \\ 0 & \rho^2(1-\sin^2\theta\sin^2\phi) & 0 \\ 0 & 0 & \rho^2\sin^2\theta \end{bmatrix} .$$

The definition of matrix **S** in Eq. (13) particularizes into the following. For $i,i'=1,2,3$,

$$S_{11}=S_{\rho\rho}=1,\quad S_{22}=S_{\theta\theta}=\rho^2,\quad S_{33}=S_{\phi\phi}=\rho^2\sin^2\theta,\quad S_{ii'\,(\neq i)}=0 .$$

For $i=1,2,3$ and $i'\geq 4$,

$$S_{ii'}=\sum_g a_g\frac{\partial a_g}{\partial q_i}\sum_{\lambda=1}^{n}G_{g\lambda}\frac{\partial G_{g\lambda}}{\partial q_{i'}}=0 .$$

For $i,i'=4,5,6$,

$$S_{ii'}=\sum_g\sum_{g'}a_g^2\omega_{gg',i}\omega_{gg',i'}=(a_y^2+a_z^2)\omega_{x,i}^*\omega_{x,i'}^*+(a_z^2+a_x^2)\omega_{y,i}^*\omega_{y,i'}^*+(a_x^2+a_y^2)\omega_{z,i}^*\omega_{z,i'}^* .$$

The corresponding block of matrix **S** can therefore be split into

$$\begin{bmatrix} \omega_{x,4}^*\omega_{y,4}^*\omega_{z,4}^* \\ \omega_{x,5}^*\omega_{y,5}^*\omega_{z,5}^* \\ \omega_{x,6}^*\omega_{y,6}^*\omega_{z,6}^* \end{bmatrix}\begin{bmatrix} a_y^2+a_z^2 & 0 & 0 \\ 0 & a_z^2+a_x^2 & 0 \\ 0 & 0 & a_x^2+a_y^2 \end{bmatrix}\begin{bmatrix} \omega_{x,4}^*\omega_{x,5}^*\omega_{x,6}^* \\ \omega_{y,4}^*\omega_{y,5}^*\omega_{y,6}^* \\ \omega_{z,4}^*\omega_{z,5}^*\omega_{z,6}^* \end{bmatrix}=\boldsymbol{\omega}^*\cdot\mathbf{I}\cdot\boldsymbol{\omega}^{*T} .$$

For $i,i'\geq 7$,

$$S_{ii'}=\sum_g a_g^2\sum_{g'}\sum_{g''}\Phi_{gg'}B_{g'g''}^{(ii')}\Phi_{gg''} ,$$

where

$$B_{g'g''}^{(ii')}=\sum_{\lambda=1}^{n}\frac{\partial b_{g'\lambda}}{\partial\theta_{i-3}}\frac{\partial b_{g''\lambda}}{\partial\theta_{i'-3}}$$

is a matrix element of a 3×3 matrix $\mathbf{B}^{(ii')}$. $S_{ii'}$ can be rewritten in a symmetric way,

$$\mathrm{tr}(\,\mathbf{A}\cdot\boldsymbol{\Phi}\cdot\mathbf{B}^{(ii')}\cdot\boldsymbol{\Phi}^T\cdot\mathbf{A})=Y_{i-6,i'-6} ,$$

where

$$\mathbf{A}=\begin{bmatrix} a_x & 0 & 0 \\ 0 & a_y & 0 \\ 0 & 0 & a_z \end{bmatrix} .$$

For $i=4,5,6$ and $i'\geq 7$,

$$S_{ii'}=\sum_{\lambda=1}^{n}\sum_g a_g^2\sum_{g'}\sum_{g''}\frac{\partial\Phi_{gg'}}{\partial\theta_{i-3}}\Phi_{gg''}\frac{\partial b_{g'\lambda}}{\partial\theta_{i'-3}}b_{g''\lambda} .$$

After some calculation, one obtains

$$S_{ii'}=-\sum_g\sum_{g'}\omega_{g,i}^*I_{gg}\Phi_{gg'}\gamma_{g'i'}$$

or, still, in matrix form for the corresponding block of **S**, $-\boldsymbol{\omega}^*\cdot\mathbf{I}\cdot\boldsymbol{\Phi}\cdot\boldsymbol{\gamma}$. Finally, according to the definition of the Coriolis matrix in Eq. (14),

$$C_{gi}=0 \quad \text{for } i=1,2,3 ,$$

$$C_{gi}=-2a_{g'}a_{g''}\omega_{g,i}^* \quad \text{for } i=4,5,6 ,$$

and

$$C_{gi}=-2a_{g'}a_{g''}\sum_{g'''}\Phi_{gg'''}\gamma_{g'''i} \quad \text{for } i\geq 7 ,$$

where $(gg'g'')$ is a circular permutation of (xyz).

With the help of the diagonal matrix **a** defined as

$$\mathbf{a}=\begin{bmatrix} -2a_ya_z & 0 & 0 \\ 0 & -2a_za_x & 0 \\ 0 & 0 & -2a_xa_y \end{bmatrix} ,$$

the complete matrix

$$\begin{bmatrix} \mathbf{S}(q) & \mathbf{C}^T(q) \\ \mathbf{C}(q) & \mathbf{I}(q) \end{bmatrix}$$

of Sec. II now reads, for the set of PA-hyperspherical coordinates introduced in Sec. III, as indicated in Fig. 4.

B. Lagrangian expression of the kinetic energy

The matrix in Fig. 4 can be readily split and inserted into the Lagrangian expression of the kinetic energy in terms of the PA-hyperspherical coordinates, as follows below,

$$2T=(\dot{\rho}\dot{\theta}\dot{\phi}\ \ \dot{\theta}_1\dot{\theta}_2\dot{\theta}_3\ \ \dot{\theta}_4\cdots\dot{\theta}_{3n-6}\ \ \omega_x\omega_y\omega_z)\cdot\begin{pmatrix}1 & 0 & 0 & 0\\ 0 & \boldsymbol{\omega}^* & 0 & 0\\ 0 & \boldsymbol{\gamma}^T\cdot\boldsymbol{\Phi}^T & 1 & 0\\ 0 & 0 & 0 & 1\end{pmatrix}\cdot\begin{pmatrix}\begin{matrix}1 & 0 & 0\\ 1 & 0 & 0\\ 0 & 0 & \rho^2\sin^2\theta\end{matrix} & \mathbf{0} & \mathbf{0} & \mathbf{0}\\ \mathbf{0} & \mathbf{I} & \mathbf{z}^T & \mathbf{a}\\ \mathbf{0} & \mathbf{z} & \mathbf{y} & \mathbf{0}\\ \mathbf{0} & \mathbf{a} & \mathbf{0} & \mathbf{I}\end{pmatrix}\cdot\begin{pmatrix}1 & 0 & 0 & 0\\ 0 & \boldsymbol{\omega}^{*T} & \boldsymbol{\Phi}\cdot\boldsymbol{\gamma} & 0\\ 0 & 0 & 1 & 0\\ 0 & 0 & 0 & 1\end{pmatrix}\cdot\begin{pmatrix}\dot{\rho}\\ \dot{\theta}\\ \dot{\phi}\\ \dot{\theta}_1\\ \dot{\theta}_2\\ \dot{\theta}_3\\ \dot{\theta}_4\\ \vdots\\ \dot{\theta}_{3n-6}\\ \omega_x\\ \omega y\\ \omega_z\end{pmatrix}, \tag{39}$$

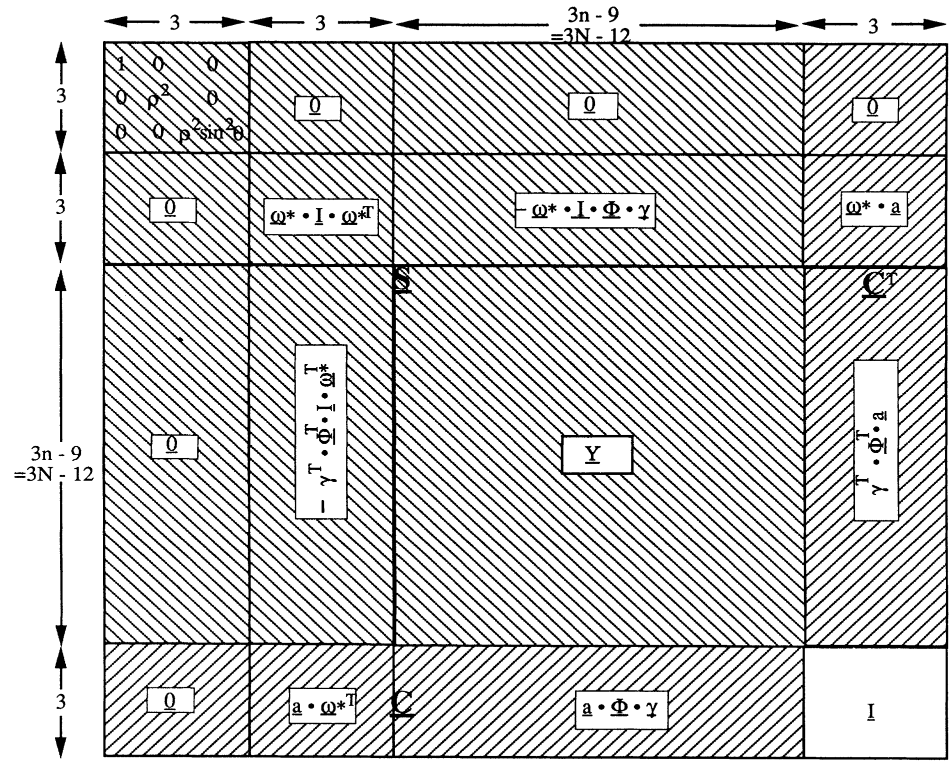

FIG. 4. Algebraic structure (defined block by block) of the symmetric matrix, defined in Eq. (9), for the set of the PA-hyperspherical coordinates. This matrix appears in the Lagrangian expression of the kinetic energy of the system in terms of the $3N-6=3n-3$ internal velocities and the three MS components of the angular velocity vector. The row and column indices of the matrix correspond to the individual coordinates, namely (i) three spherical coordinates to parametrize the three gyration radii of the system, ρ, θ, and ϕ; (ii) three Eulerian angles θ_1, θ_2, and θ_3 that are actually internal coordinates and account for the internal deformations; (iii) $3n-9$ additional generalized Eulerian angles, θ_4 to θ_{3n-6}, appearing in the expression of matrix $\bar{\mathbf{g}}$ that are also internal coordinates; and finally (iv) the three directions x, y, and z of the MS frame of reference.

where $\mathbf{y}=\mathbf{Y}+3\boldsymbol{\gamma}^T\cdot\boldsymbol{\Phi}^T\cdot\mathbf{I}\cdot\boldsymbol{\Phi}\cdot\boldsymbol{\gamma}$ and $\mathbf{z}=-2\mathbf{I}\cdot\boldsymbol{\Phi}\cdot\boldsymbol{\gamma}$. The structure of Eq. (39) strongly suggests that one introduce three quasivelocities, namely,

$$\boldsymbol{\Omega}=\begin{bmatrix}\Omega_x\\ \Omega_y\\ \Omega_z\end{bmatrix}=\boldsymbol{\omega}^{*T}(\theta_1,\theta_2)\cdot\begin{bmatrix}\dot\theta_1\\ \dot\theta_2\\ \dot\theta_3\end{bmatrix}+\boldsymbol{\Phi}(\theta_1,\theta_2,\theta_3)\cdot\boldsymbol{\gamma}(\theta_4\cdots\theta_{3n-6})\cdot\begin{bmatrix}\dot\theta_4\\ \vdots\\ \dot\theta_{3n-6}\end{bmatrix}, \tag{40}$$

so that the kinetic energy now reads

$$2T=(\dot\rho\dot\theta\dot\phi\;\;\Omega_x\Omega_y\Omega_z\;\;\dot\theta_4\cdots\dot\theta_{3n-6}\;\;\omega_x\omega_y\omega_x)\times\begin{bmatrix}\begin{matrix}1&0&0\\0&\rho^2&0\\0&0&\rho^2\sin^2\theta\end{matrix}&\mathbf{0}&\mathbf{0}&\mathbf{0}\\ \mathbf{0}&\mathbf{I}&\mathbf{z}^T&\mathbf{a}\\ \mathbf{0}&\mathbf{z}&\mathbf{y}&\mathbf{0}\\ \mathbf{0}&\mathbf{a}&\mathbf{0}&\mathbf{I}\end{bmatrix}\cdot\begin{bmatrix}\dot\rho\\ \dot\theta\\ \dot\phi\\ \Omega_x\\ \Omega_y\\ \Omega_z\\ \dot\theta_4\\ \vdots\\ \theta_{3n-6}\\ \omega_x\\ \omega_y\\ \omega_z\end{bmatrix}. \tag{41}$$

Here, the quasivelocities are introduced to facilitate the transformation of the Lagrangian into the Hamiltonian expression of the kinetic energy. Indeed (see Sec. II), all matrices appearing in Eq. (39) must be inverted, and the one defining the quasivelocities is trivially inverted, namely,

$$\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \boldsymbol{\omega}^{*T} & \boldsymbol{\Phi}\cdot\boldsymbol{\gamma} & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}^{-1} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \boldsymbol{\omega}^{*T-1} & -\boldsymbol{\omega}^{*T-1}\cdot\boldsymbol{\Phi}\cdot\boldsymbol{\gamma} & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}.$$

In this context Eckart's brightest original idea appears. Indeed, Eckart thinks of the orthogonal matrix $\bar{\mathbf{g}}$ as being defined from the Jacobi vectors; as such, *all* its elements (and not only those of the three first lines) can be invoked to define quasivelocities. In particular, if the following notation is used (see above, Sec. III):

$$\bar{\mathbf{g}} = \begin{bmatrix} [G_{g\lambda}] \\ \\ [b_{f\lambda}] \end{bmatrix} \begin{matrix} x \\ y \\ z \\ 4 \\ \vdots \\ f \\ \vdots \\ n \end{matrix}, \quad \text{columns } 1\ 2 \cdots \lambda \cdots n \tag{42}$$

where $G_{g\lambda}$ and $\mathbf{b}$ are defined in Eqs. (33); the additional quasivelocities introduced by Eckart are

$$\omega_{gf} = \sum_{\lambda=1}^{n} \dot{G}_{g\lambda} b_{f\lambda} = \sum_{\mu=1}^{3n-6} \left[\sum_{\lambda=1}^{n} \frac{\partial G_{g\lambda}}{\partial \theta_\mu} b_{f\lambda} \right] \dot{\theta}_\mu$$

$$= \sum_{\mu=4}^{3n-6} \left[\sum_{g'} \Phi_{gg'} \sum_{\lambda=1}^{n} \frac{\partial b_{g'\lambda}}{\partial \theta_\mu} b_{f\lambda} \right] \dot{\theta}_\mu$$

$$(g = x,y,z;\ f = 4, \ldots, n) \tag{43}$$

by analogy with the possible expression of Ω_g $(g = x,y,z)$:

$$\Omega_g = \sum_{\lambda=1}^{n} \dot{G}_{g'\lambda} G_{g''\lambda} = \sum_{\mu=1}^{3n-6} \left[\sum_{\lambda=1}^{n} \frac{\partial G_{g'\lambda}}{\partial \theta_\mu} G_{g''\lambda} \right] \dot{\theta}_\mu$$

$$= \sum_{\mu=1}^{3} \omega^*_{g,\mu+3} \dot{\theta}_\mu + \sum_{\mu=4}^{3n-6} \left[\sum_{g'} \Phi_{gg'} \gamma_{g'\mu} \right] \dot{\theta}_\mu \tag{44}$$

$(gg'g'')$ being a circular permutation of (xyz).

With the help of these quasivelocities, the expression of the kinetic energy takes the remarkably simple form of Eq. (45) on the following page.

In matrix form, the transformation from the internal angular velocities $\dot{\theta}_\mu$ to the angular quasivelocities is

$$\begin{bmatrix} \Omega_x \\ \Omega_y \\ \Omega_z \\ \omega_{x4} \\ \vdots \\ \omega_{zn} \end{bmatrix} = \begin{bmatrix} \boldsymbol{\omega}^{*T}(\theta_1,\theta_2) & \boldsymbol{\Phi}(\theta_1,\theta_2,\theta_3)\cdot\boldsymbol{\gamma}(\theta_4 \cdots \theta_{3n-6}) \\ 0 & \mathbf{D}^n \end{bmatrix} \cdot \begin{bmatrix} \dot{\theta}_1 \\ \dot{\theta}_2 \\ \dot{\theta}_3 \\ \dot{\theta}_4 \\ \vdots \\ \dot{\theta}_{3n-6} \end{bmatrix} \tag{46}$$

and, formally, the inverse of the matrix in Eq. (46) is

$$\begin{bmatrix} \boldsymbol{\omega}^{*T} & \boldsymbol{\Phi}\cdot\boldsymbol{\gamma} \\ 0 & \mathbf{D}^n \end{bmatrix}^{-1} = \begin{bmatrix} \boldsymbol{\omega}^{*T-1} & -\boldsymbol{\omega}^{*T-1}\cdot\boldsymbol{\Phi}\cdot\boldsymbol{\gamma}\cdot(\mathbf{D}^n)^{-1} \\ 0 & (\mathbf{D}^n)^{-1} \end{bmatrix}. \tag{47}$$

At this point, Eckart obviously suggests that one should canonically associate quasimomenta with the quasivelocities of Eq. (46), in order to preserve, in the Hamiltonian context, the wonderful simplicity of Eq. (45). All the problem thus amounts to is the question, "Does the $(3n-9)$-dimensional matrix $\mathbf{D}^n$ always have an inverse?" The answer is "yes."

C. Inversion of D^n

According to Eq. (43), a matrix element of $\mathbf{D}^n$ is by construction,

$$D^n_{gf,\mu} = \sum_{g'} \Phi_{gg'} \sum_{\lambda=1}^{n} \frac{\partial b_{g'\lambda}}{\partial \theta_\mu} b_{f\lambda}, \tag{48}$$

where $g = x,y,z$; $f = 4, \ldots, n$; and $\mu = 4, \ldots, 3n-6$.

Let us introduce $n-3$ auxiliary $3\times(3n-9)$ matrices $\boldsymbol{\delta}^f$ $(f = 4,n)$, of element

$$2T=(\dot\rho\dot\theta\dot\phi\ \ \Omega_x\Omega_y\Omega_z\ \ \omega_{x4}\cdots\omega_{xn}\ \ \omega_{y4}\cdots\omega_{yn}\ \ \omega_{z4}\cdots\omega_{zn}\ \ \omega_x\omega_y\omega_z)\cdot\left(\begin{array}{ccc|c|ccccccccc|c}
\begin{array}{ccc}1&0&0\\0&\rho^2&0\\0&0&\rho^2\sin^2\theta\end{array} & & & \mathbf{0} & & & & & \mathbf{0} & & & & & \mathbf{0}\\
\hline
\mathbf{0} & & & \mathbf{I} & & & & & \mathbf{0} & & & & & \mathbf{a}\\
\hline
 & & & & a_x^2 & & & & & & & & & \\
 & & & & & \ddots & & & & & 0 & & & \\
 & & & & & & a_x^2 & & & & & & & \\
 & & & & & & & a_y^2 & & & & & & \\
\mathbf{0} & & & \mathbf{0} & & & & & \ddots & & & & & \mathbf{0}\\
 & & & & & & & & & a_y^2 & & & & \\
 & & & & & & & & & & a_z^2 & & & \\
 & & & & & 0 & & & & & & \ddots & & \\
 & & & & & & & & & & & & a_z^2 & \\
\hline
\mathbf{0} & & & \mathbf{a} & & & & & \mathbf{0} & & & & & \mathbf{I}
\end{array}\right)\left(\begin{array}{c}\dot\rho\\ \dot\theta\\ \dot\phi\\ \Omega_x\\ \Omega_y\\ \Omega_z\\ \omega_{x4}\\ \vdots\\ \omega_{xn}\\ \omega_{y4}\\ \vdots\\ \omega_{yn}\\ \omega_{z4}\\ \vdots\\ \omega_{zn}\\ \omega_x\\ \omega_y\\ \omega_z\end{array}\right). \tag{45}$$

$$\delta^f_{g\mu}=\sum_{\lambda=1}^{n}\frac{\partial b_{g\lambda}}{\partial\theta_\mu}b_{f\lambda}=-\sum_{\lambda=1}^{n}b_{g\lambda}\frac{\partial b_{f\lambda}}{\partial\theta_\mu}\ . \tag{49}$$

Matrix $\mathbf{D}^n$ is written as

$$\mathbf{D}^n=\begin{bmatrix}\mathbf{\Phi}\cdot\boldsymbol{\delta}^4\\ \vdots\\ \mathbf{\Phi}\cdot\boldsymbol{\delta}^n\end{bmatrix}=\begin{bmatrix}\mathbf{\Phi} & & \mathbf{0}\\ & \ddots & \\ \mathbf{0} & & \mathbf{\Phi}\end{bmatrix}\cdot\begin{bmatrix}\boldsymbol{\delta}^4\\ \vdots\\ \boldsymbol{\delta}^n\end{bmatrix}\ . \tag{50}$$

We denote by $\mathbf{\Delta}^n$ the last matrix on the right in Eq. (50). In the same way as $\mathbf{D}^n$, $\mathbf{\Delta}^n$ is a $(3n-9)$-dimensional square matrix. Therefore

$$(\mathbf{D}^n)^{-1}=(\mathbf{\Delta}^n)^{-1}\cdot\begin{bmatrix}\mathbf{\Phi}^t & & \mathbf{0}\\ & \ddots & \\ \mathbf{0} & & \mathbf{\Phi}^t\end{bmatrix}, \tag{51}$$

and all the difficulty of the inversion of $\mathbf{D}^n$ is transferred to $\mathbf{\Delta}^n$.

1. Algebraic properties of $\mathbf{\Delta}^n$

Let us first show by recursion that $\mathbf{\Delta}^n$ and $\mathbf{D}^n$ are block upper triangular matrices, namely,

$$\mathbf{\Delta}^n=\begin{bmatrix}\mathbf{a}_{44} & \mathbf{a}_{45} & \cdots & \mathbf{a}_{4n}\\ \mathbf{0} & \mathbf{a}_{55} & \cdots & \mathbf{a}_{5n}\\ \vdots & & \ddots & \vdots\\ \mathbf{0} & \mathbf{0} & \cdots & \mathbf{a}_{nn}\end{bmatrix}, \tag{52a}$$

$$\mathbf{D}^n=\begin{bmatrix}\mathbf{\Phi}\cdot\mathbf{a}_{44} & \mathbf{\Phi}\cdot\mathbf{a}_{45} & \cdots & \mathbf{\Phi}\cdot\mathbf{a}_{4n}\\ \mathbf{0} & \mathbf{\Phi}\cdot\mathbf{a}_{55} & \cdots & \mathbf{\Phi}\cdot\mathbf{a}_{5n}\\ \vdots & & \ddots & \vdots\\ \mathbf{0} & \mathbf{0} & \cdots & \mathbf{\Phi}\cdot\mathbf{a}_{nn}\end{bmatrix}, \tag{52b}$$

where the current block $\mathbf{a}_{fn}$ is a 3×3 matrix. This property will turn out to be very important below (see Sec. IV C 2)

For five particles, $n=4$ and $\mathbf{b}_4=\bar{\mathbf{g}}_{(3)}=\mathbf{g}_{(3)}$,

$$\mathbf{b}_4=\begin{bmatrix}\cos\theta_6 & -\sin\theta_6 & 0 & 0\\ \cos\theta_5\sin\theta_6 & \cos\theta_5\cos\theta_6 & -\sin\theta_5 & 0\\ \cos\theta_4\sin\theta_5\sin\theta_6 & \cos\theta_4\sin\theta_5\cos\theta_6 & \cos\theta_4\cos\theta_5 & -\sin\theta_4\\ \sin\theta_4\sin\theta_5\sin\theta_6 & \sin\theta_4\sin\theta_5\cos\theta_6 & \sin\theta_4\cos\theta_5 & \cos\theta_4\end{bmatrix},$$

where the indices of the rows and columns are, by construction, respectively, x, y, z (g-type), and 4 (f-type) for the rows; and 1, 2, 3, and 4 (λ-type) for the columns. In addition, $\mathbf{D}^4=\mathbf{\Phi}\cdot\mathbf{\Delta}^4=\mathbf{\Phi}\cdot\mathbf{a}_{44}$ is a 3×3 matrix, where

$$\mathbf{\Delta}^4=\mathbf{a}_{44}=\begin{bmatrix}0 & 0 & -\sin\theta_4\sin\theta_5\\ 0 & -\sin\theta_4 & 0\\ -1 & 0 & 0\end{bmatrix}.$$

For six particles, $n=5$, $\mathbf{b}_5=\bar{\mathbf{g}}_{(3)}\cdot\bar{\mathbf{g}}_{(4)}$,

$$\mathbf{b}_5=\begin{bmatrix}\cos\theta_6 & -\sin\theta_6 & 0 & 0 & 0\\ \cos\theta_5\sin\theta_6 & \cos\theta_5\cos\theta_6 & -\sin\theta_5 & 0 & 0\\ \cos\theta_4\sin\theta_5\sin\theta_6 & \cos\theta_4\sin\theta_5\cos\theta_6 & \cos\theta_4\cos\theta_5 & -\sin\theta_4 & 0\\ \sin\theta_4\sin\theta_5\sin\theta_6 & \sin\theta_4\sin\theta_5\cos\theta_6 & \sin\theta_4\cos\theta_5 & \cos\theta_4 & 0\\ 0 & 0 & 0 & 0 & 1\end{bmatrix}$$

$$\cdot\begin{bmatrix}1 & 0 & 0 & 0 & 0\\ 0 & \cos\theta_9 & -\sin\theta_9 & 0 & 0\\ 0 & \cos\theta_8\sin\theta_9 & \cos\theta_8\cos\theta_9 & -\sin\theta_8 & 0\\ 0 & \cos\theta_7\sin\theta_8\sin\theta_9 & \cos\theta_7\sin\theta_8\cos\theta_9 & \cos\theta_7\cos\theta_8 & -\sin\theta_7\\ 0 & \sin\theta_7\sin\theta_8\sin\theta_9 & \sin\theta_7\sin\theta_8\cos\theta_9 & \sin\theta_7\cos\theta_8 & \cos\theta_7\end{bmatrix},$$

whence

$$\boldsymbol{\Delta}^5=\begin{bmatrix}\mathbf{a}_{44} & \mathbf{a}_{45}\\ \mathbf{0} & \mathbf{a}_{55}\end{bmatrix}=\begin{pmatrix}0 & 0 & -\sin\theta_4\sin\theta_5 & 0 & 0 & \sin\theta_6(\sin\theta_4\cos\theta_5\cos\theta_8+\cos\theta_4\cos\theta_7\sin\theta_8)\\ 0 & -\sin\theta_4 & 0 & 0 & \cos\theta_4\sin\theta_5\cos\theta_7 & -\cos\theta_6(\sin\theta_4\cos\theta_8+\cos\theta_4\cos\theta_5\cos\theta_7\sin\theta_8)\\ -1 & 0 & 0 & 0 & -\cos\theta_5\cos\theta_7 & -\sin\theta_5\cos\theta_6\cos\theta_7\sin\theta_8\\ 0 & 0 & 0 & 0 & 0 & \sin\theta_6\sin\theta_7\sin\theta_8\\ 0 & 0 & 0 & 0 & \sin\theta_5\sin\theta_7 & -\cos\theta_5\cos\theta_6\sin\theta_7\sin\theta_8\\ 0 & 0 & 0 & \sin\theta_4 & -\cos\theta_4\cos\theta_5\sin\theta_7 & -\cos\theta_4\sin\theta_5\cos\theta_6\times\sin\theta_7\sin\theta_8\end{pmatrix} .$$

The following points are noticeable: (i) $\boldsymbol{\Delta}^5$ is a block upper triangular matrix; (ii) $\boldsymbol{\Delta}^4=\mathbf{a}_{44}$ remains unchanged in the upper left corner of $\boldsymbol{\Delta}^5$; (iii)

$$\mathbf{a}_{55}=\begin{bmatrix}-\sin\theta_6 & 0 & 0\\ \cos\theta_5\cos\theta_6 & -\sin\theta_5 & 0\\ \cos\theta_4\sin\theta_5\cos\theta_6 & \cos\theta_4\cos\theta_5 & -\sin\theta_4\end{bmatrix}\cdot\begin{bmatrix}0 & 0 & -\sin\theta_7\sin\theta_8\\ 0 & -\sin\theta_7 & 0\\ -1 & 0 & 0\end{bmatrix};$$

(iv)

$$\mathbf{a}_{45}=\begin{bmatrix}\sin\theta_4\cos\theta_5\sin\theta_6 & 0 & \cos\theta_4\sin\theta_6\\ -\sin\theta_4\cos\theta_6 & \cos\theta_4\sin\theta_5 & -\cos\theta_4\cos\theta_5\cos\theta_6\\ 0 & -\cos\theta_5 & -\sin\theta_5\cos\theta_6\end{bmatrix}\cdot\begin{bmatrix}0 & 0 & \cos\theta_8\\ 0 & \cos\theta_7 & 0\\ 0 & 0 & \cos\theta_7\sin\theta_8\end{bmatrix} .$$

In the general case, $n\geq 5$, $\mathbf{b}_n=\overline{\mathbf{g}}_{(3)}\cdot\ \cdots\ \cdot\overline{\mathbf{g}}_{(n-2)}\cdot\overline{\mathbf{g}}_{(n-1)}=\mathbf{b}_{n-1}\cdot\overline{\mathbf{g}}_{(n-1)}$ is, in matrix form,

$$\mathbf{b}_n=\underbrace{\begin{bmatrix}\mathbf{b}_{n-1} & \mathbf{0}\\ \mathbf{0} & 1\end{bmatrix}}_{1\ \cdots\ n-1\quad n}\cdot\underbrace{\begin{bmatrix}1 & \mathbf{0}\\ \mathbf{0} & \overline{\mathbf{g}}_{(n-1)}\end{bmatrix}}_{1\ \cdots\ n-4\quad n-3\ldots n} \tag{53a}$$

or, still, with the indices appropriate for row (x,y,z, g type; and 4 to n, f type), columns (1 to n, λ type), and internal Eulerian angles (4 to $3n-6$), the matrix given in Eq. (53b) on the following page.

From Eq. (53b), on the following page, at the expense of long and intricate calculations not presented here, the three following properties are demonstrated.

(i) In $\boldsymbol{\delta}^f$, the $3\times(3n-9)$ block of $\boldsymbol{\Delta}^n$, if $f\neq n$, i.e., if $4\leq f\leq n-1$, since by construction,

$$\delta^f_{g\mu}=\sum_{\lambda=1}^{n}\frac{\partial b_n^{g,\lambda}}{\partial\theta_\mu}b_n^{f,\lambda} ,$$

if $\mu<3n-8$, i.e., for all blocks $\mathbf{a}_{fm}$ but the last one on the right ($m<n$), it is easy to check that

$$\delta^f_{x\mu}=\sum_{\lambda=1}^{n-4}\frac{\partial b_{n-1}^{x,\lambda}}{\partial\theta_\mu}b_{n-1}^{f,\lambda}+\frac{\partial b_{n-1}^{x,n-3}}{\partial\theta_\mu}b_{n-1}^{f,n-3}=\sum_{\lambda=1}^{n-1}\frac{\partial b_{n-1}^{x,\lambda}}{\partial\theta_\mu}b_{n-1}^{f,\lambda} .$$

The second equality holds because $b_{n-1}^{x,n-2}=b_{n-1}^{x,n-1}=0$. Similarly,

$$\delta^f_{y\mu}=\sum_{\lambda=1}^{n-1}\frac{\partial b_{n-1}^{y,\lambda}}{\partial\theta_\mu}b_{n-1}^{f,\lambda},\quad \delta^f_{z\mu}=\sum_{\lambda=1}^{n-1}\frac{\partial b_{n-1}^{z,\lambda}}{\partial\theta_\mu}b_{n-1}^{f,\lambda} .$$

$$
\begin{array}{c}
\left(
\begin{array}{ccccc}
b_{n-1}^{x,\lambda} & b_{n-1}^{x,n-3}\cos\theta_{3n-6} & -b_{n-1}^{x,n-3}\sin\theta_{3n-6} & 0 & 0 \\
b_{n-1}^{y,\lambda} & (b_{n-1}^{y,n-3}\cos\theta_{3n-6} & (-b_{n-1}^{y,n-3}\sin\theta_{3n-6} & -b_{n-1}^{y,n-2}\sin\theta_{3n-7} & 0 \\
 & +b_{n-1}^{y,n-2}\cos\theta_{3n-7}\sin\theta_{3n-6}) & +b_{n-1}^{y,n-2}\cos\theta_{3n-7}\cos\theta_{3n-6}) & & \\
b_{n-1}^{z,\lambda} & (b_{n-1}^{z,n-3}\cos\theta_{3n-6} & (-b_{n-1}^{z,n-3}\sin\theta_{3n-6} & (-b_{n-1}^{z,n-2}\sin\theta_{3n-7} & -b_{n-1}^{z,n-1}\sin\theta_{3n-8} \\
 & +b_{n-1}^{z,n-2}\cos\theta_{3n-7}\sin\theta_{3n-6} & +b_{n-1}^{z,n-2}\cos\theta_{3n-7}\cos\theta_{3n-6} & +b_{n-1}^{z,n-1}\cos\theta_{3n-8}\cos\theta_{3n-7}) & \\
 & +b_{n-1}^{z,n-1}\cos\theta_{3n-8}\sin\theta_{3n-7}\sin\theta_{3n-6}) & +b_{n-1}^{z,n-1}\cos\theta_{3n-8}\sin\theta_{3n-7}\cos\theta_{3n-6}) & & \\
b_{n-1}^{f,\lambda} & (b_{n-1}^{f,n-3}\cos\theta_{3n-6} & (-b_{n-1}^{f,n-3}\sin\theta_{3n-6} & (-b_{n-1}^{f,n-2}\sin\theta_{3n-7} & -b_{n-1}^{f,n-1}\sin\theta_{3n-8} \\
 & +b_{n-1}^{f,n-2}\cos\theta_{3n-7}\sin\theta_{3n-6} & +b_{n-1}^{f,n-2}\cos\theta_{3n-7}\cos\theta_{3n-6} & +b_{n-1}^{f,n-1}\cos\theta_{3n-8}\cos\theta_{3n-7}) & \\
 & +b_{n-1}^{f,n-1}\cos\theta_{3n-8}\sin\theta_{3n-7}\sin\theta_{3n-6}) & +b_{n-1}^{f,n-1}\cos\theta_{3n-8}\sin\theta_{3n-7}\cos\theta_{3n-6}) & & \\
0 & \sin\theta_{3n-8}\sin\theta_{3n-7}\sin\theta_{3n-6} & \sin\theta_{3n-8}\sin\theta_{3n-7}\cos\theta_{3n-6} & \sin\theta_{3n-8}\cos\theta_{3n-7} & \cos\theta_{3n-8}
\end{array}
\right)
\begin{array}{c}
x \\ y \\ \\ z \\ \\ \\ 4 \\ \vdots \\ n-1 \\ n
\end{array}
\\
\begin{array}{ccccc}
1\ \cdots\ n-4 & n-3 & n-2 & n-1 & n
\end{array}
\end{array}
\qquad (53b)
$$

In other words, apart from the last 3×3 block on the right, $\mathbf{a}_{fn}$, the block $\boldsymbol{\delta}^f$ of $\boldsymbol{\Delta}^n$ is the same as block $\boldsymbol{\delta}^f$ of $\boldsymbol{\Delta}^{n-1}$; or still $\boldsymbol{\Delta}^{n-1}$, as a block, remains unchanged in the upper left corner of $\boldsymbol{\Delta}^n$.

(ii) In $\boldsymbol{\Delta}^n$,

$$\boldsymbol{\delta}^n=(\mathbf{0}\ \cdots\ \mathbf{0}\ \mathbf{a}_{nn})$$

is the last $3\times(3n-9)$ submatrix, in which each indicated block is a 3×3 matrix, i.e., $\boldsymbol{\Delta}^n$ actually is an upper triangular block matrix. Moreover,

$$\mathbf{a}_{nn}=\begin{bmatrix} b_{n-1}^{x,n-3} & 0 & 0 \\ b_{n-1}^{y,n-3} & b_{n-1}^{y,n-2} & 0 \\ b_{n-1}^{z,n-3} & b_{n-1}^{z,n-2} & b_{n-1}^{z,n-1} \end{bmatrix}\cdot\begin{bmatrix} 0 & 0 & -\sin\theta_{3n-8}\sin\theta_{3n-7} \\ 0 & -\sin\theta_{3n-8} & 0 \\ -1 & 0 & 0 \end{bmatrix}$$

is recursively identified with

$$\mathbf{a}_{nn}=\begin{bmatrix} -\sin\theta_6 & 0 & 0 \\ \cos\theta_5\cos\theta_6 & -\sin\theta_5 & 0 \\ \cos\theta_4\sin\theta_5\cos\theta_6 & \cos\theta_4\cos\theta_5 & -\sin\theta_4 \end{bmatrix}$$

$$\cdots\cdots\begin{bmatrix} -\sin\theta_{3n-9} & 0 & 0 \\ \cos\theta_{3n-10}\cos\theta_{3n-9} & -\sin\theta_{3n-10} & 0 \\ \cos\theta_{3n-11}\sin\theta_{3n-10}\cos\theta_{3n-9} & \cos\theta_{3n-11}\cos\theta_{3n-10} & -\sin\theta_{3n-11} \end{bmatrix}$$

$$\cdot\begin{bmatrix} 0 & 0 & -\sin\theta_{3n-8}\sin\theta_{3n-7} \\ 0 & -\sin\theta_{3n-8} & 0 \\ -1 & 0 & 0 \end{bmatrix}, \tag{54a}$$

so that

$$\mathbf{a}_{nn}^{-1}=(-1)^{n+1}\begin{bmatrix} 0 & 0 & 1 \\ 0 & \dfrac{1}{\sin\theta_{3n-8}} & 0 \\ \dfrac{1}{\sin\theta_{3n-8}\sin\theta_{3n-7}} & 0 & 0 \end{bmatrix}\cdot\begin{bmatrix} \dfrac{1}{\sin\theta_{3n-9}} & 0 & 0 \\ \dfrac{\cos\theta_{3n-10}\cos\theta_{3n-9}}{\sin\theta_{3n-10}\sin\theta_{3n-9}} & \dfrac{1}{\sin\theta_{3n-10}} & 0 \\ \dfrac{\cos\theta_{3n-11}\cos\theta_{3n-9}}{\sin\theta_{3n-11}\sin\theta_{3n-10}\sin\theta_{3n-9}} & \dfrac{\cos\theta_{3n-11}\cos\theta_{3n-10}}{\sin\theta_{3n-11}\sin\theta_{3n-10}} & \dfrac{1}{\sin\theta_{3n-11}} \end{bmatrix}$$

$$\cdots\cdots\begin{bmatrix} \dfrac{1}{\sin\theta_6} & 0 & 0 \\ \dfrac{\cos\theta_5\cos\theta_6}{\sin\theta_5\sin\theta_6} & \dfrac{1}{\sin\theta_5} & 0 \\ \dfrac{\cos\theta_4\cos\theta_6}{\sin\theta_4\sin\theta_5\sin\theta_6} & \dfrac{\cos\theta_4\cos\theta_5}{\sin\theta_4\sin\theta_5} & \dfrac{1}{\sin\theta_4} \end{bmatrix}. \tag{54b}$$

always exists.

(iii) For $\mu=3n-8$, $3n-7$, and $3n-6$, i.e., for the last 3×3 blocks in the right of $\boldsymbol{\Delta}^n$, we have $(4\le f\le n-1)$:

$$\mathbf{a}_{fn}=\begin{bmatrix} -b_{n-1}^{x,n-3}b_{n-1}^{f,n-2} & 0 & -b_{n-1}^{x,n-3}b_{n-1}^{f,n-1} \\ -b_{n-1}^{y,n-3}b_{n-1}^{f,n-2}+b_{n-1}^{y,n-2}b_{n-1}^{f,n-3} & -b_{n-1}^{y,n-2}b_{n-1}^{f,n-1} & -b_{n-1}^{y,n-3}b_{n-1}^{f,n-1} \\ -b_{n-1}^{z,n-3}b_{n-1}^{f,n-2}+b_{n-1}^{z,n-2}b_{n-1}^{f,n-3} & -b_{n-1}^{z,n-2}b_{n-1}^{f,n-1}+b_{n-1}^{z,n-2}b_{n-1}^{f,n-1} & -b_{n-1}^{z,n-3}b_{n-1}^{f,n-1}+b_{n-1}^{z,n-1}b_{n-1}^{f,n-3} \end{bmatrix}\cdot\begin{bmatrix} 0 & 0 & \cos\theta_{3n-7} \\ 0 & \cos\theta_{3n-8} & 0 \\ 0 & 0 & \cos\theta_{3n-8}\sin\theta_{3n-7} \end{bmatrix} \tag{55a}$$

$\mathbf{a}_{fn}$ can be put in the explicit form in Eq. (55b) below:

$$\mathbf{a}_{fn}(f\le n)=\begin{bmatrix} -\sin\theta_6 & 0 & 0 \\ \cos\theta_5\cos\theta_6 & -\sin\theta_5 & 0 \\ \cos\theta_4\sin\theta_5\cos\theta_6 & \cos\theta_4\cos\theta_5 & -\sin\theta_4 \end{bmatrix}\cdots\cdots\begin{bmatrix} -\sin\theta_{3f-9} & 0 & 0 \\ \cos\theta_{3f-10}\cos\theta_{3f-9} & -\sin\theta_{3f-10} & 0 \\ \cos\theta_{3f-11}\sin\theta_{3f-10}\cos\theta_{3f-9} & \cos\theta_{3f-11}\cos\theta_{3f-10} & -\sin\theta_{3f-11} \end{bmatrix}$$

$$\cdot\begin{bmatrix} -\sin\theta_{3f-8}\cos\theta_{3f-7}\sin\theta_{3f-6} & 0 & \cos\theta_{3f-8}\sin\theta_{3f-6} \\ -\sin\theta_{3f-8}\cos\theta_{3f-6} & \cos\theta_{3f-8}\sin\theta_{3f-7} & -\cos\theta_{3f-8}\cos\theta_{3f-7}\cos\theta_{3f-6} \\ 0 & -\cos\theta_{3f-7} & -\sin\theta_{3f-7}\cos\theta_{3f-6} \end{bmatrix}$$

$$\cdot\begin{bmatrix} \sin\theta_{3f-4}\sin\theta_{3f-3} & 0 & 0 \\ \cos\theta_{3f-5}\cos\theta_{3f-3} & \sin\theta_{3f-5}\sin\theta_{3f-4} & -\sin\theta_{3f-5}\cos\theta_{3f-4}\cos\theta_{3f-3} \\ -\cos\theta_{3f-5}\cos\theta_{3f-4}\sin\theta_{3f-3} & 0 & \sin\theta_{3f-5}\sin\theta_{3f-3} \end{bmatrix}$$

$$\cdots\cdots\begin{bmatrix} \sin\theta_{3n-10}\sin\theta_{3n-9} & 0 & 0 \\ \cos\theta_{3n-11}\cos\theta_{3n-9} & \sin\theta_{3n-11}\sin\theta_{3n-10} & -\sin\theta_{3n-11}\cos\theta_{3n-10}\cos\theta_{3n-9} \\ -\cos\theta_{3n-11}\cos\theta_{3n-10}\sin\theta_{3n-9} & 0 & \sin\theta_{3n-11}\sin\theta_{3n-9} \end{bmatrix}\cdot\begin{bmatrix} 0 & 0 & \cos\theta_{3n-7} \\ 0 & \cos\theta_{3n-8} & 0 \\ 0 & 0 & \cos\theta_{3n-8}\sin\theta_{3n-7} \end{bmatrix}. \tag{55b}$$

This form holds whatever $n \geq 5$ and $4 \leq f \leq n-1$, provided that the formula is limited as indicated below if $f=4$ or $f=n-1$.

Indeed, six 3×3 matrices are explicitly displayed in Eq. (55b). It should be noted that there are two implicit series products between the first and the second matrices on the one hand, and between the fourth and the fifth ones on the other hand. If $f=4$, the matrix product in Eq. (55b) must be restricted to the third, fourth, and fifth matrices (θ_{3f-8}, θ_{3f-7}, and θ_{3f-6} are then θ_4, θ_5, and θ_6, respectively) whereas, if $f=n-1$, it must be restricted to the first three matrices (θ_{3f-8}, θ_{3f-7}, and θ_{3f-6} are then θ_{3n-11}, θ_{3n-10}, and θ_{3n-9}, respectively).

2. Inversion of $\boldsymbol{\Delta}^n$

Since $\boldsymbol{\Delta}^{n-1}$ is an invariant block in $\boldsymbol{\Delta}^n$, the following open (i.e., nonfinite) upper triangular block matrix is introduced:

$$\boldsymbol{\Delta}=\begin{bmatrix} \mathbf{a}_{44} & \mathbf{a}_{45} & \mathbf{a}_{46} & \mathbf{a}_{47} & \mathbf{a}_{48} & & \cdots \\ \mathbf{0} & \mathbf{a}_{55} & \mathbf{a}_{56} & \mathbf{a}_{57} & \mathbf{a}_{58} & & \cdots \\ \mathbf{0} & \mathbf{0} & \mathbf{a}_{66} & \mathbf{a}_{67} & \mathbf{a}_{68} & & \cdots \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{a}_{77} & \mathbf{a}_{78} & & \cdots \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{a}_{88} & & \cdots \\ & & & & & & \mathbf{a}_{fn}(f\leq n) \\ \vdots & \vdots & \vdots & \vdots & \vdots & & \ddots \\ & & & & & \mathbf{0} & \end{bmatrix} . \tag{56a}$$

Formally, the inverse of $\boldsymbol{\Delta}$ is Eq. (56b) on the following page, where $\mathbf{A}_{fn}$ is the current 3×3 block of $\boldsymbol{\Delta}^{-1}$.

In the same way as $\boldsymbol{\Delta}^{n-1}$ is block invariant in $\boldsymbol{\Delta}^n$, according to known algebraic properties of block triangular matrices, $(\boldsymbol{\Delta}^{n-1})^{-1}$ is block invariant in $(\boldsymbol{\Delta}^n)^{-1}$. Indeed,

$$(\boldsymbol{\Delta}^n)^{-1}=\begin{bmatrix} & \vdots \\ \boldsymbol{\Delta}^{n-1} & \mathbf{a}_{kn} \\ & \vdots \\ \mathbf{0} & \mathbf{a}_{nn} \end{bmatrix}^{-1}=\begin{bmatrix} & \vdots \\ (\boldsymbol{\Delta}^{n-1})^{-1} & \mathbf{A}_{fn} \\ & \vdots \\ \mathbf{0} & \mathbf{a}_{nn}^{-1} \end{bmatrix}, \tag{57}$$

where

$$\mathbf{A}_{fn}=-\sum_{k=f}^{n-1}\mathbf{A}_{fk}\cdot\mathbf{a}_{kn}\cdot\mathbf{a}_{nn}^{-1}(4\leq f\leq n-1) \tag{58}$$

and $\mathbf{A}_{fk}$ in the right-hand side (rhs) of Eq. (58) is a block of $(\boldsymbol{\Delta}^{n-1})^{-1}$.

The explicit result (recursively derived, demonstration not given) is

$$\begin{aligned}\mathbf{A}_{fn}=-\Bigg[\mathbf{B}_{fn}&-\sum_{k=f+1}^{n-1}\mathbf{B}_{fk}\cdot\mathbf{B}_{kn}+\sum_{k=f+1}^{n-2}\sum_{l=k+1}^{n-1}\mathbf{B}_{k}\cdot\mathbf{B}_{kl}\cdot\mathbf{B}_{ln}-\cdots\\ &+(-1)^i\sum_{k=f+1}^{n-i}\sum_{l=k+1}^{n-i+1}\cdots\sum_{u}^{n-2}\sum_{v=u+1}^{n-1}\mathbf{B}_{fk}\cdot\mathbf{B}_{kl}\cdot\ \cdots\ \cdot\mathbf{B}_{uv}\cdot\mathbf{B}_{vn}+\cdots\\ &+(-1)^{n-f-1}\mathbf{B}_{f,f+1}\cdot\mathbf{B}_{f+1,f+2}\cdot\ \cdots\ \cdot\mathbf{B}_{n-2,n-1}\cdot\mathbf{B}_{n-1,n}\Bigg]\cdot\mathbf{a}_{nn}^{-1}\quad(n\geq 5;\ n-1\geq f\geq 4)\ ,\end{aligned} \tag{59}$$

where $\mathbf{B}_{fn}$ (3×3) stands for $\mathbf{a}_{ff}^{-1}\cdot\mathbf{a}_{fn}$ ($n\geq 5$; $n-1\geq f\geq 4$). Moreover,

$$\mathbf{A}_{nn}=\mathbf{a}_{nn}^{-1}\quad(n\geq 4)\ . \tag{60}$$

In the expression of $\mathbf{A}_{fn}$, Eq. (59), there are $(n-f)$ terms in large parentheses, and the ith term ($i=0,\ldots,n-f-1$) is in turn a sum of $\binom{n-f-1}{i}$ 3×3 matrix products, so that, on the whole, there are

$$\sum_{i=0}^{n-f-1}\binom{n-f-1}{i}=2^{n-f-1}$$

matrix products in $\mathbf{A}_{fn}$.

Let us particularize the results for five and six bodies ($n=4$ and 5, respectively):

$$
\Delta^{-1}=\left(\begin{array}{cccccc}
\mathbf{a}_{44}^{-1} & -\mathbf{a}_{44}^{-1}\mathbf{a}_{45}\mathbf{a}_{55}^{-1} & \begin{array}{c}-\mathbf{a}_{44}^{-1}(\mathbf{a}_{46}\\ -\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{56})\mathbf{a}_{66}^{-1}\end{array} & \begin{array}{c}-\mathbf{a}_{44}^{-1}[\mathbf{a}_{47}\\ -(\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{57}+\mathbf{a}_{46}\mathbf{a}_{66}^{-1}\mathbf{a}_{67})\\ +\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{56}\mathbf{a}_{66}^{-1}\mathbf{a}_{67}]\mathbf{a}_{77}^{-1}\end{array} & \begin{array}{c}-\mathbf{a}_{44}^{-1}\{\mathbf{a}_{48}\\ -[\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{58}+\mathbf{a}_{46}\mathbf{a}_{66}^{-1}\mathbf{a}_{68}+\mathbf{a}_{47}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}]\\ +[\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{56}\mathbf{a}_{66}^{-1}\mathbf{a}_{68}+\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{57}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}\\ +\mathbf{a}_{46}\mathbf{a}_{66}^{-1}\mathbf{a}_{67}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}]\\ -\mathbf{a}_{45}\mathbf{a}_{55}^{-1}\mathbf{a}_{56}\mathbf{a}_{66}^{-1}\mathbf{a}_{67}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}\}\mathbf{a}_{88}^{-1}\end{array} & \cdots \\
0 & \mathbf{a}_{55}^{-1} & -\mathbf{a}_{55}^{-1}\mathbf{a}_{56}\mathbf{a}_{66}^{-1} & \begin{array}{c}-\mathbf{a}_{55}^{-1}\{\mathbf{a}_{57}\\ -\mathbf{a}_{56}\mathbf{a}_{66}^{-1}\mathbf{a}_{67}\}\mathbf{a}_{77}^{-1}\end{array} & \begin{array}{c}-\mathbf{a}_{55}^{-1}\{\mathbf{a}_{58}\\ -[\mathbf{a}_{56}\mathbf{a}_{66}^{-1}\mathbf{a}_{68}+\mathbf{a}_{57}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}]\\ +\mathbf{a}_{56}\mathbf{a}_{66}^{-1}\mathbf{a}_{67}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}\}\mathbf{a}_{88}^{-1}\end{array} & \cdots \\
0 & 0 & \mathbf{a}_{66}^{-1} & -\mathbf{a}_{66}^{-1}\mathbf{a}_{67}\mathbf{a}_{77}^{-1} & -\mathbf{a}_{66}^{-1}\{\mathbf{a}_{68}-\mathbf{a}_{67}\mathbf{a}_{77}^{-1}\mathbf{a}_{78}\}\mathbf{a}_{88}^{-1} & \cdots \\
0 & 0 & 0 & \mathbf{a}_{77}^{-1} & -\mathbf{a}_{77}^{-1}\mathbf{a}_{78}\mathbf{a}_{88}^{-1} & \cdots \\
0 & 0 & 0 & 0 & \mathbf{a}_{88}^{-1} & \cdots \\
 & & & & & \mathbf{A}_{fn} \\
\vdots & \vdots & \vdots & \vdots & \vdots & 0 \quad \ddots
\end{array}\right) \tag{56b}
$$

$$\mathbf{A}_{44}=\mathbf{a}_{44}^{-1}=-\begin{bmatrix}0 & 0 & 1\\ 0 & \frac{1}{\sin\theta_4} & 0\\ \frac{1}{\sin\theta_4\sin\theta_5} & 0 & 0\end{bmatrix},$$

$$\mathbf{A}_{55}=\mathbf{a}_{55}^{-1}=\begin{bmatrix}0 & 0 & 1\\ 0 & \frac{1}{\sin\theta_7} & 0\\ \frac{1}{\sin\theta_7\sin\theta_8} & 0 & 0\end{bmatrix}\cdot\begin{bmatrix}\frac{1}{\sin\theta_6} & 0 & 0\\ \cot\theta_5\cot\theta_6 & \frac{1}{\sin\theta_5} & 0\\ \frac{\cot\theta_4\cot\theta_6}{\sin\theta_5} & \cot\theta_4\cot\theta_5 & \frac{1}{\sin\theta_4}\end{bmatrix},$$

$$\mathbf{A}_{45}=-\mathbf{a}_{44}^{-1}\mathbf{a}_{45}\mathbf{a}_{55}^{-1}$$
$$=-\mathbf{B}_{45}\mathbf{a}_{55}^{-1}$$
$$=-\begin{bmatrix}0 & \cot\theta_5 & +\frac{\cot\theta_6}{\sin\theta_5}\\ \cot\theta_6 & -\cot\theta_4 & 0\\ -\cot\theta_5 & 0 & -\frac{\cot\theta_4}{\sin\theta_5}\end{bmatrix}\cdot\begin{bmatrix}\frac{\cot\theta_8}{\sin\theta_7} & 0 & 0\\ 0 & \cot\theta_7 & 0\\ \cot\theta_7 & 0 & 0\end{bmatrix}.$$

$\mathbf{A}_{45}$ turns out to be the simplest particular case of a totally general analytical expression of $\mathbf{A}_{fn}$ that we have actually derived at the expense of terrible calculations (demonstration not given), namely,

$$A_{fn}=(-1)^n\begin{bmatrix}0 & \cot\theta_{3f-7} & \frac{\cot\theta_{3f-6}}{\sin\theta_{3f-7}}\\ \cot\theta_{3f-6} & -\cot\theta_{3f-8} & 0\\ -\cot\theta_{3f-7} & 0 & -\frac{\cot\theta_{3f-8}}{\sin\theta_{3f-7}}\end{bmatrix}\cdot\begin{bmatrix}\frac{1}{\sin\theta_{3f-9}} & 0 & 0\\ 0 & \frac{1}{\sin\theta_{3f-10}} & \cot\theta_{3f-10}\cot\theta_{3f-9}\\ 0 & 0 & \frac{1}{\sin\theta_{3f-9}}\end{bmatrix}$$
$$\cdots\cdots\begin{bmatrix}\frac{1}{\sin\theta_6} & 0 & 0\\ 0 & \frac{1}{\sin\theta_5} & \cot\theta_5\cot\theta_6\\ 0 & 0 & \frac{1}{\sin\theta_6}\end{bmatrix}\cdot\begin{bmatrix}\frac{1}{\sin\theta_{3f-4}} & 0 & \cot\theta_{3f-5}\cot\theta_{3f-4}\\ 0 & \frac{1}{\sin\theta_{3f-5}} & 0\\ 0 & 0 & \frac{1}{\sin\theta_{3f-5}}\end{bmatrix}$$
$$\cdots\cdots\begin{bmatrix}\frac{1}{\sin\theta_{3n-10}} & 0 & \cot\theta_{3n-11}\cot\theta_{3n-10}\\ 0 & \frac{1}{\sin\theta_{3n-11}} & 0\\ 0 & 0 & \frac{1}{\sin\theta_{3n-11}}\end{bmatrix}\cdot\begin{bmatrix}\frac{\cot\theta_{3n-7}}{\sin\theta_{3n-8}} & 0 & 0\\ 0 & \cot\theta_{3n-8} & 0\\ \cot\theta_{3n-8} & 0 & 0\end{bmatrix}, \quad (61)$$

where the expression must be restricted if $f=4$ or $f=n-1$. If $f=4$, the second and third matrices in Eq. (61), and the implicit product in between them, must be ignored. If $f=n-1$, the fourth and fifth matrices, and the implicit product in between, must in turn be ignored.

Remarkably, $\mathbf{A}_{fn}$ is a triangular 3×3 matrix of the type

$$\begin{bmatrix}\neq0 & \neq0 & 0\\ \neq0 & \neq0 & 0\\ \neq0 & 0 & 0\end{bmatrix}.$$

Since $\mathbf{A}_{nn}=\mathbf{a}_{nn}^{-1}$ is of the type

$$\begin{bmatrix}\neq0 & \neq0 & \neq0\\ \neq0 & \neq0 & 0\\ \neq0 & 0 & 0\end{bmatrix}.$$

[see Eq. (54b)], the property is also true for $\mathbf{A}_{fn}$, whatever f and n.

Finally, it is worth reminding the reader that $(\mathbf{D}^n)^{-1}$—in the same way as $(\boldsymbol{\Delta}^n)^{-1}$—is the left upper block of the following open matrix:

$$\mathbf{D}^{-1}=\begin{bmatrix} \mathbf{A}_{44}\cdot\mathbf{\Phi}^T & \mathbf{A}_{45}\cdot\mathbf{\Phi}^T & \mathbf{A}_{46}\cdot\mathbf{\Phi}^T & \cdots \\ 0 & \mathbf{A}_{55}\cdot\mathbf{\Phi}^T & \mathbf{A}_{56}\cdot\mathbf{\Phi}^T & \cdots \\ 0 & 0 & \mathbf{A}_{66}\cdot\mathbf{\Phi}^T & \cdots \\ & & & \ddots\ \mathbf{A}_{fn}\cdot\mathbf{\Phi}^T \\ \vdots & \vdots & \vdots & 0 \end{bmatrix} . \tag{62}$$

D. Hamiltonian expression of the kinetic energy

Now that $\mathbf{D}^n$ is inverted, the Eckart quasivelocities in Eq. (46) are used to define conjugate quasimomenta (cf. Sec. II), $P'_K=\partial T/\partial V'_K$ $(K=1,3n)$, with

$$V'_K=\{\dot\rho,\dot\theta,\dot\phi,\Omega_x,\Omega_y,\Omega_z,\omega_{x4},\ldots,\omega_{zn},\omega_x,\omega_y,\omega_z\} .$$

From Eq. (45), we obtain Eq. (63) on the following page.

The remaining inversion problem thus reduces to the following 2×2 one:

$$\begin{bmatrix} a_{g'}^2+a_{g''}^2 & -2a_{g'}a_{g''} \\ -2a_{g'}a_{g''} & a_{g'}^2+a_{g''}^2 \end{bmatrix}^{-1} =(a_{g'}^2-a_{g''}^2)^{-2}\begin{bmatrix} a_{g'}^2+a_{g''}^2 & 2a_{g'}a_{g''} \\ 2a_{g'}a_{g''} & a_{g'}^2+a_{g''}^2 \end{bmatrix} = \begin{bmatrix} B_g^{-1} & C_g^{-1} \\ C_g^{-1} & B_g^{-1} \end{bmatrix} , \tag{64a}$$

where $(gg'g''=xyz,yzx,zxy)$:

$$B_g=\frac{(a_{g'}^2-a_{g''}^2)^2}{a_{g'}^2+a_{g''}^2},\quad C_g=\frac{(a_{g'}^2-a_{g''}^2)^2}{2a_{g'}a_{g''}} . \tag{64b}$$

In terms of the quasimomenta of Eq. (63), the Hamiltonian form of the kinetic energy thus reads

$$2T=P_\rho^2+\frac{1}{\rho^2}\left[P_\theta^2+\frac{1}{\sin^2\theta}P_\phi^2\right] +\sum_g\left[\frac{1}{B_g}(K_g^2+J_g^2)+\frac{2}{C_g}K_gJ_g+\frac{1}{a_g^2}\sum_{\alpha=4}^{n}N_{g\alpha}^2\right] , \tag{65}$$

where ρ is the mass-weighted hyperradius defined in Eq. (20a).

This exceptionally concise expression of the kinetic energy of an N-particle system presents several noticeable characteristics.

(i) The usual rotational and Coriolis energies reduce, respectively, to $\frac{1}{2}\sum_g(1/B_g)J_g^2$ and $\sum_g(1/C_g)J_gK_g$, all the rest being pure internal deformation kinetic energy.

(ii) All coefficients in Eq. (65) depend solely on ρ, θ, and ϕ. All the other (internal) coordinates, namely, θ_i $(i=1,3n-6)$, are contained in the quasimomenta themselves, respectively, K_g and $N_{g\alpha}$ $(g=x,y,z;\ \alpha=4,n)$.

(iii) From the matrix in Eq. (46) that transforms the velocities into quasivelocities, and from the fact that the matrix transforming the conjugate momenta into quasimomenta is the transpose inverse of the previous one [cf. Sec. II, Eqs. (10a) and (10b)], one obtains [see Eq. (36b)]

$$\begin{bmatrix} K_x \\ K_y \\ K_z \\ N_{x4} \\ \vdots \\ N_{zn} \end{bmatrix} = \begin{bmatrix} \boldsymbol{\omega}^{*-1} & 0 \\ -(\mathbf{D}^{nT})^{-1}\cdot\boldsymbol{\gamma}^T\cdot\mathbf{\Xi}^T & (\mathbf{D}^{nT})^{-1} \end{bmatrix}\cdot\begin{bmatrix} P_{\theta_1} \\ P_{\theta_2} \\ P_{\theta_3} \\ P_{\theta_4} \\ \vdots \\ P_{\theta_{3n-6}} \end{bmatrix} , \tag{66a}$$

where

$$\boldsymbol{\omega}^{*-1}(\theta_1,\theta_2)=\begin{bmatrix} -\sin\theta_1\cot\theta_2 & \cos\theta_1 & \sin\theta_1/\sin\theta_2 \\ -\cos\theta_1\cot\theta_2 & -\sin\theta_1 & \cos\theta_1/\sin\theta_2 \\ 1 & 0 & 0 \end{bmatrix} \tag{66b}$$

and

$$\mathbf{\Phi}^T\cdot\boldsymbol{\omega}^{*-1}=\mathbf{\Xi}^T(\theta_2,\theta_3) =\begin{bmatrix} \sin\theta_3/\sin\theta_2 & \cos\theta_3 & -\cot\theta_2\sin\theta_3 \\ -\cos\theta_3/\sin\theta_2 & \sin\theta_3 & \cot\theta_2\cos\theta_3 \\ 0 & 0 & 1 \end{bmatrix} . \tag{66c}$$

As a mathematical consequence of the factorization of a three-dimensional Eulerian rotation $g_{(1)}(\theta_1)g_{(2)}(\theta_2,\theta_3)$ on the left side of g [see Eqs. (27) and (28)], the expression of the general rotation of $\mathbb{R}^n$ (see Sec. III B), one has

$$\mathbf{K}^T=\begin{bmatrix} K_x \\ K_y \\ K_z \end{bmatrix}=\boldsymbol{\omega}^{*-1}(\theta_1,\theta_2)\begin{bmatrix} P_{\theta_1} \\ P_{\theta_2} \\ P_{\theta_3} \end{bmatrix} . \tag{67}$$

Algebraically, the vector $\mathbf{K}$ is an *angular momentum*. This is clearly seen by analogy with $\mathbf{J}$, cf. Eq. (3):

$$\mathbf{J}^T=\begin{bmatrix} J_x \\ J_y \\ J_z \end{bmatrix}=\boldsymbol{\omega}^{*-1}\left[\gamma-\frac{\pi}{2},\beta\right]\begin{bmatrix} P_\gamma \\ P_\beta \\ P_\alpha \end{bmatrix} .$$

This is why, in the PA-hyperspherical Hamiltonian expression of the kinetic energy, Eq. (65), both $\mathbf{K}$ and $\mathbf{J}$ are on the same footing. We call $\mathbf{K}$ "pseudo-angular-momentum," in accordance with our previous work on four-particle systems in which $\mathbf{K}$ was identified for the first time.[36] Obviously, the pseudo-angular-momentum $\mathbf{K}$ is not a rotational angular momentum, since it refers to the internal coordinates θ_1, θ_2, and θ_3, and not to the rotational Eulerian angles α, β, and γ. In particular, *it is not a constant of the motion.* However, being defined by the same explicit relationships as the true angular

$$
\begin{pmatrix} P_\rho \\ P_\theta \\ P_\phi \\ K_x \\ K_y \\ K_z \\ N_{x4} \\ \vdots \\ N_{xn} \\ N_{y4} \\ \vdots \\ N_{yn} \\ N_{z4} \\ \vdots \\ N_{zn} \\ J_x \\ J_y \\ J_z \end{pmatrix}
=
\left(\begin{array}{ccc|ccc|ccccccccc|ccc}
1 & 0 & 0 & & & & & & & & & & & & & & & \\
0 & \rho^2 & 0 & & \mathbf{0} & & & & & & \mathbf{0} & & & & & & \mathbf{0} & \\
0 & 0 & \rho^2\sin^2\theta & & & & & & & & & & & & & & & \\
 & & & a_y^2+a_z^2 & 0 & 0 & & & & & & & & & & -2a_ya_z & 0 & 0 \\
 & \mathbf{0} & & 0 & a_z^2+a_x^2 & 0 & & & & & \mathbf{0} & & & & & 0 & -2a_za_x & 0 \\
 & & & 0 & 0 & a_x^2+a_y^2 & & & & & & & & & & 0 & 0 & -2a_xa_y \\
 & & & & & & a_x^2 & & & & & & & & & & & \\
 & & & & & & & \ddots & & & & & & \mathbf{0} & & & & \\
 & & & & & & & & a_x^2 & & & & & & & & & \\
 & & & & & & & & & a_y^2 & & & & & & & & \\
 & \mathbf{0} & & & \mathbf{0} & & & & & & \ddots & & & & & & \mathbf{0} & \\
 & & & & & & & & & & & a_y^2 & & & & & & \\
 & & & & & & & & & & & & a_z^2 & & & & & \\
 & & & & & & & \mathbf{0} & & & & & & \ddots & & & & \\
 & & & & & & & & & & & & & & a_z^2 & & & \\
 & & & -2a_ya_z & 0 & 0 & & & & & & & & & & a_y^2+a_z^2 & 0 & 0 \\
 & \mathbf{0} & & 0 & -2a_za_x & 0 & & & & & \mathbf{0} & & & & & 0 & a_z^2+a_x^2 & 0 \\
 & & & 0 & 0 & -2a_xa_y & & & & & & & & & & 0 & 0 & a_x^2+a_y^2
\end{array}\right)
\cdot
\begin{pmatrix} \dot\rho \\ \dot\theta \\ \dot\phi \\ \Omega_x \\ \Omega_y \\ \Omega_z \\ \omega_{x4} \\ \vdots \\ \omega_{xn} \\ \omega_{y4} \\ \vdots \\ \omega_{yn} \\ \omega_{z4} \\ \vdots \\ \omega_{zn} \\ \omega_x \\ \omega_y \\ \omega_z \end{pmatrix} . \qquad (63)
$$

momentum, in quantum mechanics, its components as operators obey the same commutation rules as those of the angular momentum, namely $[\hat{K}_x,\hat{K}_y]=-i\hbar\hat{K}_z$, and so on, so that[35]

$$\hat{K}^2|K,k\rangle=\hbar^2K(K+1)|K,k\rangle ,$$
$$\hat{K}_z|K,k\rangle=\hbar k|K,k\rangle ,$$
$$(\hat{K}_x\mp i\hat{K}_y)|K,k\rangle=\hbar\sqrt{K(K+1)-k(k\pm1)}|K,k\pm1\rangle ,$$

where K is a positive integer (or zero), $k\in[-K,+K]$ and

$$\langle\theta_1,\theta_2,\theta_3|K,k\rangle=\mathcal{D}^K_{0k}(\theta_3,\theta_2,\theta_1) ,$$

where $\mathcal{D}^K_{0k}$ denotes a Wigner matrix element.

(iv) $2T$ can be rewritten as

$$2T=P_\rho^2+\frac{\Lambda^2}{\rho^2} , \tag{68a}$$

where

$$\Lambda^2=P_\theta^2+\frac{1}{\sin^2\theta}P_\phi^2+\sum_g\left[(e_{g'}^2-e_{g''}^2)^{-2}[b_g(K_g^2+J_g^2)+2d_gK_gJ_g]+e_g^{-2}\sum_{\alpha=4}^{n}N_{g\alpha}^2\right] \tag{68b}$$

is the PA-hyperspherical version of the so-called "grand angular momentum,"[2–4,21–23,30] and

$$e_x=\sin\theta\cos\phi,\quad e_y=\sin\theta\sin\phi,\quad e_z=\cos\theta ,$$
$$b_x=1-\sin^2\theta\cos^2\phi,\quad b_y=1-\sin^2\theta\sin^2\phi,\quad b_z=\sin^2\theta , \tag{68c}$$
$$d_x=\sin2\theta\sin\phi,\quad d_y=\sin2\theta\cos\phi,\quad d_z=\sin2\theta\sin2\phi .$$

It should be noted here that there are no K_g and no $N_{g\alpha}$ for three particles, and that there are even no $N_{g\alpha}$ for four particles, thus indicating that the general N-particle regime settles in, according to the PA-hyperspherical description, for *five particles*.

(v) The following completely diagonal expression of Λ^2 is worth mentioning:

$$\Lambda^2=P_\theta^2+\frac{1}{\sin^2\theta}P_\phi^2+\sum_g\left[(e_{g'}^2-e_{g''}^2)^{-2}b_g(J_g')^2+b_g^{-1}K_g^2+e_g^{-2}\sum_{\alpha=4}^{n}N_{g\alpha}^2\right] , \tag{69a}$$

where

$$J_g'=J_g+d_gb_g^{-1}K_g \quad (g=x,y,z) \tag{69b}$$

is a generalized angular momentum component of the Darling-Dennison type[34] (see Sec. II), adapted to large amplitude deformations of the system described in terms of PA-hyperspherical coordinates.

V. CONCLUSION

The aim of the present article is twofold. First, an explicit definition of a set of PA-hyperspherical coordinates has been proposed. The basic algebraic ingredients are not new: we owe them to Eckart, in a very old and surprisingly ignored article,[28] the rediscovery of which we owe to Robert and Baudon.[29,30] Our contribution has been to create coordinates and completely calculate all the formal quantities identified by Eckart; in particular, the quasivelocities $\Omega_x,\Omega_y,\Omega_z,\omega_{x4},\ldots,\omega_{zn}$ and the quasimomenta $K_x,K_y,K_z,N_{x4},\ldots,N_{zn}$, thus allowing utilization of Eckart's ideas in practice. No singularities have appeared in the course of the extremely long and tricky calculations, thus proving that the PA-hyperspherical scheme is always applicable to the N-body problem formulated in terms of $3N-6$ internal coordinates and the three components of the total angular momentum measured in the body-fixed frame.

Second, the Hamiltonian expression of the kinetic energy expressed with the help of the quasimomenta has been put in a very concise form, which is well adapted to quantization. To quantize Eq. (65), nothing new is needed for both J_g—the usual body-fixed component of the total angular momentum—and K_g—the internal pseudo-angular-momentum (cf. Sec. IV D). All the difficulty therefore lies in the quasimomentum operators that are to be associated with the $N_{g\alpha}$'s. This is planned to be the subject of a future article.[31]

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