Appendix A

Angular Momentum of the Diatomic Rotor

The square of the angular momentum vector in SP coordinates can be easily calculated

$$\begin{aligned} j^2 &= j_x^2 + j_y^2 + j_z^2 \\ &= p_\theta^2 + \left(\frac{p_\phi}{\sin\theta}\right)^2. \end{aligned} \tag{A.1}$$

From this result, the rigid rotor Hamiltonian can be rewritten as

$$H = \frac{j^2}{2I}. \tag{A.2}$$

Using the Poisson Brackets (PB) formalism [20] is possible to calculate the time-dependence of the angular momentum for the rigid rotor. The time derivative of the magnitude of the angular momentum j in terms of the PB is

$$\frac{\mathrm{d}j}{\mathrm{d}t} = [j, H] + \frac{\partial j}{\partial t}, \tag{A.3}$$

where the square brackets are the PB of j and H with respect to the canonical variables θ, ϕ, p_θ and p_ϕ. Using (A.2) and the fact that j does not depend explicitly on t (depends on t only through the canonical variables) is obtained from the last equation

$$\begin{aligned} \frac{\mathrm{d}j}{\mathrm{d}t} &= \left[j, \frac{j^2}{2I}\right] \\ &= \frac{1}{2I}\left[j, j^2\right] \\ &= \frac{1}{2I}\sum_i \left(\frac{\partial j}{\partial q_i}\frac{\partial j^2}{\partial p_i} - \frac{\partial j}{\partial p_i}\frac{\partial j^2}{\partial p_i}\right) \\ &= \frac{j}{I}\sum_i \left(\frac{\partial j}{\partial q_i}\frac{\partial j}{\partial p_i} - \frac{\partial j}{\partial p_i}\frac{\partial j}{\partial p_i}\right) = 0, \end{aligned} \tag{A.4}$$

with the index i standing for $\{\theta, \phi\}$, $q_\theta = \theta$ and $q_\phi = \phi$. This result is indicating that the magnitude of the total angular momentum, j, is a constant of motion for the rigid rotor.

It is also important to identify the z-component of the angular momentum as a constant of motion. Using the PB formalism, the time derivative of $m = p_\phi$ is

$$\begin{aligned} \frac{\mathrm{d}m}{\mathrm{d}t} &= [m, H] + \frac{\partial m}{\partial t} \\ &= \frac{j}{I} \sum_i \left(\frac{\partial m}{\partial q_i} \frac{\partial j}{\partial p_i} - \frac{\partial m}{\partial p_i} \frac{\partial j}{\partial p_i} \right) \\ &= -\frac{j}{I} \frac{\partial j}{\partial \phi} = 0, \end{aligned} \tag{A.5}$$

since that ϕ is an ignorable coordinate in the rigid rotor Hamiltonian (A.2).

Since m and j are constants of motion, any function of them will be also a constant of motion. A particularly interesting case is the angle between the positive lab-fixed z-axis and the $\boldsymbol{j}$ vector

$$\bar{\theta} = \arccos \frac{m}{j}. \tag{A.6}$$

Another important function of j and m is their square difference, $j^2 - m^2$, which can be written in terms of the lab-fixed components of $\boldsymbol{j}$

$$j^2 - m^2 = j_x^2 + j_y^2 = \text{constant}. \tag{A.7}$$

According to this equation, the projection of the vector $\boldsymbol{j}$ onto the lab-fixed xy-plane is restricted to move on a circle of radius $\sqrt{j^2 - m^2}$. The angle between the lab-fixed x-axis and this projection of $\boldsymbol{j}$ is

$$\bar{\phi} = \arctan{(j_y/j_x)}. \tag{A.8}$$

It can be shown using PB formalism that the time derivative of the ratio j_y/j_x is zero. This result means that $\bar{\phi}$ is a constant of motion for the rigid rotor.

Another important result for the rigid rotor is that $\boldsymbol{r}$ and $\boldsymbol{j}$ are perpendicular to one another. This can be demonstrated easily by calculating, either in cartesian or SP coordinates, the scalar product

$$\boldsymbol{r}\cdot\boldsymbol{j} = xj_x + yj_y + zj_z = 0. \tag{A.9}$$

Equations (A.6)-(A.9) indicate that the rotor is moving in a circular orbit that lies on a plane with normal vector proportional to $\boldsymbol{j}$. Besides, since m is a constant (positive or negative), the system is rotating about $\boldsymbol{j}$ in the same direction all the time. At the turning points in the θ coordinate, where the rotor reaches its smallest and largest values, $\dot{\theta}$ must be zero and equation (2.43a) gives $p_\theta = 0$. Replacing this into equation (A.1) and solving for θ casts two possible solutions

$$\theta_0 = \begin{cases} \arcsin\left(m/j\right), \\ \pi - \arcsin\left(m/j\right). \end{cases} \tag{A.10}$$

This gives the smallest and largest values of θ respectively. Using equation (2.45), at the smallest value of θ, θ_0, the position vector in SP coordinates is

$$\boldsymbol{r}_0 = r\left(\frac{m}{j}\cos\phi, \frac{m}{j}\cos\phi, \sqrt{1-\frac{m^2}{j^2}}\right). \tag{A.11}$$

This vector $\boldsymbol{r}_0$ is lying in the same plane together with the z-axis ($\boldsymbol{e}_z$) and $\boldsymbol{j}$. This can be shown comparing the cross products $\boldsymbol{e}_z \times \boldsymbol{r}_0$ and $\boldsymbol{j}\times\boldsymbol{e}_z$

$$\boldsymbol{e}_z \times \boldsymbol{r}_0 = \frac{rm}{j}\left(-\sin\phi, \cos\phi, 0\right), \tag{A.12a}$$

$$\boldsymbol{j}\times\boldsymbol{e}_z = \left(j_y, -j_x, 0\right). \tag{A.12b}$$

Using (2.52) in the second of the equations above and evaluating at θ_0 will produce

$$\boldsymbol{j}\times\boldsymbol{e}_z = j\sqrt{1-\frac{m^2}{j^2}}\left(-\sin\phi, \cos\phi, 0\right). \tag{A.13}$$

The vectors (A.12a) and (A.13) will be identical after normalization.

From equation (2.13c) the z coordinate of the point at θ_0 is given by

$$\begin{aligned} z_0 &= r\cos\theta_0 \\ &= r\cos\left(\arcsin\left(m/j\right)\right) \\ &= \frac{r}{j}\sqrt{j^2 - m^2}. \end{aligned} \tag{A.14}$$

It is clear that $z_0 \geq 0$ all the time.

Replacing (A.14) into equation (A.9) and using the definition of the SP coordinate r it is obtained a system of equations to solve for x and y

$$xj_x + yj_y + r\frac{j_z}{j}\sqrt{j^2 - m^2} = 0, \tag{A.15a}$$

$$x^2 + y^2 + \frac{r^2}{j^2}\left(j^2 - m^2\right) = r^2. \tag{A.15b}$$

It is more convenient to solve the first equation for y and replace it into the second one to get a quadratic equation in x

$$\left[1 + \left(\frac{j_x}{j_y}\right)^2\right] x^2 + \left(\frac{2mrj_x}{jj_y^2}\sqrt{j^2 - m^2}\right) x + \frac{m^2r^2}{j^2}\left(\frac{j_x}{j_y}\right)^2 = 0. \tag{A.16}$$

This equation has only one solution for x, x_0, which after plugging, together with z_0, into (A.15a) gives the coordinates of the point P_0 at which θ has its smallest value θ_0

$$P_0 = -\frac{mr}{j\sqrt{j^2 - m^2}}\left(j_x, j_y, -\frac{j^2 - m^2}{m}\right). \tag{A.17}$$

With this point and the direction of $\boldsymbol{j}$ it is possible to define a cartesian coordinate frame with the z axis along the direction of $\boldsymbol{j}$ and the x axis defined by the position of P_0. Since that the distance from any point along the trajectory to the origin is always r, the unit-vector from the origin to P_0 is

$$\boldsymbol{e}_{\bar{x}} = \frac{P_0}{\|P_0\|} = -\frac{m}{j\sqrt{j^2 - m^2}}\left(j_x, j_y, -\frac{j^2 - m^2}{m}\right). \tag{A.18}$$

The $\boldsymbol{e}_{\bar{z}}$ unit-vector is defined with the same direction of $\boldsymbol{j}$

$$\boldsymbol{e}_{\bar{z}} = \frac{\boldsymbol{j}}{j} = \frac{1}{j}\left(j_x, j_y, m\right). \tag{A.19}$$

Finally, the last unit-vector, $\boldsymbol{e}_{\bar{y}}$, is obtained from

$$\boldsymbol{e}_{\bar{y}} = \boldsymbol{e}_{\bar{z}} \times \boldsymbol{e}_{\bar{x}} = \frac{1}{\sqrt{j^2 - m^2}}\left(j_y, -j_x, 0\right). \tag{A.20}$$

With these three unit-vectors now it is possible to define an angle $\bar{\psi}$ for the motion of the rotor in the $\bar{x}\bar{y}$ plane relative to the $\bar{x}$ axis

$$\begin{aligned}\bar{\psi} &= \arctan \frac{\boldsymbol{e}_{\bar{y}} \cdot \boldsymbol{r}}{\boldsymbol{e}_{\bar{x}} \cdot \boldsymbol{r}} \\ &= \arctan \frac{x j_y - y j_x}{z j} \\ &= \arctan \frac{\sqrt{j^2 \sin^2\theta - m^2}}{j \cos\theta}.\end{aligned} \tag{A.21}$$

The time derivative of $\bar{\psi}$ is calculated using PB formalism to get

$$\frac{\mathrm{d}\bar{\psi}}{\mathrm{d}t} = \frac{j}{I}. \tag{A.22}$$

This is indicating that the rigid rotor is moving on a plane perpendicular to $\boldsymbol{j}$ with constant angular velocity that depends only on j.

Appendix B

Cartesian Representation of the Coordinates

B.1 Spherical Polar Coordinates

Since the numerical integration has to be performed in cartesian coordinates and the study of the dynamics is easier to understand in SP or action-angle variables it is necessary to get the expressions for the SP (and action-angle) coordinates, momenta and phase space flow in terms of their cartesian analogues.

The inverse of equations (2.13) can be easily calculated to get

$$r = \sqrt{x^2 + y^2 + z^2}, \tag{B.1a}$$

$$\theta = \arccos(z/r), \tag{B.1b}$$

$$\phi = \begin{cases} \arctan(y/x), & x > 0, \\ \phi = \pi + \arctan(y/x), & x < 0. \end{cases} \tag{B.1c}$$

Using these equations the transformation matrix S, defined by (2.16), becomes

$$\mathsf{S} = \begin{pmatrix} x/r & xz/\tilde{r} & -y \\ y/r & yz/\tilde{r} & x \\ z/r & -\tilde{r} & 0 \end{pmatrix}, \tag{B.2}$$

where $\tilde{r} = \sqrt{x^2 + y^2}$, and the inverse S^{-1} is

$$\mathsf{S}^{-1} = \begin{pmatrix} x/r & y/r & z/r \\ xz/\tilde{r}r^2 & yz/\tilde{r}r^2 & -\tilde{r}/r^2 \\ -y/\tilde{r}^2 & x/\tilde{r}^2 & 0 \end{pmatrix}. \tag{B.3}$$

The vector, $\dot{\boldsymbol{\theta}}$, of velocities in SP coordinates is given by the inverse of equation (2.15)

$$\dot{\boldsymbol{\theta}} = \mathsf{S}^{-1}\dot{\boldsymbol{r}}, \tag{B.4}$$

or more explicitly

$$\begin{pmatrix} \dot{r} \\ \dot{\theta} \\ \dot{\phi} \end{pmatrix} = \begin{pmatrix} x/r & y/r & z/r \\ xz/\tilde{r}r^2 & yz/\tilde{r}r^2 & -\tilde{r}/r^2 \\ -y/\tilde{r}^2 & x/\tilde{r}^2 & 0 \end{pmatrix} \begin{pmatrix} \dot{x} \\ \dot{y} \\ \dot{z} \end{pmatrix}. \tag{B.5}$$

The momentum vector, $\boldsymbol{p}_\theta$, in SP coordinates can be found with the help of equation (A.7). The metric tensor g in cartesian coordinates is

$$\mathsf{g} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & r^2 & 0 \\ 0 & 0 & \tilde{r}^2 \end{pmatrix}, \tag{B.6}$$

and $\boldsymbol{p}_\theta$ is finally given by

$$\boldsymbol{p}_\theta = \mathsf{g}\mathsf{S}^{-1}\boldsymbol{p}, \tag{B.7}$$

which is

$$\begin{pmatrix} p_r \\ p_\theta \\ p_\phi \end{pmatrix} = \begin{pmatrix} x/r & y/r & z/r \\ xz/\tilde{r} & yz/\tilde{r} & -\tilde{r} \\ -y & x & 0 \end{pmatrix} \begin{pmatrix} p_x \\ p_y \\ p_z \end{pmatrix}. \tag{B.8}$$

The dynamics in phase space can be described in terms of the vector $\boldsymbol{z}$ which, in any canonical set of variables, is simply the concatenation of $\boldsymbol{r}$ and $\boldsymbol{p}$. The time derivative of $\boldsymbol{z}$, $\dot{\boldsymbol{z}}$, is the phase space flow vector which for $\boldsymbol{z}$ in cartesian coordinates is given by

$$\dot{\boldsymbol{z}} = \begin{pmatrix} \dot{\boldsymbol{r}} \\ \dot{\boldsymbol{p}} \end{pmatrix}. \tag{B.9}$$

Similarly in SP coordinates the phase space flow vector $\dot{\boldsymbol{z}}_\theta$ is defined as

$$\dot{\boldsymbol{z}}_\theta = \begin{pmatrix} \dot{\boldsymbol{\theta}} \\ \\ \dot{\boldsymbol{p}}_\theta \end{pmatrix}. \tag{B.10}$$

These two phase space flow vectors are related through

$$\dot{\boldsymbol{z}}_\theta = \mathsf{D}\dot{\boldsymbol{z}}, \tag{B.11}$$

where D is the Jacobian transformation matrix with elements

$$D_{ij} = \frac{\partial z_{\theta,i}}{\partial z_j}. \tag{B.12}$$

These elements are partially obtained from equation (B.5) and completed using the time derivative of (B.8). The gradients of the SP momenta with respect to $\boldsymbol{r}$ and $\boldsymbol{p}$ are

$$\nabla_{\boldsymbol{r}} p_r = -\frac{\boldsymbol{r} \times \boldsymbol{j}}{r^3}, \tag{B.13a}$$

$$\nabla_{\boldsymbol{p}} p_r = \frac{\boldsymbol{r}}{r}, \tag{B.13b}$$

$$\nabla_{\boldsymbol{r}} p_\theta = \frac{1}{\tilde{r}} \left(-\frac{yzj_z}{\tilde{r}^2} - xp_z, \; \frac{xzj_z}{\tilde{r}^2} - yp_z, xp_x + yp_y \right), \tag{B.13c}$$

$$\nabla_{\boldsymbol{p}} p_\theta = \frac{1}{\tilde{r}} \left(xz, yz, -\tilde{r}^2 \right), \tag{B.13d}$$

$$\nabla_{\boldsymbol{r}} p_\phi = \left(p_y, -p_x, 0 \right), \tag{B.13e}$$

$$\nabla_{\boldsymbol{p}} p_\phi = \left(-y, x, 0 \right). \tag{B.13f}$$

Using these gradients the matrix $\mathsf{g}\mathsf{S}^{-1}$ can be written as a row concatenation of the gradients with respect to $\boldsymbol{p}$

$$\mathsf{g}\mathsf{S}^{-1} = \begin{pmatrix} \nabla_{\boldsymbol{p}} p_r \\ \\ \nabla_{\boldsymbol{p}} p_\theta \\ \\ \nabla_{\boldsymbol{p}} p_\phi \end{pmatrix}. \tag{B.14}$$

Seemingly is defined the matrix

$$\mathsf{B} = \begin{pmatrix} \nabla_{\boldsymbol{r}} p_r \\ \nabla_{\boldsymbol{r}} p_\theta \\ \nabla_{\boldsymbol{r}} p_\phi \end{pmatrix}. \tag{B.15}$$

resulting of concatenation by rows of the gradients with respect to $\boldsymbol{r}$.

With these definitions the time derivative of the SP momentum vector can be written as

$$\dot{\boldsymbol{p}}_\theta = \mathsf{B}\dot{\boldsymbol{r}} + \mathsf{g}\mathsf{S}^{-1}\dot{\boldsymbol{p}}, \tag{B.16}$$

and equation (B.11) is

$$\dot{\boldsymbol{z}}_\theta = \begin{pmatrix} \mathsf{S}^{-1} & 0 \\ \mathsf{B} & \mathsf{g}\mathsf{S}^{-1} \end{pmatrix} \dot{\boldsymbol{z}}. \tag{B.17}$$

B.2 Action-Angle Variables

The action variable j can be written directly in terms of cartesian coordinates

$$\begin{aligned} j &= \sqrt{j_x^2 + j_y^2 + j_z^2} \\ &= \left[(yp_z - zp_y)^2 + (xp_z - zp_y)^2 + (xp_y - yp_x)^2 \right]^{1/2}. \end{aligned} \tag{B.18}$$

Seemingly for m

$$m = xp_y - yp_x. \tag{B.19}$$

The angle q_j can be evaluated in term of cartesian coordinates resorting to equation (A.21)

$$\begin{aligned} q_j = \bar{\psi} &= \arctan \frac{xj_y - yj_x}{zj} \\ &= \arctan \frac{x\,(zp_x - xp_z) - y\,(yp_z - zp_y)}{zj}, \end{aligned} \tag{B.20}$$

with j defined above. Finally, the angle variable q_m can be expressed in cartesian coordinates with the use of equation (A.8)

$$\begin{aligned} q_m &= \bar{\phi} = \arctan j_y/j_x \\ &= \arctan \frac{zp_x - xp_z}{yp_z - zp_y}. \end{aligned} \tag{B.21}$$

The time derivative of the j action is given by

$$\frac{\mathrm{d}j}{\mathrm{d}t} = \frac{1}{j}\left(\dot{\boldsymbol{r}} \cdot \boldsymbol{p} \times \boldsymbol{j} - \dot{\boldsymbol{p}} \cdot \boldsymbol{r} \times \boldsymbol{j}\right). \tag{B.22}$$

Using equations (B.13e) and (B.13f) the time derivative of m is

$$\frac{\mathrm{d}m}{\mathrm{d}t} = \nabla_{\boldsymbol{r}} p_\phi \cdot \dot{\boldsymbol{r}} + \nabla_{\boldsymbol{p}} p_\phi \cdot \dot{\boldsymbol{p}}. \tag{B.23}$$

For the angle q_j it is suitable first to define u as the argument of the arctan in (B.20)

$$u \equiv \frac{[\boldsymbol{r} \times \boldsymbol{j}]_z}{zj} = \frac{xj_y - yj_x}{zj}. \tag{B.24}$$

The gradient of u with respect to $\boldsymbol{z}$ is given by

$$\frac{\partial u}{\partial x} = \frac{1}{zj}\left(j_y - xp_z - \frac{1}{j^2}[\boldsymbol{r} \times \boldsymbol{j}]_z[\boldsymbol{p} \times \boldsymbol{j}]_x\right), \tag{B.25a}$$

$$\frac{\partial u}{\partial y} = \frac{1}{zj}\left(-j_x - yp_z - \frac{1}{j^2}[\boldsymbol{r} \times \boldsymbol{j}]_z[\boldsymbol{p} \times \boldsymbol{j}]_y\right), \tag{B.25b}$$

$$\frac{\partial u}{\partial z} = \frac{1}{zj}\left(xp_x + yp_y - \frac{1}{z}[\boldsymbol{r} \times \boldsymbol{j}]_z - \frac{1}{j^2}[\boldsymbol{r} \times \boldsymbol{j}]_z[\boldsymbol{p} \times \boldsymbol{j}]_z\right), \tag{B.25c}$$

$$\frac{\partial u}{\partial p_x} = \frac{1}{zj}\left(xz + \frac{1}{j^2}[\boldsymbol{r} \times \boldsymbol{j}]_z[\boldsymbol{p} \times \boldsymbol{j}]_x\right), \tag{B.25d}$$

$$\frac{\partial u}{\partial p_y} = \frac{1}{zj}\left(yz + \frac{1}{j^2}[\boldsymbol{r} \times \boldsymbol{j}]_z[\boldsymbol{p} \times \boldsymbol{j}]_y\right), \tag{B.25e}$$

$$\frac{\partial u}{\partial p_y} = \frac{1}{zj}\left(-x^2 - y^2 + \frac{1}{j^2}[\boldsymbol{r} \times \boldsymbol{j}]_z[\boldsymbol{p} \times \boldsymbol{j}]_z\right). \tag{B.25f}$$

The time derivative of q_j is then given by

$$\dot{q}_j = \frac{1}{1+u^2}\left(\nabla_{\boldsymbol{r}} u \cdot \dot{\boldsymbol{r}} + \nabla_{\boldsymbol{p}} u \cdot \dot{\boldsymbol{p}}\right). \tag{B.26}$$

Following a similar procedure in the case of the angle q_m, v is defined as the argument of the arctan in (B.21)

$$v = \frac{j_y}{j_x} = \frac{zp_x - xp_z}{yp_z - zp_y}. \tag{B.27}$$

The components of the gradient of v with respect to $\boldsymbol{z}$ are

$$\nabla_{\boldsymbol{r}} v = -\frac{p_z}{j_x^2}\boldsymbol{j}, \tag{B.28a}$$

$$\nabla_{\boldsymbol{p}} v = \frac{z}{j_x^2}\boldsymbol{j}. \tag{B.28b}$$

With these results the time derivative of q_m becomes

$$\dot{q}_m = \frac{1}{1+v^2}\left(\nabla_{\boldsymbol{r}} v \cdot \dot{\boldsymbol{r}} + \nabla_{\boldsymbol{p}} v \cdot \dot{\boldsymbol{p}}\right). \tag{B.29}$$

Appendix C

Stability Matrix

The position vector in phase space is simply defined $\boldsymbol{z} = (\varphi, j)$. The equations of motion for φ and j can be written as

$$\dot{\boldsymbol{z}} = \boldsymbol{f}(\boldsymbol{z}; t), \tag{C.1}$$

with $\boldsymbol{f}(\boldsymbol{z}; t)$ as the vector

$$\boldsymbol{f}(\boldsymbol{z}; t) = \left(\partial H/\partial j, -\partial H/\partial \varphi\right). \tag{C.2}$$

Let $\boldsymbol{z}_t = \boldsymbol{z}(\boldsymbol{z}_i, t)$ be a trajectory with initial condition $\boldsymbol{z}_i$. If the initial condition is slightly displaced by $\delta\boldsymbol{z}_i$ (according to the imposed constraints), this originates a second trajectory $\boldsymbol{z}'_t$ that can be written in terms of $\boldsymbol{z}_t$ like

$$\begin{aligned}
\boldsymbol{z}'_t &= \boldsymbol{z}(\boldsymbol{z}_i + \delta\boldsymbol{z}_i, t) \\
&= \boldsymbol{z}(\boldsymbol{z}_i, t) + \frac{\partial \boldsymbol{z}}{\partial \boldsymbol{z}_i} \cdot \delta\boldsymbol{z}_i + \mathcal{O}(|\delta\boldsymbol{z}_i|^2) \\
&= \boldsymbol{z}_t(\boldsymbol{z}_i, t) + \frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{z}_i} \cdot \delta\boldsymbol{z}_i + \mathcal{O}(|\delta\boldsymbol{z}_i|^2).
\end{aligned} \tag{C.3}$$

Taking the difference between these two trajectories is obtained, up to first order in the displacement,

$$\begin{aligned}
\delta\boldsymbol{z}_t &\equiv \boldsymbol{z}'_t - \boldsymbol{z}_t \\
&= \frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{z}_i} \cdot \delta\boldsymbol{z}_i.
\end{aligned} \tag{C.4}$$

For the $\boldsymbol{z}'_t$, from the flow equation (C.1), it is obtained

$$\begin{aligned}
\frac{\mathrm{d}\boldsymbol{z}'_t}{\mathrm{d}t} &= \boldsymbol{f}(\boldsymbol{z}'_t) \\
\frac{\mathrm{d}}{\mathrm{d}t}\left(\boldsymbol{z}_t + \delta\boldsymbol{z}_t\right) &= \boldsymbol{f}(\boldsymbol{z}_t + \delta\boldsymbol{z}_t) \\
&= \boldsymbol{f}(\boldsymbol{z}_t) + \frac{\partial \boldsymbol{f}}{\partial \boldsymbol{z}}(\boldsymbol{z}_t) \cdot \delta\boldsymbol{z}_t + \mathcal{O}(|\delta\boldsymbol{z}_t|^2),
\end{aligned} \tag{C.5}$$

where the definition given in equation (C.4) has been used. Resorting to (C.1), and keeping up to first order terms, this equation simplifies to

$$\frac{\mathrm{d}\delta \boldsymbol{z}_t}{\mathrm{d}t} = \frac{\partial \boldsymbol{f}}{\partial \boldsymbol{z}}(\boldsymbol{z}_t) \cdot \delta \boldsymbol{z}_t. \tag{C.6}$$

Using the result of equation (C.4), and eliminating the time independent $\delta \boldsymbol{z}_i$, it is obtained

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{z}_i} = \frac{\partial \boldsymbol{f}}{\partial \boldsymbol{z}}(\boldsymbol{z}_t) \cdot \frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{z}_i}. \tag{C.7}$$

This is the equation of motion for the stability matrix, $\mathsf{M}_t \equiv \partial \boldsymbol{z}_t / \partial \boldsymbol{z}_i$, in terms of the derivative of the flow vector. Explicitly this matrix is

$$\mathsf{M}_t = \begin{pmatrix} \partial\varphi_t/\partial\varphi_i & \partial\varphi_t/\partial j_i \\ \partial j_t/\partial\varphi_i & \partial j_t/\partial j_i \end{pmatrix}. \tag{C.8}$$

In terms of the Hessian matrix of the Hamiltonian the first matrix on the right hand side of (C.7) can be written as

$$\frac{\partial \boldsymbol{f}}{\partial \boldsymbol{z}} = \begin{pmatrix} \frac{\partial^2 H}{\partial\varphi\partial j} & \frac{\partial^2 H}{\partial j^2} \\ -\frac{\partial^2 H}{\partial\varphi^2} & -\frac{\partial^2 H}{\partial j\partial\varphi} \end{pmatrix} \tag{C.9}$$

Equation (C.7) can be written more compactly as

$$\dot{\mathsf{M}}(t) = \mathsf{J}\mathsf{H}_{z,z}(t)\mathsf{M}(t), \tag{C.10}$$

with J representing the symplectic matrix [20]

$$\mathsf{J} = \begin{pmatrix} 0 & \mathsf{E} \\ -\mathsf{E} & 0 \end{pmatrix}, \tag{C.11}$$

where E is the $n \times n$ identity matrix.

C.1 Calculation of the Stability Matrix in Cartesian Coordinates

Let $\boldsymbol{r} = (r_1, r_2, r_3)$ and $\boldsymbol{p} = (p_1, p_2, p_3)$ be the cartesian position and momentum vectors respectively. The vector $\boldsymbol{x}$ is given by the horizontal concatenation of these two vectors, $\boldsymbol{x} = (\boldsymbol{r}, \boldsymbol{p})$. The flow equation for the constraint system is given by

$$\dot{\boldsymbol{x}} = \boldsymbol{f}(\boldsymbol{x}; \lambda), \tag{C.12}$$

with λ as the Lagrange multiplier

$$\lambda = \frac{1}{2r^2}\left(\boldsymbol{r} \cdot \nabla_{\boldsymbol{r}} V - \frac{p^2}{\mu}\right), \tag{C.13}$$

with μ the reduced mass of the system, and V as the potential energy in cartesian coordinates. In cartesian coordinates the time evolution of the stability matrix follows the equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial \boldsymbol{x}_t}{\partial \boldsymbol{x}_0} = \frac{\partial \boldsymbol{f}}{\partial \boldsymbol{x}}(\boldsymbol{x}_t) \cdot \frac{\partial \boldsymbol{x}_t}{\partial \boldsymbol{x}_0}. \tag{C.14}$$

The derivative of the flow is explicitly given by the matrix

$$\frac{\partial \boldsymbol{f}}{\partial \boldsymbol{x}} = \begin{pmatrix} \mathsf{0}_{3\times3} & \mu^{-1}\mathsf{E}_{3\times3} \\ -\frac{\partial^2 V}{\partial \boldsymbol{r}^2} + 2\left(\lambda \mathsf{E}_{3\times3} + \boldsymbol{r}\nabla_{\boldsymbol{r}}\lambda\right) & 2\boldsymbol{r}\nabla_{\boldsymbol{p}}\lambda \end{pmatrix}, \tag{C.15}$$

with $\boldsymbol{r}\nabla_{\boldsymbol{r}}\lambda$ and $\boldsymbol{r}\nabla_{\boldsymbol{p}}\lambda$ as direct (dyadic) product of the vector $\boldsymbol{r}$ and the gradients of λ with respect to $\boldsymbol{r}$ or $\boldsymbol{p}$ respectively [94].

The stability matrix in a given coordinate system, $\partial \boldsymbol{z}_t / \partial \boldsymbol{z}_0$, is obtained from the cartesian matrix, $\partial \boldsymbol{x}_t / \partial \boldsymbol{x}_0$, using the chain rule

$$\frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{z}_0} = \frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{x}_t} \cdot \frac{\partial \boldsymbol{x}_t}{\partial \boldsymbol{x}_0} \cdot \frac{\partial \boldsymbol{x}_0}{\partial \boldsymbol{z}_0}. \tag{C.16}$$

The first factor is the Jacobian matrix of the transformation $\boldsymbol{z}(\boldsymbol{x})$ evaluated at time t on the trajectory, while the last factor is the Jacobian for the inverse transformation, $\boldsymbol{x}(\boldsymbol{z})$, evaluated at the initial conditions.

Appendix D

Jacobi Elliptic Theta Functions

In terms of the nome η, and ζ, the four Jacobi elliptic theta functions are defined [79]

$$\vartheta_1(\eta,\zeta) \equiv \sum_{n\in\mathbb{Z}} (-1)^{n-1/2} \eta^{(n+1/2)^2} \mathrm{e}^{(2n+1)\mathrm{i}\zeta}, \tag{D.1a}$$

$$\vartheta_2(\eta,\zeta) \equiv \sum_{n\in\mathbb{Z}} \eta^{(n+1/2)^2} \mathrm{e}^{(2n+1)\mathrm{i}\zeta}, \tag{D.1b}$$

$$\vartheta_3(\eta,\zeta) \equiv \sum_{n\in\mathbb{Z}} \eta^{n^2} \mathrm{e}^{2n\mathrm{i}\zeta}, \tag{D.1c}$$

$$\vartheta_4(\eta,\zeta) \equiv \sum_{n\in\mathbb{Z}} (-1)^n \eta^{n^2} \mathrm{e}^{2n\mathrm{i}\zeta}. \tag{D.1d}$$

In terms of trigonometric functions these can be written as

$$\vartheta_1(\eta,\zeta) = 2\sum_{n\in\mathbb{Z}_0^+} (-1)^n \eta^{(n+1/2)^2} \sin\left[(2n+1)\zeta\right], \tag{D.2a}$$

$$\vartheta_2(\eta,\zeta) = 2\sum_{n\in\mathbb{Z}_0^+} \eta^{(n+1/2)^2} \cos\left[(2n+1)\zeta\right], \tag{D.2b}$$

$$\vartheta_3(\eta,\zeta) = 1 + 2\sum_{n\in\mathbb{Z}^+} \eta^{n^2} \cos 2n\zeta, \tag{D.2c}$$

$$\vartheta_4(\eta,\zeta) = 1 + 2\sum_{n\in\mathbb{Z}^+} (-1)^n \eta^{n^2} \cos 2n\zeta, \tag{D.2d}$$

where the sets $\mathbb{Z}_0^+$ and $\mathbb{Z}^+$ contain all the integers $n \geq 0$, and $n > 0$ respectively.

From equations (D.2) it is easy to find that

$$\vartheta_4(\eta,\zeta) = \vartheta_3(\eta,\zeta+\pi/2), \tag{D.3a}$$

$$\vartheta_3(\eta,\zeta) = \vartheta_3(\eta,\zeta+\pi). \tag{D.3b}$$

In equations (D.1) it is seen that the ϑ_i functions can be evaluated at $\eta = 0$

producing

$$\vartheta_1(\eta,\zeta) = 0, \tag{D.4a}$$

$$\vartheta_2(\eta,\zeta) = 0, \tag{D.4b}$$

$$\vartheta_3(\eta,\zeta) = 1, \tag{D.4c}$$

$$\vartheta_4(\eta,\zeta) = 1. \tag{D.4d}$$

Finally for $\zeta = 0$

$$\vartheta_1(\eta,0) = 0, \tag{D.5a}$$

$$\vartheta_2(\eta,0) = \sum_{n\in\mathbb{Z}} \eta^{(n+1/2)^2}, \tag{D.5b}$$

$$\vartheta_3(\eta,0) = \sum_{n\in\mathbb{Z}} \eta^{n^2}, \tag{D.5c}$$

$$\vartheta_4(\eta,0) = \sum_{n\in\mathbb{Z}} (-1)^n \eta^{n^2}. \tag{D.5d}$$

Appendix E

Matrix Element $(z_t''|\cos^\alpha\phi\,|z_t')$ for Periodic Coherent States

For α=1 equation (3.85) reads

$$\begin{aligned}\langle\cos q_j\rangle_\psi(t) &= (z_0|\,U_{HK}^\dagger(t,0)\cos q_j U_{HK}(t,0)\,|z_0)\\ &= \frac{1}{(2\pi\hbar)^2}\int_0^{2\pi}\mathrm{d}\phi_0''\int_{\mathbb{R}}\mathrm{d}j_0''\int_0^{2\pi}\mathrm{d}\phi_0'\int_{\mathbb{R}}\mathrm{d}j_0' f(t;z_0,z_0',z_0''),\end{aligned} \tag{E.1}$$

with

$$f(t;z_0,z_0',z_0'') = (z_0|z_0'')\,(z_0'|z_0)\,C_t(z_0'')^* C_t(z_0')\exp\left[(\mathrm{i}/\hbar)\left(S_t(z_0')-S_t(z_0'')\right)\right](z_t''|\cos\phi\,|z_t')\,. \tag{E.2}$$

The last term is given explicitly by

$$\begin{aligned}(z_t''|\cos\phi\,|z_t') &= \sum_{n,m\in\mathbb{Z}}\int_0^{2\pi}\mathrm{d}\phi\,\langle j_t'',\phi_t''+2n\pi|\phi\rangle\cos\phi\,\langle\phi|j_t',\phi_t'+2m\pi\rangle\\ &= \sum_{n\in\mathbb{Z}}\int_{\mathbb{R}}\mathrm{d}q\,\langle j_t'',\phi_t''+2n\pi|q\rangle\cos q\,\langle q|j_t'\phi_t'\rangle\,.\end{aligned} \tag{E.3}$$

Expressing the $\cos q$ in exponential form and using equation (3.50) the integral splits in two terms

$$(z_t''|\cos\phi\,|z_t') = \left(\frac{\gamma}{4\pi}\right)^{1/2}\sum_{n\in\mathbb{Z}}\int_{\mathbb{R}}\mathrm{d}q\left\{\exp\left[g_{1n+}(q,z_t',z_t'')\right]+\exp\left[g_{1n-}(q,z_t',z_t'')\right]\right\}, \tag{E.4}$$

with the exponents

$$g_{1n\pm} = -\frac{\gamma}{2}\left[(q-\phi_t')^2+(q-\phi_t''-2n\pi)^2\right]+\frac{\mathrm{i}}{\hbar}\left[j_t'(q-\phi_t')-j_t''(q-\phi_t''-2n\pi)\pm\hbar q\right]. \tag{E.5}$$

Completing square for the q and integrating produces

$$(z_t''|\cos\phi\,|z_t') = \frac{1}{2}\left\{\vartheta_3(\eta,\zeta_{1+})\,\mathrm{e}^{G_{1+}(z_t',z_t'')}+\vartheta_3(\eta,\zeta_{1-})\,\mathrm{e}^{G_{1-}(z_t',z_t'')}\right\}, \tag{E.6}$$

where

$$G_{1\pm}(z_t', z_t'') = -\frac{(\Delta j)^2}{4\gamma\hbar^2} - \frac{\gamma(\Delta q)^2}{4} + \frac{\pm\Delta j - \hbar/2}{2\gamma\hbar} + \mathrm{i}\left[\pm\bar{q} + \bar{j}\Delta q/\hbar\right], \tag{E.7}$$

$\Delta q = q_t'' - q_t'$, $\Delta j = j_t'' - j_t'$, $\bar{q} = (q_t'' + q_t')/2$, $\bar{j} = (j_t'' + j_t')/2$, $\eta = \exp(-\gamma\pi^2)$, and

$$\begin{aligned} \zeta_{1\pm} &= \frac{\pi}{2\hbar}\left[2\bar{j} + \mathrm{i}\hbar\gamma\Delta q \pm \hbar\right] \\ &= \zeta \pm \tfrac{\pi}{2}. \end{aligned}$$

Comparing equation (E.7) with the overlap formula (3.61), and using the identity $\vartheta_4(\eta,\zeta) = \vartheta_3(\eta,\zeta \pm \pi/2)$ is finally obtained

$$(z_t''|\cos\phi\,|z_t') = \mathrm{e}^{-1/4\gamma}\frac{\vartheta_4(\eta,\zeta)}{\vartheta_3(\eta,\zeta)}\,(z_t''|z_t')\cosh\left(\frac{\Delta j}{2\gamma\hbar} + \mathrm{i}\bar{q}\right). \tag{E.8}$$

For the alignment the use of $\cos^2\phi = (1+\cos 2\phi)/2$ gives

$$(z_t''|\cos^2\phi\,|z_t') = \frac{1}{4}\left[2\,(z_t''|z_t') + \vartheta_3\left(\eta,\zeta_{2+}\right)\mathrm{e}^{G_{2+}(z_t',z_t'')} + \vartheta_3\left(\eta,\zeta_{2-}\right)\mathrm{e}^{G_{2-}(z_t',z_t'')} +\right], \tag{E.9}$$

with

$$G_{2\pm}(z_t', z_t'') = -\frac{(\Delta j)^2}{4\gamma\hbar^2} - \frac{\gamma(\Delta q)^2}{4} + \frac{\pm\Delta j - \hbar}{\gamma\hbar} + \mathrm{i}\left[\pm 2\bar{q} + \bar{j}\Delta q/\hbar\right], \tag{E.10}$$

and

$$\begin{aligned} \zeta_{2\pm} &= \frac{\pi}{2\hbar}\left[\mathrm{i}\hbar\gamma\Delta q + 2\bar{j} \pm 2\hbar\right] \\ &= \zeta \pm \pi. \end{aligned}$$

Again, comparing (E.10) with the overlap formula (3.61) gives

$$(z_t''|\cos^2\phi\,|z_t') = \frac{\mathrm{e}^{-1/\gamma}}{2}\,(z_t''|z_t')\left[\mathrm{e}^{1/\gamma} + \cosh\left(\frac{\Delta j}{\gamma\hbar} + 2\mathrm{i}\bar{q}\right)\right], \tag{E.11}$$

here the identity $\vartheta_3(\eta,\zeta) = \vartheta_3(\eta,\zeta \pm \pi)$ was used.

Appendix F

Rotation Operators and Wigner D-functions

F.1 Rotation Operators

A rotation about a vector $\boldsymbol{n}$ by an angle θ can be represented using the rotation operator

$$\hat{R}(\theta, \boldsymbol{n}) = \mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}}, \tag{F.1}$$

with $\hat{\boldsymbol{J}}$ as the angular momentum (vector) operator [95]. According to Euler's theorem of the rigid body, this rotation can be written in terms of three consecutive rotations involving the Euler angles α, β, and γ [20, 95]

$$\begin{aligned} \mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}} &= \hat{R}_\gamma \hat{R}_\beta \hat{R}_\alpha \\ &= \mathrm{e}^{-\mathrm{i}\gamma \hat{J}_{z''}} \mathrm{e}^{-\mathrm{i}\beta \hat{J}_{y'}} \mathrm{e}^{-\mathrm{i}\alpha \hat{J}_z}. \end{aligned} \tag{F.2}$$

The first rotation is performed above the lab-fixed z-axis by an angle α; this defines a new frame with axes (x', y', z'). The second rotation is done above the y'-axis by an angle β; this gives a new set of frames (x'', y'', z''). The last one is a rotation above the z''-axis by an angle γ.

The second rotation can be written in terms of rotations on the original frame and the third rotation in terms of rotations on the prime frame

$$\mathrm{e}^{-\mathrm{i}\beta \hat{J}_{y'}} = \mathrm{e}^{-\mathrm{i}\alpha \hat{J}_z} \mathrm{e}^{-\mathrm{i}\beta \hat{J}_y} \mathrm{e}^{\mathrm{i}\alpha \hat{J}_z}, \tag{F.3a}$$

$$\mathrm{e}^{-\mathrm{i}\gamma \hat{J}_{z''}} = \mathrm{e}^{-\mathrm{i}\beta \hat{J}_{y'}} \mathrm{e}^{-\mathrm{i}\gamma \hat{J}_{z'}} \mathrm{e}^{\mathrm{i}\beta \hat{J}_{y'}}. \tag{F.3b}$$

Plugging the second equation into (F.3) produces

$$\mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}} = \mathrm{e}^{-\mathrm{i}\beta \hat{J}_{y'}} \mathrm{e}^{-\mathrm{i}\gamma \hat{J}_{z'}} \mathrm{e}^{-\mathrm{i}\alpha \hat{J}_z}. \tag{F.4}$$

Now inserting the first of equations (F.3) into (F.2) gives

$$\mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}} = \mathrm{e}^{-\mathrm{i}\alpha\hat{J}_z}\mathrm{e}^{-\mathrm{i}\beta\hat{J}_y}\mathrm{e}^{\mathrm{i}\alpha\hat{J}_z}\mathrm{e}^{-\mathrm{i}\gamma\hat{J}_{z'}}\mathrm{e}^{-\mathrm{i}\alpha\hat{J}_z}. \tag{F.5}$$

Writing the second rotation of (F.5) as

$$\mathrm{e}^{-\mathrm{i}\gamma\hat{J}_{z'}} = \mathrm{e}^{-\mathrm{i}\alpha\hat{J}_z}\mathrm{e}^{-\mathrm{i}\gamma\hat{J}_{z'}}\mathrm{e}^{\mathrm{i}\alpha\hat{J}_z}, \tag{F.6}$$

and plugging into (F.5) produces finally

$$\mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}} = \mathrm{e}^{-\mathrm{i}\alpha\hat{J}_z}\mathrm{e}^{-\mathrm{i}\beta\hat{J}_y}\mathrm{e}^{-\mathrm{i}\gamma\hat{J}_z}. \tag{F.7}$$

This is the equation for rotations above the lab-fixed axis only.

F.2 Wigner D-functions

Using the previous result, the action of the rotation operator $\mathrm{e}^{-i\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}}$ on a $|JM\rangle$ state will produce

$$\begin{aligned}
\mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}}|JM\rangle &= \sum_{M'=-J}^{J} |JM'\rangle\langle JM'|\,\mathrm{e}^{-\mathrm{i}\theta \boldsymbol{n}\cdot\hat{\boldsymbol{J}}}|JM\rangle \\
&= \sum_{M'=-J}^{J} |JM'\rangle\langle JM'|\,\mathrm{e}^{-\mathrm{i}\alpha\hat{J}_z}\mathrm{e}^{-\mathrm{i}\beta\hat{J}_y}\mathrm{e}^{-\mathrm{i}\gamma\hat{J}_z}|JM\rangle \\
&= \sum_{M'=-J}^{J} |JM'\rangle\,\mathrm{e}^{-\mathrm{i}\alpha M'}\langle JM'|\,\mathrm{e}^{-\mathrm{i}\beta\hat{J}_y}|JM\rangle\,\mathrm{e}^{-\mathrm{i}\gamma M} \\
&= \sum_{M'=-J}^{J} |JM'\rangle\, d^J_{M'M}(\beta)\mathrm{e}^{-\mathrm{i}(\alpha M'+\gamma M)} \\
&= \sum_{M'=-J}^{J} |JM'\rangle\, D^J_{M'M}(\alpha,\beta,\gamma),
\end{aligned} \tag{F.8}$$

where D^J_{MJ} and d^J_{MJ} are Wigner's D-functions and reduced d-functions [95, 96].

The functions $d^J_{MJ}(\beta)$ are given explicitly by the series [95]

$$\begin{aligned}
d^J_{MJ}(\beta) = {}&[(2J)!(J+M)!(J-M)!]^{1/2} \\
&\times \sum_k \frac{(-1)^k\left(\cos\frac{\beta}{2}\right)^{3J-M-2k}\left(-\sin\frac{\beta}{2}\right)^{M-J+2k}}{(J-M-k)!(2J-k)!(k+M-J)!k!},
\end{aligned} \tag{F.9}$$

with the sum over all the integers k solutions of

$$k \geq 0, \tag{F.10a}$$

$$J - M - k \geq 0, \tag{F.10b}$$

$$2J - k \geq 0, \tag{F.10c}$$

$$k + M - J \geq 0. \tag{F.10d}$$

The only possible solution is $k = J - M$, and equation (F.9) is simplified to

$$d^J_{MJ}(\beta) = \frac{\sqrt{(2J)!}}{\sqrt{(J+M)!(J-M)!}} \left(\sin\frac{\beta}{2}\right)^{J-M} \left(\cos\frac{\beta}{2}\right)^{J+M}. \tag{F.11}$$

The derivative of the $d^J_{MJ}(\beta)$ functions is given by

$$\frac{\mathrm{d}}{\mathrm{d}\beta} d^J_{MJ}(\beta) = \frac{J\cos\beta - M}{\sin\beta} d^J_{MJ}(\beta). \tag{F.12}$$

From equation (F.9) is obtained the useful relation

$$\begin{aligned} d^J_{JJ}(\beta) &= \cos^{2J}(\beta/2) \\ &= d^1_{1,1}(\beta)^J. \end{aligned} \tag{F.13}$$

Appendix G

HK-type Propagator in Rotational Coherent States: Important Derivations

G.1 Equation (3.121)

Using equation (3.111), the time derivative of the rotation operator is

$$\frac{\partial}{\partial t}\hat{R} = -\mathrm{i}\left(\dot{q}_m \hat{J}_z \hat{R} + \dot{\bar{\theta}} \mathrm{e}^{-\mathrm{i}q_m \hat{J}_z} \mathrm{e}^{-\mathrm{i}\bar{\theta}\hat{J}_y} \hat{J}_y \mathrm{e}^{-\mathrm{i}q_j \hat{J}_z} + \dot{q}_j \hat{R}\hat{J}_z\right), \tag{G.1}$$

where $\hat{R} = \hat{R}(\boldsymbol{\alpha})$. With the operator $\hat{J}_y$ in terms of the spherical components of $\hat{\boldsymbol{J}}$ [81],

$$\hat{J}_y = \frac{1}{2\mathrm{i}}(\hat{J}_+ - \hat{J}_-), \tag{G.2}$$

the action of each of the terms of the operator (G.1) on $|JJ\rangle$ gives

$$\hat{J}_z \hat{R} \left|JJ\right\rangle = \sum_M \left|JM\right\rangle M D^J_{MJ}, \tag{G.3a}$$

$$\mathrm{e}^{-\mathrm{i}q_m \hat{J}_z} \mathrm{e}^{-\mathrm{i}\bar{\theta}\hat{J}_y} \hat{J}_y \mathrm{e}^{-\mathrm{i}q_j \hat{J}_z} \left|JJ\right\rangle = -\frac{\sqrt{2J}}{2\mathrm{i}} \mathrm{e}^{-\mathrm{i}q_j} \sum_M \left|JM\right\rangle D^J_{M,J-1} \tag{G.3b}$$

$$\hat{R}\hat{J}_z \left|JJ\right\rangle = J \sum_M \left|JM\right\rangle D^J_{MJ}, \tag{G.3c}$$

with $D^J_{MM'} = D^J_{MM'}(\boldsymbol{\alpha})$. In terms of the Wigner d-functions the second equation is

$$\begin{aligned} -\frac{\sqrt{2J}}{2\mathrm{i}} \mathrm{e}^{-\mathrm{i}q_j} \sum_M \left|JM\right\rangle D^J_{M,J-1} &= -\frac{\sqrt{2J}}{2\mathrm{i}} \sum_M \left|JM\right\rangle \mathrm{e}^{-\mathrm{i}Mq_m} d^J_{M,J-1}(\bar{\theta}) \mathrm{e}^{-\mathrm{i}Jq_j} \\ &= -\frac{\sqrt{2J}}{2\mathrm{i}} \sum_M \left|JM\right\rangle \mathrm{e}^{-\mathrm{i}Mq_m} d^J_{J-1,M}(-\bar{\theta}) \mathrm{e}^{-\mathrm{i}Jq_j}, \end{aligned} \tag{G.4}$$

where the identity $d^J_{MM'}(\beta) = d^J_{M'M}(-\beta)$ was used [95]. The $d^J_{J-1,M}(-\bar{\theta})$ function is explicitly [96, 97]

$$d^J_{J-1,M}(-\bar{\theta}) = \frac{2(J\cos\bar{\theta} - M)}{\sin\bar{\theta}} \frac{\sqrt{(2J-1)!}}{\sqrt{(J+M)!(J-M)!}} \left(\cos\frac{\bar{\theta}}{2}\right)^{J+M} \left(\sin\frac{\bar{\theta}}{2}\right)^{J-M}, \tag{G.5}$$

using the results of appendix F this is rewritten

$$d^J_{J-1,M}(-\bar{\theta}) = \frac{2}{\sqrt{2J}} \frac{J\cos\bar{\theta} - M}{\sin\bar{\theta}} d^J_{MJ}(\bar{\theta}). \tag{G.6}$$

Replacing into equation (G.4) is obtained

$$-\frac{\sqrt{2J}}{2\mathrm{i}} \mathrm{e}^{-\mathrm{i}q_j} \sum_M |JM\rangle\, D^J_{M,J-1} = \mathrm{i} \sum_M |JM\rangle\, \mathrm{e}^{-\mathrm{i}Mq_m} \mathrm{e}^{-\mathrm{i}Jq_j} \frac{J\cos\bar{\theta} - M}{\sin\bar{\theta}} d^J_{MJ}(\bar{\theta}). \tag{G.7}$$

The action of the operator (G.1) on $|JJ\rangle$ is given by

$$\begin{aligned} \frac{\partial}{\partial t}\hat{R}\,|JJ\rangle &= -\mathrm{i} \sum_M |JM\rangle\, D^J_{MJ} \left[\dot{q}_m M + \mathrm{i}\dot{\bar{\theta}} \frac{J\cos\bar{\theta} - M}{\sin\bar{\theta}} + \dot{q}_j J\right] \\ &= \sum_M |JM\rangle\, D^J_{MJ}\left[JF + MG\right], \end{aligned} \tag{G.8}$$

with F and G

$$F = \dot{\bar{\theta}}\cot\bar{\theta} - \mathrm{i}\dot{q}_j, \tag{G.9a}$$

$$G = -\frac{\dot{\bar{\theta}}}{\sin\bar{\theta}} - \mathrm{i}\dot{q}_m. \tag{G.9b}$$

Using $\bar{\theta} = \arccos(m/j)$, $\dot{\bar{\theta}}$ is obtained explicitly

$$\dot{\bar{\theta}} = \frac{1}{j\sqrt{j^2 - m^2}} \left(m\frac{\mathrm{d}j}{\mathrm{d}t} - j\frac{\mathrm{d}m}{\mathrm{d}t}\right). \tag{G.10}$$

Plugging this into equations (G.9) gives

$$F = \frac{m/j}{j^2 - m^2} \left(m\frac{\mathrm{d}j}{\mathrm{d}t} - j\frac{\mathrm{d}m}{\mathrm{d}t}\right) - \mathrm{i}\dot{q}_j, \tag{G.11a}$$

$$G = -\frac{1}{j^2 - m^2} \left(m\frac{\mathrm{d}j}{\mathrm{d}t} - j\frac{\mathrm{d}m}{\mathrm{d}t}\right) - \mathrm{i}\dot{q}_m. \tag{G.11b}$$

G.2 Equation (3.124)

Equation (3.123) is

$$\langle z_t'| \frac{\partial}{\partial \mathrm{t}} |z_t\rangle = \left(\mathrm{i}\dot{\mu} - \frac{1}{2}\frac{\mathrm{d}j}{\mathrm{d}t} \right) \langle z_t'|z_t\rangle + \mathrm{e}^{\mathrm{i}\Delta\mu - \bar{\jmath}} \sum_{J,M} \frac{\tilde{\jmath}^J}{J!} D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) \left[J\bar{F} + MG \right], \tag{G.12}$$

with the functions $\bar{F}$ and G

$$\bar{F} = \frac{1}{2j}\frac{\mathrm{d}j}{\mathrm{d}t} + \frac{m/j}{j^2 - m^2}\left(m\frac{\mathrm{d}j}{\mathrm{d}t} - j\frac{\mathrm{d}m}{\mathrm{d}t} \right) - \mathrm{i}\dot{q}_j, \tag{G.13a}$$

$$G = -\frac{1}{j^2 - m^2}\left(m\frac{\mathrm{d}j}{\mathrm{d}t} - j\frac{\mathrm{d}m}{\mathrm{d}t} \right) - \mathrm{i}\dot{q}_m. \tag{G.13b}$$

For the terms involving a J factor in the summation

$$\begin{aligned}
\sum_{JM} \frac{\tilde{\jmath}^J}{J!} D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) J\bar{F} &= \bar{F} \sum_{M,J=1} \frac{\tilde{\jmath}^J}{(J-1)!} D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) \\
&= \bar{F} \sum_{J=1} \frac{\tilde{\jmath}^J}{(J-1)!} \sum_{M} D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) \\
&= \bar{F} \sum_{J=1} \frac{\tilde{\jmath}^J}{(J-1)!} D^J_{JJ}(\tilde{\boldsymbol{\alpha}}) \\
&= \bar{F} \sum_{J=1} \frac{\tilde{\jmath}^J}{(J-1)!} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})^J \\
&= \tilde{\jmath}\bar{F} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) \sum_{J=0} \frac{\tilde{\jmath}^J}{J!} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})^J \\
&= \tilde{\jmath}\bar{F} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) \exp\left[\tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) \right].
\end{aligned} \tag{G.14}$$

In the second line is used the combined rotation $\hat{R}(\tilde{\boldsymbol{\alpha}}) \equiv \hat{R}(\boldsymbol{\alpha}')^{-1}\hat{R}(\boldsymbol{\alpha})$.

The terms involving a M factor in the sum

$$\begin{aligned}
\sum_{JM} \frac{\tilde{\jmath}^J}{J!} D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) M G &= G \sum_J \frac{\tilde{\jmath}^J}{J!} \sum_M M D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) \\
&= \mathrm{i} G \sum_J \frac{\tilde{\jmath}^J}{J!} \sum_M \frac{\partial}{\partial q_m} D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) \\
&= \mathrm{i} G \sum_J \frac{\tilde{\jmath}^J}{J!} \frac{\partial}{\partial q_m} \sum_M D^J_{MJ}(\boldsymbol{\alpha}')^* D^J_{MJ}(\boldsymbol{\alpha}) \\
&= \mathrm{i} G \sum_J \frac{\tilde{\jmath}^J}{J!} \frac{\partial}{\partial q_m} D^J_{JJ}(\tilde{\boldsymbol{\alpha}}) \\
&= \mathrm{i} G \frac{\partial}{\partial q_m} \sum_J \frac{\tilde{\jmath}^J}{J!} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})^J \\
&= \mathrm{i} G \frac{\partial}{\partial q_m} \exp\left[\tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right] \\
&= \mathrm{i} \tilde{\jmath} G \exp\left[\tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right] \frac{\partial}{\partial q_m} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}).
\end{aligned} \tag{G.15}$$

Putting results (G.14) and (G.15) into (G.12) is obtained

$$\begin{aligned}
\langle z_t'| \frac{\partial}{\partial \mathrm{t}} |z_t\rangle = \left(\mathrm{i}\dot{\mu} - \frac{1}{2}\frac{\mathrm{d}j}{\mathrm{d}t}\right) \langle z_t'|z_t\rangle + \tilde{\jmath} \exp\left[\mathrm{i}\Delta\mu - \bar{\jmath} + \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right] \\
\times \left(\bar{F} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) + \mathrm{i} G \frac{\partial}{\partial q_m} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right).
\end{aligned} \tag{G.16}$$

Finally, using equation (3.117)

$$\langle z_t'| \frac{\partial}{\partial \mathrm{t}} |z_t\rangle = \langle z_t'|z_t\rangle \left[\mathrm{i}\dot{\mu} - \frac{1}{2}\frac{\mathrm{d}j}{\mathrm{d}t} + \tilde{\jmath} \left(\bar{F} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) + \mathrm{i} G \frac{\partial}{\partial q_m} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right)\right]. \tag{G.17}$$

G.3 **Equation** (3.127)

From the derivative of the power series of the exponential function is obtained the equality

$$\begin{aligned}
\mathrm{e}^x &= \sum_{n=0} \frac{n}{n!} x^{n-1}, \\
x\mathrm{e}^x &= \sum_{n=0} \frac{n}{n!} x^n;
\end{aligned} \tag{G.18}$$

for the second derivative

$$\begin{aligned} \mathrm{e}^x &= \sum_{n=0} \frac{n(n-1)}{n!} x^{n-2}, \\ x^2\mathrm{e}^x &= \sum_{n=0} \frac{n(n-1)}{n!} x^{n}, \\ x^2\mathrm{e}^x &= \sum_{n=0} \frac{n^2}{n!} x^{n} - x\mathrm{e}^x, \\ x(x+1)\mathrm{e}^x &= \sum_{n=0} \frac{n^2}{n!} x^{n}, \end{aligned} \tag{G.19}$$

where the result (G.18) was used to get the third line. Combining the last lines of (G.18) and (G.19) gives

$$\sum_{n=0} \frac{n(n+1)}{n!} x^n = x(x+2)\mathrm{e}^x. \tag{G.20}$$

Equation (3.126) reads

$$\langle z_t'| \hat{J}^2 |z_t\rangle = \mathrm{e}^{\mathrm{i}\Delta\mu - \bar{\jmath}} \sum_J \frac{J(J+1)}{J!} \left[\tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right]^J. \tag{G.21}$$

Using the result (G.20) with $x = \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})$ in equation (G.21) gives

$$\langle z_t'| \hat{J}^2 |z_t\rangle = \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) \left[2 + \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right] \exp\left[\mathrm{i}\Delta\mu - \bar{\jmath} + \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right]. \tag{G.22}$$

Finally resorting to (3.117) this is simplified to

$$\langle z_t'| \hat{J}^2 |z_t\rangle = \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}}) \left[2 + \tilde{\jmath} D^1_{1,1}(\tilde{\boldsymbol{\alpha}})\right] \langle z_t'|z_t\rangle. \tag{G.23}$$

Appendix H

Symmetric Top

H.1 Gradients with Respect to $\boldsymbol{j}$ and $\tilde{\mathsf{C}}$

Only the non-zero components of the gradients are displayed.

H.1.1 Euler Angles and Conjugate Momenta

Euler angles in terms of $\boldsymbol{j}$ and $\tilde{\mathsf{C}}$

$$\theta = \arccos \tilde{C}_{33}, \tag{H.1a}$$

$$\phi = \arctan \frac{\tilde{C}_{31}\tilde{C}_{12} - \tilde{C}_{32}\tilde{C}_{11}}{\tilde{C}_{13}}, \tag{H.1b}$$

$$\psi = \arctan \frac{\tilde{C}_{32}}{-\tilde{C}_{31}}, \tag{H.1c}$$

$$p_\theta = \frac{\tilde{C}_{32} j_1 - \tilde{C}_{31} j_2}{(1 - \tilde{C}_{33}^2)^{1/2}}, \tag{H.1d}$$

$$p_\phi = \tilde{C}_{31} j_1 + \tilde{C}_{32} j_2 + \tilde{C}_{33} j_3, \tag{H.1e}$$

$$p_\psi = j_3. \tag{H.1f}$$

Gradient of θ

$$\frac{\partial \theta}{\partial \tilde{C}_{33}} = -\left(1 - \tilde{C}_{33}^2\right)^{-1/2}. \tag{H.2}$$

Gradient of ϕ

$$\frac{\partial \phi}{\partial \tilde{C}_{11}} = -\frac{\tilde{C}_{32}\tilde{C}_{13}}{\tilde{C}_{33}^2}, \tag{H.3a}$$

$$\frac{\partial \phi}{\partial \tilde{C}_{12}} = \frac{\tilde{C}_{31}\tilde{C}_{13}}{\tilde{C}_{33}^2}, \tag{H.3b}$$

$$\frac{\partial \phi}{\partial \tilde{C}_{13}} = -\frac{\tilde{C}_{31}\tilde{C}_{12} - \tilde{C}_{32}\tilde{C}_{11}}{\tilde{C}_{33}^2}, \tag{H.3c}$$

$$\frac{\partial \phi}{\partial \tilde{C}_{31}} = \frac{\tilde{C}_{12}\tilde{C}_{13}}{\tilde{C}_{33}^2}, \tag{H.3d}$$

$$\frac{\partial \phi}{\partial \tilde{C}_{32}} = -\frac{\tilde{C}_{11}\tilde{C}_{32}}{\tilde{C}_{33}^2}. \tag{H.3e}$$

Gradient of ψ

$$\frac{\partial \psi}{\partial \tilde{C}_{31}} = \frac{\tilde{C}_{32}}{1 - \tilde{C}_{33}^2}, \tag{H.4a}$$

$$\frac{\partial \psi}{\partial \tilde{C}_{32}} = -\frac{\tilde{C}_{31}}{1 - \tilde{C}_{33}^2}. \tag{H.4b}$$

Gradient of p_θ

$$\frac{\partial p_\theta}{\partial j_1} = \frac{\tilde{C}_{32}}{(\tilde{C}_{31}^2 + \tilde{C}_{32}^2)^{1/2}}, \tag{H.5a}$$

$$\frac{\partial p_\theta}{\partial j_1} = -\frac{\tilde{C}_{31}}{(\tilde{C}_{31}^2 + \tilde{C}_{32}^2)^{1/2}}, \tag{H.5b}$$

$$\frac{\partial p_\theta}{\partial \tilde{C}_{31}} = -\frac{\tilde{C}_{32}(\tilde{C}_{31}j_1 + \tilde{C}_{32}j_2)}{(\tilde{C}_{31}^2 + \tilde{C}_{32}^2)^{3/2}}, \tag{H.5c}$$

$$\frac{\partial p_\theta}{\partial \tilde{C}_{31}} = \frac{\tilde{C}_{31}(\tilde{C}_{31}j_1 + \tilde{C}_{32}j_2)}{(\tilde{C}_{31}^2 + \tilde{C}_{32}^2)^{3/2}}. \tag{H.5d}$$

Gradient of p_ϕ

$$\frac{\partial p_\phi}{\partial j_1} = \tilde{C}_{31}, \tag{H.6a}$$
$$\frac{\partial p_\phi}{\partial j_2} = \tilde{C}_{32}, \tag{H.6b}$$
$$\frac{\partial p_\phi}{\partial j_3} = \tilde{C}_{33}, \tag{H.6c}$$
$$\frac{\partial p_\phi}{\partial \tilde{C}_{31}} = j_1, \tag{H.6d}$$
$$\frac{\partial p_\phi}{\partial \tilde{C}_{32}} = j_2, \tag{H.6e}$$
$$\frac{\partial p_\phi}{\partial \tilde{C}_{33}} = j_3. \tag{H.6f}$$

Gradient of p_ψ

$$\frac{\partial p_\psi}{\partial j_3} = 1. \tag{H.7}$$

H.1.2 Action-Angle Variables

Action-Angle Variables in terms of $\boldsymbol{j}$ and $\tilde{\mathsf{C}}$

$$q_j = \arctan \frac{j(j_2\tilde{C}_{31} - j_1\tilde{C}_{32})}{j^2\tilde{C}_{33} - mk}, \tag{H.8a}$$
$$q_m = \arctan \frac{\tilde{\boldsymbol{C}}_2 \cdot \boldsymbol{j}}{\tilde{\boldsymbol{C}}_1 \cdot \boldsymbol{j}}, \tag{H.8b}$$
$$q_k = \arctan \frac{j_2}{j_1}, \tag{H.8c}$$
$$j = \sqrt{j_1^2 + j_2^2 + j_3^3}, \tag{H.8d}$$
$$m = \tilde{\boldsymbol{C}}_3 \cdot \boldsymbol{j}, \tag{H.8e}$$
$$k = j_3, \tag{H.8f}$$

with

$$\tilde{\boldsymbol{C}}_1 = \left(\tilde{C}_{11}, \tilde{C}_{12}, \tilde{C}_{13}\right), \tag{H.9a}$$

$$\tilde{\boldsymbol{C}}_3 = \left(\tilde{C}_{31}, \tilde{C}_{32}, \tilde{C}_{33}\right), \tag{H.9b}$$

$$\tilde{\boldsymbol{C}}_2 = \tilde{\boldsymbol{C}}_3 \times \tilde{\boldsymbol{C}}_1. \tag{H.9c}$$

Gradient of q_j

For the $\arctan u(t)$,

$$\frac{\mathrm{d}}{\mathrm{d}t} \arctan u(t) = \frac{1}{1+u^2} u'(t). \tag{H.10}$$

With

$$U_j = \frac{j(j_2\tilde{C}_{31} - j_1\tilde{C}_{32})}{j^2\tilde{C}_{33} - mk}, \tag{H.11}$$

the common factor of the derivatives is

$$\frac{1}{1+U_j^2} = \frac{u_j^2}{(j^2-m^2)(j^2-k^2)}, \tag{H.12}$$

with the definition $u = j^2\tilde{C}_{33} - mk$.

The gradient of U_j has components

$$\frac{\partial U_j}{\partial j_1} = u_j^{-2}\left[\left(\frac{j_1}{j}\tau_{33} - j\tilde{C}_{32}\right)u_j - \left(2j_1\tilde{C}_{33} - k\tilde{C}_{31}\right)j\tau_{33}\right], \tag{H.13a}$$

$$\frac{\partial U_j}{\partial j_2} = u_j^{-2}\left[\left(\frac{j_2}{j}\tau_{33} + j\tilde{C}_{31}\right)u_j - \left(2j_2\tilde{C}_{33} - k\tilde{C}_{32}\right)j\tau_{33}\right], \tag{H.13b}$$

$$\frac{\partial U_j}{\partial j_3} = u_j^{-2}\left[\frac{j_3}{j}\tau_{33}u_j - \left(k\tilde{C}_{33} - m\right)j\tau_{33}\right], \tag{H.13c}$$

$$\frac{\partial U_j}{\partial \tilde{C}_{31}} = u_j^{-2}\left[jj_2u_j + kjj_1\tau_{33}\right], \tag{H.13d}$$

$$\frac{\partial U_j}{\partial \tilde{C}_{32}} = u_j^{-2}\left[-jj_1u_j + kjj_2\tau_{33}\right], \tag{H.13e}$$

$$\frac{\partial U_j}{\partial \tilde{C}_{33}} = u_j^{-2}\left[-j\left(j^2-k^2\right)\tau_{33}\right], \tag{H.13f}$$

where τ_{ij} is the j-th component of the $\boldsymbol{\tau}_i$ vector

$$\boldsymbol{\tau}_i = \tilde{\boldsymbol{C}}_i \times \boldsymbol{j}. \tag{H.14}$$

Gradient of q_m

Again equation (H.10) is necessary, but now with

$$U_m = \frac{\tilde{\boldsymbol{C}}_2 \cdot \boldsymbol{j}}{\tilde{\boldsymbol{C}}_1 \cdot \boldsymbol{j}}, \tag{H.15}$$

which gives the common factor

$$\frac{1}{1+U_m^2} = \frac{j^2 - m^2}{(\tilde{\boldsymbol{C}}_1 \cdot \boldsymbol{j})^2}. \tag{H.16}$$

The gradient of U_m has components

$$\frac{\partial U_m}{\partial j_1} = -\frac{\tau_{13}}{(\tilde{\boldsymbol{C}}_1 \cdot \boldsymbol{j})^2}, \tag{H.17a}$$
$$\frac{\partial U_m}{\partial j_2} = -\frac{\tau_{23}}{(\tilde{\boldsymbol{C}}_1 \cdot \boldsymbol{j})^2}, \tag{H.17b}$$
$$\frac{\partial U_m}{\partial j_3} = -\frac{\tau_{33}}{(\tilde{\boldsymbol{C}}_1 \cdot \boldsymbol{j})^2}, \tag{H.17c}$$
$$\frac{\partial U_m}{\partial \tilde{C}_{11}} = \frac{(-j_1\tau_{31} - j_1 j_2)}{j_1^2}, \tag{H.17d}$$
$$\frac{\partial U_m}{\partial \tilde{C}_{12}} = \frac{(-j_1\tau_{32} - j_2^2)}{j_1^2}, \tag{H.17e}$$
$$\frac{\partial U_m}{\partial \tilde{C}_{13}} = \frac{(-j_1\tau_{33} - j_2 j_3)}{j_1^2}, \tag{H.17f}$$
$$\frac{\partial U_m}{\partial \tilde{C}_{31}} = \frac{\tau_{11}}{j_1}, \tag{H.17g}$$
$$\frac{\partial U_m}{\partial \tilde{C}_{32}} = \frac{\tau_{12}}{j_1}, \tag{H.17h}$$
$$\frac{\partial U_m}{\partial \tilde{C}_{33}} = \frac{\tau_{13}}{j_1}. \tag{H.17i}$$

Gradient of q_k

$$\frac{\partial q_k}{\partial j_1} = -\frac{j_2}{j^2 - k^2}, \tag{H.18a}$$
$$\frac{\partial q_k}{\partial j_2} = \frac{j_1}{j^2 - k^2}. \tag{H.18b}$$

Gradient of j

$$\frac{\partial j}{\partial j_1} = \frac{j_1}{j}, \tag{H.19a}$$
$$\frac{\partial j}{\partial j_2} = \frac{j_2}{j}, \tag{H.19b}$$
$$\frac{\partial j}{\partial j_3} = \frac{j_3}{j}. \tag{H.19c}$$

Gradient of m

Given by equation (H.6) with $p_\phi = m$.

Gradient of k

Given by equation (H.7) with $p_\psi = k$.

Appendix I

Constant m Sampling for Asymmetric Tops in Collinear Fields

In the body fixed angular momentum space the intersecting surfaces are the plane

$$\begin{aligned} m &= \tilde{\boldsymbol{C}}_3 \cdot \boldsymbol{j} \\ &= \tilde{C}_{31} j_1 + \tilde{C}_{32} j_2 + \tilde{C}_{33} j_3, \end{aligned} \tag{I.1}$$

and the ellipsoid

$$2\left[E - V(\tilde{\boldsymbol{C}}_3)\right] = (\mathsf{I}^{-1}\boldsymbol{j}) \cdot \boldsymbol{j}. \tag{I.2}$$

Since m is constant, it is convenient to obtain the intersection of these surfaces in terms of the space fixed components of the angular momentum. Body and space fixed angular momentum vectors are related by

$$\boldsymbol{j} = \mathsf{C}\boldsymbol{l}. \tag{I.3}$$

Using this into the ellipsoid equation gives

$$2\left[E - V(\tilde{\boldsymbol{C}}_3)\right] = (\mathsf{I}^{-1}\mathsf{C}\boldsymbol{l}) \cdot (\mathsf{C}\boldsymbol{l}). \tag{I.4}$$

The matrix $\mathsf{I}^{-1}\mathsf{C}$ can be written as the row concatenation of the vectors

$$\begin{aligned} \boldsymbol{c}_i &= I_i^{-1}\boldsymbol{C}_i \\ &= I_i^{-1}\left(C_{i1}, C_{i2}, C_{i3}\right), \end{aligned} \tag{I.5}$$

with $i = 1, 2, 3$. The product $\mathsf{C}\boldsymbol{l}$ gives the vector $(\boldsymbol{C}_1 \cdot \boldsymbol{l}, \boldsymbol{C}_2 \cdot \boldsymbol{l}, \boldsymbol{C}_3 \cdot \boldsymbol{l})$, and using equation (I.5), the product $(\mathsf{I}^{-1}\mathsf{C}\boldsymbol{l})$ is the vector $(\boldsymbol{c}_1 \cdot \boldsymbol{l}, \boldsymbol{c}_2 \cdot \boldsymbol{l}, \boldsymbol{c}_3 \cdot \boldsymbol{l})$. These two vectors in (I.4) produce

$$\begin{aligned}2\left[E - V(\tilde{\boldsymbol{C}}_3)\right] &= c_{ij}C_{ik}l_jl_k \\ &= I_i^{-1}C_{ij}C_{ik}l_jl_k \\ &= I_i^{-1}\tilde{C}_{ji}\tilde{C}_{ki}l_jl_k \\ &= (\mathsf{I}^{-1}\tilde{\boldsymbol{C}}_j)\cdot\tilde{\boldsymbol{C}}_kl_jl_k \\ &= A_{jk}l_jl_k,\end{aligned} \tag{I.6}$$

with summation over the repeated indices implied, and $A_{jk} \equiv (\mathsf{I}^{-1}\tilde{\boldsymbol{C}}_j)\cdot\tilde{\boldsymbol{C}}_k$. In the $\boldsymbol{l}$ space, this equation, together with $l_3 = m$, define an ellipse lying on the $l_3 = m$ plane. This summation can be rearranged to get

$$2\left[E - V(\tilde{\boldsymbol{C}}_3)\right] = A_{33}m^2 + 2mA_{3j}l_j + A_{jk}l_jl_k, \tag{I.7}$$

where $j, k = 1, 2$, summation over repeated indices implied, and the symmetry $A_{ij} = A_{ji}$ was used. This shows that the center of the ellipse is not on the l_3 axis, and the presence of l_1l_2 terms implies that the principal axes of the ellipse are not parallel to the (l_1, l_2) axes. The principal axes of the ellipse define a frame (L_1, L_2) in which there would not be L_1L_2 terms, this frame is related to the axes (l_1, l_2) by a rotation

$$\begin{pmatrix} l_1 \\ l_2 \end{pmatrix} = \begin{pmatrix} \cos\Theta & -\sin\Theta \\ \sin\Theta & \cos\Theta \end{pmatrix}\begin{pmatrix} L_1 \\ L_2 \end{pmatrix}. \tag{I.8}$$

After substituting into (I.7) it is clear that only the last term of this equation could involve the product L_1L_2, in the rotated system this term after simplifying gives

$$\begin{aligned}A_{jk}l_jl_k = &\left(A_{11}\cos^2\Theta + A_{12}\sin 2\Theta + A_{22}\sin^2\Theta\right)L_1^2 \\ &+ \left(A_{22}\cos^2\Theta - A_{12}\sin 2\Theta + A_{11}\sin^2\Theta\right)L_2^2 \\ &+ \left[2A_{12}\cos 2\Theta + (A_{22} - A_{11})\sin 2\Theta\right]L_1L_2.\end{aligned} \tag{I.9}$$

The last term will vanish for Θ such that

$$\cot 2\Theta = \frac{A_{11} - A_{22}}{2A_{12}}. \tag{I.10}$$

With the ellipse correctly aligned now it is necessary to make its center to coincide with the l_3 axis. In terms of (L_1, L_2), the second term of the LHS in equation (I.7) is

$$2mA_{3j}l_j = 2m\left[\left(A_{13}\cos\Theta + A_{23}\sin\Theta\right)L_1 + \left(A_{23}\cos\Theta - A_{13}\sin\Theta\right)L_2\right]. \tag{I.11}$$

Now Equation (I.7) can be written as

$$B_{11}L_1^2 + B_{22}L_2^2 + B_{31}L_1 + B_{32}L_2 + B_{33} = 0, \tag{I.12}$$

with

$$B_{11} = A_{11}\cos^2\Theta + A_{12}\sin 2\Theta + A_{22}\sin^2\Theta, \tag{I.13a}$$

$$B_{22} = A_{22}\cos^2\Theta - A_{12}\sin 2\Theta + A_{11}\sin^2\Theta, \tag{I.13b}$$

$$B_{31} = A_{13}\cos\Theta + A_{23}\sin\Theta, \tag{I.13c}$$

$$B_{32} = A_{23}\cos\Theta - A_{13}\sin\Theta, \tag{I.13d}$$

$$B_{33} = A_{33}m^2 - 2\left[E - V(\tilde{\boldsymbol{C}}_3)\right]. \tag{I.13e}$$

Completing squares in equation (I.12) gives

$$\rho_1^2\left(L_1 + \Lambda_1\right)^2 + \rho_2^2\left(L_2 + \Lambda_2\right)^2 = 1, \tag{I.14}$$

with

$$\Lambda_1 = \frac{B_{31}}{2B_{11}}, \tag{I.15a}$$

$$\Lambda_2 = \frac{B_{32}}{2B_{22}}, \tag{I.15b}$$

$$\rho_1 = \left[B_{11} \left(\frac{B_{31}^2}{2B_{11}} + \frac{B_{32}^2}{2B_{22}} - B_{33} \right)^{-1} \right]^{1/2}, \tag{I.15c}$$

$$\rho_2 = \left[B_{22} \left(\frac{B_{31}^2}{2B_{11}} + \frac{B_{32}^2}{2B_{22}} - B_{33} \right)^{-1} \right]^{1/2}. \tag{I.15d}$$

Defining $\lambda_1 = L_1 + \Lambda_1$ and $\lambda_2 = L_1 + \Lambda_2$, casts equation (I.14) into

$$\rho_1^2 \lambda_1^2 + \rho_2^2 \lambda_2^2 = 1, \tag{I.16}$$

which allows the parametrization

$$\lambda_1 = \cos \vartheta, \tag{I.17a}$$

$$\lambda_2 = \cos \vartheta. \tag{I.17b}$$

The sampling is done fixing m and $\tilde{\boldsymbol{C}}_3$, then finding Θ using equation (I.10), which allows to obtain the coefficients of equation (I.13). These coefficients determine completely ρ_i and Λ_i. Now, given ϑ, the two λ_i are obtained using equation (I.17); the λ_i together with the Λ_i give the L_i from their definition. With the two L_i, and the angle Θ, it is possible to obtained the space fixed components of the angular momentum, l_i, which give via the Euler rotations the j_i that are used finally in the numerical integration.

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