Chapter 4

Collective Quantum Dynamics of Two and Three Interacting Heavy-Ions

In the fusion of heavy-ions the nuclei can stick together for a time of 10^{-19} or longer forming thus quasi-molecular states. Since such times are comparable or even much longer than the life time of collective states, it is then interesting to study the spectroscopy of such systems.

4.1 Geometrical Hamiltonian of Dinuclear Systems

4.1.1 Coordinate systems

For a dinuclear molecule we consider the following 7 degrees of freedom: $\widetilde{\omega}_i = (\widetilde{\alpha}_i, \widetilde{\beta}_i, \widetilde{\gamma}_i)$, $(i = 1, 2)$ the Euler angles of each nucleus; $\boldsymbol{R} = (R, \Theta, \Phi)$ defining the relative vector as displayed in Fig.3.4 for two deformed nuclei. The axis z' is defined as the molecular axis of the whole system. Then the intrinsic axes of each deformed nucleus are referred to this molecular system. The angles $\Theta = \theta_2$ and $\Phi = \theta_1$ are giving the orientation of the z'-axis and by the above definition also the orientation of $\boldsymbol{R}$ for that system. Since the system of principal axes of the nuclei does not necessarily coincide with the molecular one, a new set of Euler angles, $\widetilde{\omega}_i = (\alpha_i, \beta_i, \gamma_i), \quad (i = 1, 2)$, must be introduced in order to describe the orientation of the interacting nuclei in the molecular frame by means of the following transformation:

$$\mathcal{R}(\omega_i) = \mathcal{R}(\Phi, \Theta, 0)\mathcal{R}(\widetilde{\omega}_i) \tag{4.1}$$

where $\mathcal{R}$ denote Euler rotations. The variables γ_i are set to zero because we suppose that no quantum rotations around the symmetry axes of the nuclei are allowed. Further the variables α_1 and α_2 are combined into the variables $\theta_3 = (\alpha_1 + \alpha_2)/2$, which describes rotations of the quasi-molecular system around the molecular axis z', and $\alpha = (\alpha_1 - \alpha_2)/2$ which describes twisting of the two nuclei with respect to the same axis. Then the new set of 7 degrees of freedom is

$$q_i = (\theta_1, \theta_2, \theta_3, \alpha, R, \beta_1, \beta_2) \tag{4.2}$$

It is possible to define a different molecular frame, whose axes are the principal axes of the total system. However such a system proves to be inconvenient in analyses of decay

properties. Its z'-axis does not necessarily coincide with the direction of the relative vector $\boldsymbol{R}$ and consequently a complicated transformation must be done between the Euler angles of the total system and the angular variables of the vector $\boldsymbol{R}$.

4.1.2 The classical kinetic energy and its quantization

In the laboratory system the classical kinetic energy of the two center-of-masses motions and the rotational motions are

$$T_{\text{cl}} = \frac{1}{2}m_1\dot{\boldsymbol{r}}_1^2 + \frac{1}{2}m_2\dot{\boldsymbol{r}}_2^2 + \frac{1}{2}{}^t\boldsymbol{\omega}_1\boldsymbol{\mathcal{J}}_1\boldsymbol{\omega}_1 + \frac{1}{2}{}^t\boldsymbol{\omega}_2\boldsymbol{\mathcal{J}}_2\boldsymbol{\omega}_2 \tag{4.3}$$

where m_1 and m_2 are the masses of the two nuclei, and $\boldsymbol{r}_{1,2}$ denote the vector positions of their c.m.. The vectors $\boldsymbol{\omega}_{1,2}$ are the angular velocities of the rotational motions of the two nuclei. The inertia tensors $\boldsymbol{\mathcal{J}}_1$ and $\boldsymbol{\mathcal{J}}_2$ are defined in the coordinate frame of their principal axes, i.e. they are diagonal.

In order to separate the c.m. motion, the c.m. position vector $\boldsymbol{R}_{\text{c.m.}}$ and the relative vector between the two nuclear c.m. $\boldsymbol{R}$ are introduced:

$$\boldsymbol{R}_{\text{c.m.}} = \frac{m_1\boldsymbol{r}_1 + m_2\boldsymbol{r}_2}{m_1 + m_2} \tag{4.4}$$

$$\boldsymbol{R} = \boldsymbol{r}_2 - \boldsymbol{r}_1 \tag{4.5}$$

Then the c.m. energies of the two nuclei can be written as the c.m. energy of the total system and the relative energy between the two nuclei

$$\frac{1}{2}m_1\dot{\boldsymbol{r}}_1^2 + \frac{1}{2}m_2\dot{\boldsymbol{r}}_2^2 = \frac{1}{2}M\dot{\boldsymbol{R}}_{\text{c.m.}}^2 + \frac{1}{2}\mu\dot{\boldsymbol{R}}^2 \tag{4.6}$$

where $M = m_1 + m_2$ is the total mass and $\mu = m_1m_2/(m_1 + m_2)$ the relative mass. The relative energy in its turn can be rewritten as the sum of a radial(translational) motion and a relative(orbital) rotation. For that we write $\boldsymbol{R}$ in terms of spherical components

$$R_m = \sum_{m'=-1}^{+1} D^1_{mm'}{}^*(\theta_j)\bar{R}_{m'} \tag{4.7}$$

The intrinsic components $\bar{R}_{m'}$ are by definition

$$\bar{R}_0 = R, \quad , \bar{R}_{+1} = \bar{R}_{-1} = 0 \tag{4.8}$$

i.e. $\boldsymbol{R}$ lies along the molecular z'-axis. Then (4.7) reduces to

$$R_m = D^1_{m0}{}^*(\theta_j)R \tag{4.9}$$

Calculating the time derivative of $\boldsymbol{R}$ we get for the relative energy

$$\begin{aligned}\frac{1}{2}\mu\dot{\boldsymbol{R}}^2 &= \frac{1}{2}\mu\dot{R}^2 + \frac{1}{2}\mu R^2(\dot{\theta}_2^2 + \dot{\theta}_1^2\sin\theta_2^2) \\ &= \frac{1}{2}\mu\dot{R}^2 + \frac{1}{2}{}^t\boldsymbol{\omega}'\boldsymbol{\mathcal{J}}_{\text{rel}}\boldsymbol{\omega}'\end{aligned} \tag{4.10}$$

where

$$\boldsymbol{\mathcal{J}}_{\text{rel}} = \begin{pmatrix} \mu R^2 & 0 & 0 \\ 0 & \mu R^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{4.11}$$

Thus, the total classical energy, ommiting the c.m. energy of the total system, is put in the new form

$$T_{\text{cl}} = \frac{1}{2}\mu\dot{R}^2 + \frac{1}{2}{}^t\boldsymbol{\omega}'\boldsymbol{\mathcal{J}}_{\text{rel}}\boldsymbol{\omega}' + \frac{1}{2}{}^t\boldsymbol{\omega}_1\boldsymbol{\mathcal{J}}_1\boldsymbol{\omega}_1 + \frac{1}{2}{}^t\boldsymbol{\omega}_2\boldsymbol{\mathcal{J}}_2\boldsymbol{\omega}_2 \tag{4.12}$$

The angular velocities of the constituent nuclei $\boldsymbol{\omega}_i$ are referred to the laboratory system. It is neccesary to express them in the molecular coordinate system, i.e. in terms of the angular velocity of the molecular frame $\boldsymbol{\omega}'$ and those referred to the molecular frame $\boldsymbol{\omega}_i''$. As shown in [1] the angular velocities $\boldsymbol{\omega}'$ are expressed in terms of the time-derivative of the Euler angles as follows

$$\omega_j' = \sum_{l=1}^{3} V_{jl}(\theta_2, \theta_3)\frac{d\theta_l}{dt} \tag{4.13}$$

where the transformation matrix V is given by

$$V_{jl}(\theta_2, \theta_3) = \begin{pmatrix} -\sin\theta_2\cos\theta_3 & \sin\theta_3 & 0 \\ \sin\theta_2\sin\theta_3 & \cos\theta_3 & 0 \\ \cos\theta_2 & 0 & 1 \end{pmatrix} \tag{4.14}$$

For the angular velocities of the constituent nuclei referred to the molecular frame, Uegaki and Abe [2] obtained

$$\omega_j'' = \sum_{l=1}^{3} V_{jl}(\psi_2, \psi_3)\frac{d\psi_l}{dt} \tag{4.15}$$

where ψ_l with $l = 1, 2, 3$ denote the Euler angles α_i, β_i and γ_i, respectively.

The relation between the three angular velocities is given by

$$\boldsymbol{\omega}_i = R(\alpha_i, \beta_i, \gamma_i)\boldsymbol{\omega}' + \boldsymbol{\omega}_i'' \tag{4.16}$$

where $R(\alpha_i, \beta_i, \gamma_i)$ denotes the transformation matrix which connects the axes of the molecular frame and the principal axes of each nucleus. Explicitely it is given by

$$R(\alpha_i, \beta_i, \gamma_i) =$$

$$\begin{pmatrix} \cos\alpha_i\cos\beta_i\cos\gamma_i - \sin\alpha_i\sin\gamma_i & \sin\alpha_i\cos\beta_i\cos\gamma_i + \cos\alpha_i\sin\gamma_i & -\sin\beta_i\cos\gamma_i \\ -\cos\alpha_i\cos\beta_i\sin\gamma_i - \sin\alpha_i\cos\gamma_i & -\sin\alpha_i\cos\beta_i\sin\gamma_i + \cos\alpha_i\cos\gamma_i & \sin\beta_i\sin\gamma_i \\ \cos\alpha_i\cos\beta_i & \sin\alpha_i\sin\beta_i & \cos\beta_i \end{pmatrix} \tag{4.17}$$

Inserting (4.16) in (4.12), the kinetic energy is expressed as a sum of three parts:

$$T_{\text{cl}} = T_{\text{rot}} + T_{\text{int}} + T_{\text{C}} \tag{4.18}$$

where

$$T_{\text{rot}} = \frac{1}{2}{}^t\boldsymbol{\omega}'\boldsymbol{\mathcal{I}}_{\text{s}}\boldsymbol{\omega}' \tag{4.19}$$

is the total rotational energy of the interacting nuclei as a whole. The inertia tensor is given by

$$\boldsymbol{\mathcal{I}}_{\text{s}} = \boldsymbol{\mathcal{J}}_{\text{rel}} + {}^tR(\alpha_1,\beta_1,\gamma_1)\boldsymbol{\mathcal{J}}_1R(\alpha_1,\beta_1,\gamma_1) + {}^tR(\alpha_2,\beta_2,\gamma_2)\boldsymbol{\mathcal{J}}_2R(\alpha_2,\beta_2,\gamma_2) \tag{4.20}$$

Since we assume that the deformations of the constituent nuclei are axially symmetric, the inertia tensors $\mathcal{J}_{1,2}$ are diagonal in the coordinate frames of the principal axes of the constituent nuclei

$$\mathcal{J}_1 = \begin{pmatrix} J_A & 0 & 0 \\ 0 & J_A & 0 \\ 0 & 0 & J_a \end{pmatrix}, \quad \mathcal{J}_2 = \begin{pmatrix} J_B & 0 & 0 \\ 0 & J_B & 0 \\ 0 & 0 & J_b \end{pmatrix} \tag{4.21}$$

Substituting these last two expressions in (4.20), the components of the symmetric matrix $\boldsymbol{\mathcal{I}}_{\text{s}}$ are obtained

$$\begin{aligned}
I_{11} &= \mu R^2 + J_A + J_B + (J_a - J_A)\cos^2\alpha_1\sin^2\beta_1 + (J_b - J_B)\cos^2\alpha_2\sin^2\beta_2 \\
I_{12} &= (J_a - J_A)\sin\alpha_1\cos\alpha_1\sin^2\beta_1 + (J_b - J_B)\sin\alpha_2\cos\alpha_2\sin^2\beta_2 \\
I_{13} &= (J_a - J_A)\cos\alpha_1\sin\beta_1\cos\beta_1 + (J_b - J_B)\cos\alpha_2\sin\beta_2\cos\beta_2 \\
I_{22} &= \mu R^2 + J_A + J_B + (J_a - J_A)\sin^2\alpha_1\sin^2\beta_1 + (J_b - J_B)\sin^2\alpha_2\sin^2\beta_2 \\
I_{23} &= (J_a - J_A)\sin\alpha_1\sin\beta_1\cos\beta_1 + (J_b - J_B)\sin\alpha_2\sin\beta_2\cos\beta_2 \\
I_{33} &= J_A + J_B + (J_a - J_A)\cos^2\beta_1 + (J_b - J_B)\cos^2\beta_2
\end{aligned} \tag{4.22}$$

In case the nuclei have no non-axial deformations and since in quantum mechanics the rotations around the symmetry axis are forbidden $I_a = I_b = 0$ and since $\alpha_1 = -\alpha_2 = \alpha$, the above expressions are rewritting:

$$\begin{aligned}
I_{11} &= \mu R^2 + J_A + J_B - (J_A\sin^2\beta_1 + J_B\sin^2\beta_2)\cos^2\alpha \\
I_{12} &= (-J_A\sin^2\beta_1 + J_B\sin^2\beta_2)\sin\alpha\cos\alpha \\
I_{13} &= -(J_A\sin\beta_1\cos\beta_1 + J_B\sin\beta_2\cos\beta_2)\cos\alpha \\
I_{22} &= \mu R^2 + J_A + J_B - (J_A\sin^2\beta_1 + J_B\sin^2\beta_2)\sin^2\alpha \\
I_{23} &= (-J_A\sin\beta_1\cos\beta_1 + J_B\sin\beta_2\cos\beta_2)\sin\alpha \\
I_{33} &= J_A\sin^2\beta_1 + J_B\sin^2\beta_2
\end{aligned} \tag{4.23}$$

Now introducing (4.13) in (4.19) we obtain

$$T_{\text{rot}} = \frac{1}{2}\sum_{1\le i,j\le 3} g_{ij}^{\text{rot}}\dot\theta_i\dot\theta_j \tag{4.24}$$

where the elements of the symmetric tensor $\boldsymbol{g}^{\text{rot}}$ are given in terms of the matrix elements I_{ij}

$$\begin{aligned}
g_{11}^{\text{rot}} &= I_{11}\sin^2\theta_2\cos^2\theta_2 + I_{22}\sin^2\theta_2\sin^2\theta_3 + I_{33}\cos^2\theta_2 \\
&- 2I_{12}\sin^2\theta_2\sin\theta_3\cos\theta_3 - 2I_{13}\sin\theta_2\cos\theta_2\cos\theta_3 + 2I_{23}\sin\theta_2\cos\theta_2\sin\theta_3 \\
g_{12}^{\text{rot}} &= (I_{22}-I_{11})\sin\theta_2\sin\theta_3\cos\theta_3 + I_{12}\sin\theta_2(\sin^2\theta_3-\cos^2\theta_3) \\
&+ I_{13}\cos\theta_2\sin\theta_3 + I_{23}\cos\theta_2\cos\theta_3 \\
g_{13}^{\text{rot}} &= I_{33}\cos\theta_2 - I_{13}\sin\theta_2\cos\theta_3 + I_{23}\sin\theta_2\sin\theta_3 \\
g_{22}^{\text{rot}} &= I_{11}\sin^2\theta_3 + I_{22}\cos^2\theta_3 + 2I_{12}\sin\theta_3\cos\theta_3 \\
g_{23}^{\text{rot}} &= I_{13}\sin\theta_3 + I_{23}\cos\theta_3 \\
g_{33}^{\text{rot}} &= I_{33}
\end{aligned} \tag{4.25}$$

The second term in (4.18) is the intrinsic kinetic energy T_{int}

$$T_{\text{int}} = \frac{1}{2}\mu\dot{\boldsymbol{R}}^2 + \frac{1}{2}{}^t\boldsymbol{\omega}_1''\boldsymbol{\mathcal{J}}_1\boldsymbol{\omega}_1'' + \frac{1}{2}{}^t\boldsymbol{\omega}_2''\boldsymbol{\mathcal{J}}_2\boldsymbol{\omega}_2'' \tag{4.26}$$

By substituing the angular velocities $\boldsymbol{\omega}_i''$, whose components are given in (4.15) we obtain

$$\begin{aligned}
T_{\text{int}} &= \frac{1}{2}\mu\dot{R}^2 + \frac{1}{2}\left[J_A(\dot{\alpha}_1^2\sin^2\beta_1 + \dot{\beta}_1^2) + J_a(\dot{\alpha}_1\cos\beta_1 + \dot{\gamma}_1^2)^2\right] \\
&\quad + \frac{1}{2}\left[J_B(\dot{\alpha}_2^2\sin^2\beta_2 + \dot{\beta}_2^2) + J_b(\dot{\alpha}_2\cos\beta_2 + \dot{\gamma}_2^2)^2\right] \\
&= \frac{1}{2}\mu\dot{R}^2 + \frac{1}{2}(J_A\sin^2\beta_1 + J_B\sin^2\beta_2)\dot{\alpha}^2 + \frac{1}{2}(J_A\dot{\beta}_1^2 + J_B\dot{\beta}_2^2)
\end{aligned} \tag{4.27}$$

Thus

$$T_{\text{int}} = \frac{1}{2}\sum_{1\le i,j\le 4} g_{ij}^{\text{int}}\dot{q}_i\dot{q}_j \tag{4.28}$$

where the only non vanishing elements of the symmetric tensor $\boldsymbol{g}^{\text{int}}$ for the internal variables are given by

$$\begin{aligned}
g_{11}^{\text{int}} &= J_A\sin^2\beta_1 + J_B\sin^2\beta_2 \\
g_{22}^{\text{int}} &= \mu \\
g_{33}^{\text{int}} &= J_A \\
g_{44}^{\text{int}} &= J_B
\end{aligned} \tag{4.29}$$

The third term in (4.18) is the Coriolis coupling term T_{C}

$$T_{\text{C}} = {}^t\boldsymbol{\omega}'\left({}^tR(\alpha_1,\beta_1,\gamma_1)\boldsymbol{\mathcal{I}}_1\boldsymbol{\omega}_1'' + {}^tR(\alpha_2,\beta_2,\gamma_2)\boldsymbol{\mathcal{I}}_2\boldsymbol{\omega}_2''\right) \tag{4.30}$$

We obtain upon substitution

$$T_{\text{C}} = \frac{1}{2}\sum_{\substack{1\le i\le 3\\ 1\le j\le 4}} g_{ij}^{\text{C}}\dot{\theta}_i\dot{q}_j \tag{4.31}$$

where q_j are the subset of four variables $(\alpha, R, \beta_1\beta_2)$. The non-zero elements of the metric tensor g^{C}_{ij} are given by

$$\begin{aligned}
g^{\mathrm{C}}_{11} &= (J_A \sin\beta_1 \cos\beta_1 - J_B \sin\beta_2 \cos\beta_2) \cos\alpha \sin\theta_2 \cos\theta_3 \\
&- (J_A \sin\beta_1 \cos\beta_1 + J_B \sin\beta_2 \cos\beta_2) \sin\alpha \sin\theta_2 \sin\theta_3 \\
&+ (J_A \sin^2\beta_1 - J_B \sin^2\beta_2) \cos\theta_2 \\
g^{\mathrm{C}}_{13} &= J_A(\sin\alpha \sin\theta_2 \cos\theta_3 + \cos\alpha \sin\theta_2 \sin\theta_3) \\
g^{\mathrm{C}}_{14} &= J_B(-\sin\alpha \sin\theta_2 \cos\theta_3 + \cos\alpha \sin\theta_2 \sin\theta_3) \\
g^{\mathrm{C}}_{21} &= (-J_A \sin\beta_1 \cos\beta_1 + J_B \sin\beta_2 \cos\beta_2) \cos\alpha \sin\theta_3 \\
&- (J_A \sin\beta_1 \cos\beta_1 + J_B \sin\beta_2 \cos\beta_2) \sin\alpha \cos\theta_3 \\
g^{\mathrm{C}}_{23} &= J_A(-\sin\alpha \sin\theta_3 + \cos\alpha \cos\theta_3) \\
g^{\mathrm{C}}_{24} &= J_B(\sin\alpha \sin\theta_3 + \cos\alpha \cos\theta_3) \\
g^{\mathrm{C}}_{31} &= J_A \sin^2\beta_1 - J_B \sin^2\beta_2
\end{aligned} \tag{4.32}$$

Next the classical kinetic energy must be quantized. In curvilinear coordinate the quantized form reads [1]

$$\widehat{T} = -\frac{\hbar^2}{2} \sum_{ij} \frac{1}{\sqrt{g}} \frac{\partial}{\partial q_i} \sqrt{g} (g^{-1})_{ij} \frac{\partial}{\partial q_j} \tag{4.33}$$

The total 7×7 metric g_{ij} tensor can be arranged in term of the sub-matrices g^{rot}, g^{int} and g^{C}

$$(g)_{ij} = \begin{pmatrix} g^{\mathrm{rot}} & g^{\mathrm{C}} \\ g^{\mathrm{C}} & g^{\mathrm{int}} \end{pmatrix} \tag{4.34}$$

whose determinat is given by

$$g = 4\mu^3 R^4 J_A^2 \sin^2\beta_1 J_B^2 \sin^2\beta_2 \tag{4.35}$$

The volume element is given by

$$d\tau = \sqrt{g} dq_1 \ldots dq_n = \sqrt{g} d\tau' \tag{4.36}$$

Some times is more convenient to use as volume element $d\tau'$ instead. The total volume element can be factored in volume elements for rotational and vibrational variables

$$d\tau = d\Omega \cdot dV \tag{4.37}$$

where

$$d\Omega = 2 \sin\theta_2 d\theta_1 d\theta_2 d\theta_3 d\alpha \tag{4.38}$$

$$dV = D dR d\beta_1 d\beta_2 \tag{4.39}$$

with the weight function D as

$$D = \mu^{3/2} R^2 J_A \sin\beta_1 J_B \sin\beta_2 \tag{4.40}$$

We have to redefine the kinetic energy $\hat{T}$ and the wave function ψ of the original Schrödinger equation via

$$\begin{aligned} \widetilde{T} &\rightarrow \widehat{T}' = g^{\frac{1}{2}}\widehat{H}g^{-\frac{1}{2}} \\ \psi &\rightarrow \phi = g^{\frac{1}{2}}\psi \end{aligned} \tag{4.41}$$

so that

$$\int \psi^* \widehat{T}\psi d\tau = \int \phi^* \widehat{T}'\phi d\tau'$$

holds. The potential commutes with g because it does not contain differential operators. The kinetic energy simplifies to

$$\widehat{T} = \widehat{\widetilde{T}} + \widehat{V}_{\text{add}} \tag{4.42}$$

where

$$\widehat{\widetilde{T}} = -\frac{\hbar^2}{2}\sum_{\mu\nu}\frac{\partial}{\partial q_\mu}(g^{-1})_{\mu\nu}\frac{\partial}{\partial q_\nu} \tag{4.43}$$

$$\widehat{V}_{\text{add}} = -\frac{\hbar^2}{8}\sum_{\mu\nu}\left\{\frac{3}{4}(g^{-1})_{\mu\nu}\frac{1}{g^2}\frac{\partial g}{\partial q_\nu}\frac{\partial g}{\partial q_\nu} - \frac{1}{g}\frac{\partial (g^{-1})_{\mu\nu}}{\partial q_\mu}\frac{\partial g}{\partial q_\nu} - (g^{-1})_{\mu\nu}\frac{1}{g}\frac{\partial^2 g}{\partial q_\nu \partial q_\mu}\right\} \tag{4.44}$$

The quantum mechanical kinetic energy is divided into three terms, i.e. $\widehat{T}_{\text{rot}}$, $\widehat{T}_{\text{vib}}$ and $\widehat{T}_{\text{C}}$. $\widehat{T}_{\text{rot}}$ is given by that part of the sum (4.33) with $i, j = 1\ 3$, i.e. it is given in terms of the differential operators of the angles θ_i. The quantum rotational energy in the three variables θ_i was worked out in [1] and the result is

$$\widehat{T}_{\text{rot}} = \frac{\hbar^2}{2}\sum_{1\leq i,j\leq 3}\mu_{ij}\widehat{L}'_i\widehat{L}'_j \tag{4.45}$$

where the three components of the angular momentum operators are referred to the molecular frame and are given by

$$\widehat{L}'_1 = -i\left(-\frac{\cos\theta_3}{\sin\theta_2}\frac{\partial}{\partial\theta_1} + \sin\theta_3\frac{\partial}{\partial\theta_2} + \cot\theta_2\cos\theta_3\frac{\partial}{\partial\theta_3}\right) \tag{4.46}$$

$$\widehat{L}'_2 = -i\left(\frac{\sin\theta_3}{\sin\theta_2}\frac{\partial}{\partial\theta_1} + \cos\theta_3\frac{\partial}{\partial\theta_2} - \cot\theta_2\sin\theta_3\frac{\partial}{\partial\theta_3}\right) \tag{4.47}$$

$$\widehat{L}'_3 = -i\frac{\partial}{\partial\theta_3} \tag{4.48}$$

where the μ_{ij} coefficients are given as

$$\mu_{11} = \mu_{22} = \frac{1}{\mu R^2},\ \ \mu_{12} = 0 \tag{4.49}$$

$$\mu_{13} = \frac{1}{2\mu R^2}\cos\alpha(\cot\beta_1 + \cot\beta_2) \tag{4.50}$$

$$\mu_{23} = \frac{1}{2\mu R^2}\cos\alpha(\cot\beta_1 - \cot\beta_2) \tag{4.51}$$

$$\begin{aligned}\mu_{33} &= \frac{1}{4}\left[\left(\frac{1}{J_A}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_1}+\left(\frac{1}{J_B}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_2}\right]\\ &+ \frac{1}{2\mu R^2}(\cos 2\alpha\cot\beta_1\cot\beta_2 - 1)\end{aligned} \tag{4.52}$$

Next, we add to $\widehat{T}_{\text{rot}}$ the kinetic energy associated with α, i.e. add the terms with $i = 4$ and/or $j = 4$. The elements of g^{-1} with $i = 1-4$, $j = 4$ are given by

$$\begin{aligned}(g^{-1})_{1\le i\le 3, j=4} &= \begin{pmatrix}(-\mu_{14}\cos\theta_3 + \mu_{24}\sin\theta_3/\sin\theta_2)\\ \mu_{14}\sin\theta_3 + \mu_{24}\cos\theta_3\\ (\mu_{14}\cos\theta_3 - \mu_{24}\cos\theta_2\sin\theta_3)/\sin\theta_2 + \mu_{34}\end{pmatrix}\\ (g^{-1})_{44} &= \mu_{44}\end{aligned} \tag{4.53}$$

where the coefficients are given by

$$\mu_{14} = \frac{1}{2\mu R^2}\cos\alpha(\cot\beta_1 - \cot\beta_2) \tag{4.54}$$

$$\mu_{24} = \frac{1}{2\mu R^2}\sin\alpha(\cot\beta_1 + \cot\beta_2) \tag{4.55}$$

$$\mu_{34} = \frac{1}{4}\left[\left(\frac{1}{J_A}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_1}-\left(\frac{1}{J_B}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_2}\right] \tag{4.56}$$

$$\begin{aligned}\mu_{44} &= \frac{1}{4}\left[\left(\frac{1}{J_A}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_1}+\left(\frac{1}{J_B}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_2}\right]\\ &- \frac{1}{2\mu R^2}(\cos 2\alpha\cot\beta_1\cot\beta_2 + 1)\end{aligned} \tag{4.57}$$

Substituing the matrix elements $(g^{-1})_{1-4,j=4}$ into (4.33) we obtain the kinetic energy associated with α

$$\widehat{T}_{\text{rot}}(\alpha) = -\frac{\hbar^2}{2}\left\{\sum_{1\le k\le 3}\left[\widehat{L}'_k\mu_{k4}\left(-i\frac{\partial}{\partial\alpha}\right)+\left(-i\frac{\partial}{\partial\alpha}\right)\mu_{k4}\widehat{L}'_k\right]+\left(-i\frac{\partial}{\partial\alpha}\right)\mu_{44}\left(-i\frac{\partial}{\partial\alpha}\right)\right\} \tag{4.58}$$

and thus the new $\widehat{T}_{\text{rot}}$, is expressed in the compact form

$$\widehat{T}^4_{\text{rot}} = \frac{\hbar^2}{2}\sum_{1\le i,j\le 4}\widehat{L}'_i\mu_{ij}\widehat{L}'_j \tag{4.59}$$

where $\widehat{L}'_4$ is given by

$$\widehat{L}'_4 \equiv \widehat{L}'_\alpha = -i\frac{\partial}{\partial\alpha} \tag{4.60}$$

$\widehat{T}_{\text{vib}}$ is given by that part of the sum (4.33) with $i, j = 5-7$, i.e. it is given in terms of the differential operators of the internal variables (R, β_1, β_2). The strategy of calculating $\widehat{T}_{\text{vib}}$ consists in removing the weight function D from the vibrational volume element dV by multiplying the wave function by $\sqrt{D}$. Accordingly the vibrational kinetic energy is transformed into

$$\widehat{T}_{\text{vib}} = -\frac{\hbar^2}{2}\sum_{5\le i,j\le 7}\frac{1}{\sqrt{D}}\frac{\partial}{\partial q_i}D(g^{-1})_{ij}\frac{\partial}{\partial q_j}\frac{1}{\sqrt{D}} = \widehat{\widetilde{T}}_{\text{vib}} + \widehat{V}_{\text{add}} \tag{4.61}$$

where $\widehat{\widetilde{T}}_{\text{vib}}$ and $\widehat{V}_{\text{add}}$ denote the modified vibrational kinetic energy and "additional potential" originated from D, and are given by

$$\widehat{\widetilde{T}}_{\text{vib}} = -\frac{\hbar^2}{2}\sum_{5\le i,j\le 7}\frac{\partial}{\partial q_i}(g^{-1})_{ij}\frac{\partial}{\partial q_j}, \tag{4.62}$$

$$\widehat{V}_{\text{add}} = \frac{\hbar^2}{4D}\sum_{5\le i,j\le 7}\left[\frac{\partial(g^{-1})_{ij}}{\partial q_i}\frac{\partial D}{\partial q_j} + (g^{-1})_{ij}\frac{\partial^2 D}{\partial q_i\partial q_j} - (g^{-1})_{ij}\frac{1}{2D}\frac{\partial D}{\partial q_i}\frac{\partial D}{\partial q_j}\right] \tag{4.63}$$

and the substition of the matrix elements $(g)_{5\le i,j\le 7}$ results in

$$\begin{aligned}\widehat{\widetilde{T}}_{\text{vib}} = & -\frac{\hbar^2}{2}\left[\frac{1}{\mu}\frac{\partial^2}{\partial R^2} + \left(\frac{1}{J_A}+\frac{1}{\mu R^2}\right)\frac{\partial^2}{\partial\beta_1^2} + \left(\frac{1}{J_B}+\frac{1}{\mu R^2}\right)\frac{\partial^2}{\partial\beta_2^2}\right.\\ & \left. + \frac{2\cos 2\alpha}{\mu R^2}\frac{\partial^2}{\partial\beta_1\partial\beta_2}\right]\end{aligned} \tag{4.64}$$

$$\begin{aligned}\widehat{V}_{\text{add}} = & -\frac{\hbar^2}{8}\left[\left(\frac{1}{J_A}+\frac{1}{\mu R^2}\right)\left(\frac{1}{\sin^2\beta_1}+1\right) + \left(\frac{1}{J_B}+\frac{1}{\mu R^2}\right)\left(\frac{1}{\sin^2\beta_2}+1\right)\right.\\ & \left. - \frac{2\cos 2\alpha}{\mu R^2}\cot\beta_1\cot\beta_2\right]\end{aligned} \tag{4.65}$$

The coupling term $\widehat{T}_{\text{C}}$, given by the sum (4.31) is also transformed into a new one $\widehat{\widetilde{T}}_{\text{C}}$ due to the change of the volume element as follows

$$\widehat{T}_{\text{C}} = -\frac{\hbar^2}{2}\left[\sum_{\substack{1\le i\le 4\\ 5\le j\le 7}}\frac{1}{\sqrt{D}\sin\theta_2}\left(\frac{\partial}{\partial q_i}D\sin\theta_2(g^{-1})_{ij}\frac{\partial}{\partial q_j} + \frac{\partial}{\partial q_j}D\sin\theta_2(g^{-1})_{ji}\frac{\partial}{\partial q_i}\right)\frac{1}{\sqrt{D}}\right] \tag{4.66}$$

where the elements g^{-1} are obtained as

$$
\begin{aligned}
& (g^{-1})_{i=5,j=1-4} = 0 \\
(g^{-1})_{61} &= (-\sin\alpha\cos\theta_3 - \cos\alpha\sin\theta_3)/\mu R^2 \sin\theta_2 \\
(g^{-1})_{62} &= (\sin\alpha\sin\theta_3 - \cos\alpha\cos\theta_3)/\mu R^2 \\
(g^{-1})_{63} &= \left[(\sin\alpha\cos\theta_3 + \cos\alpha\sin\theta_3)\cot\theta_2 + \frac{1}{2}\sin 2\alpha\cot\beta_2\right]/\mu R^2 \\
(g^{-1})_{64} &= -\frac{1}{2}\sin 2\alpha\cot\beta_2/\mu R^2 \\
(g^{-1})_{71} &= (\sin\alpha\cos\theta_3 - \cos\alpha\sin\theta_3)/\mu R^2 \sin\theta_2 \\
(g^{-1})_{72} &= (-\sin\alpha\sin\theta_3 - \cos\alpha\cos\theta_3)/\mu R^2 \\
(g^{-1})_{73} &= \left[(-\sin\alpha\cos\theta_3 + \cos\alpha\sin\theta_3)\cot\theta_2 - \frac{1}{2}\sin 2\alpha\cot\beta_1\right]/\mu R^2 \\
(g^{-1})_{74} &= -\frac{1}{2}\sin 2\alpha\cot\beta_1/\mu R^2
\end{aligned}
\tag{4.67}
$$

Substituting these matrix elements in (4.66) results in

$$
\begin{aligned}
\widehat{T}_{\mathrm{C}} &= -\frac{\hbar^2}{2}\left[\sum_{\substack{1\le i\le 4\\ 5\le j\le 7}} (g^{-1})_{ij}\frac{\partial}{\partial\theta_i}\left(\frac{\partial}{\partial q_j} - \frac{1}{2D}\frac{\partial D}{\partial q_j}\right) + \sum_{j=6,7}\frac{\partial}{\partial\alpha}(g^{-1})_{4,j}\left(\frac{\partial}{\partial q_j} - \frac{1}{2D}\frac{\partial D}{\partial q_j}\right)\right. \\
&+ \left.\sum_{\substack{i=6,7\\ 1\le j\le 3}}\left(\frac{\partial}{\partial q_i} + \frac{1}{2D}\frac{\partial D}{\partial q_i}\right)(g^{-1})_{ij}\frac{\partial}{\partial\theta_j} + \sum_{j=6,7}\left(\frac{\partial}{\partial q_i} + \frac{1}{2D}\frac{\partial D}{\partial q_i}\right)(g^{-1})_{i,4}\frac{\partial}{\partial\alpha}\right]
\end{aligned}
$$

$$
\begin{aligned}
&= \frac{\hbar^2}{\mu R^2}\left[i\sin\alpha\left(-\frac{\partial}{\partial\beta_1} + \frac{\partial}{\partial\beta_2}\right)\widehat{L}'_1 + i\cos\alpha\left(\frac{\partial}{\partial\beta_1} + \frac{\partial}{\partial\beta_2}\right)\widehat{L}'_2\right. \\
&+ \frac{i}{2}\sin 2\alpha\left(-\cot\beta_2\frac{\partial}{\partial\beta_1} + \cot\beta_1\frac{\partial}{\partial\beta_2}\right)\widehat{L}'_3 \\
&+ \frac{i}{4}\sin 2\alpha\left(\cot\beta_2\frac{\partial}{\partial\beta_1} + \cot\beta_1\frac{\partial}{\partial\beta_2}\right)\widehat{L}'_4 \\
&+ \left.\frac{i}{4}\widehat{L}'_4\sin 2\alpha\left(\cot\beta_2\frac{\partial}{\partial\beta_1} + \cot\beta_1\frac{\partial}{\partial\beta_2}\right) - \frac{1}{2}\cos 2\alpha\cot\beta_1\cot\beta_2\right]
\end{aligned}
\tag{4.68}
$$

Adding to the above expression of the Coriolis kinetic energy the K-mixing terms from $\widehat{T}^4_{\mathrm{rot}}$ we obtain a new coupling $\widehat{T}'_{\mathrm{C}}$ which is divided into two terms, one which preserves the K-quantum number and a second one which does not:

$$
\widehat{T}'_{\mathrm{C}} = \widehat{T}_{\mathrm{C}}(K) + \widehat{T}_{\mathrm{C}}(K, K') \tag{4.69}
$$

where

$$\begin{aligned}\widehat{T}_{\rm C}(K) &= \frac{\hbar^2}{4\mu R^2}\left[-(e^{2i\alpha}+e^{-2i\alpha})\cot\beta_1\cot\beta_2\right.\\ &+ \cot\beta_2\frac{\partial}{\partial\beta_1}\left(-e^{2i\alpha}(\widehat{L}'_3-\widehat{L}'_4-1)+(\widehat{L}'_3-\widehat{L}'_4-1)e^{-2i\alpha}\right)\\ &+ \left.\cot\beta_1\frac{\partial}{\partial\beta_2}\left(e^{2i\alpha}(\widehat{L}'_3+\widehat{L}'_4+1)+(\widehat{L}'_3+\widehat{L}'_4+1)e^{-2i\alpha}\right)\right]\end{aligned} \tag{4.70}$$

$$\begin{aligned}\widehat{T}_{\rm C}(K,K') &= \frac{\hbar^2}{4\mu R^2}\left[e^{i\alpha}\widehat{L}'_-\left((\widehat{L}'_3+\widehat{L}'_4+1)\cot\beta_1-2\frac{\partial}{\partial\beta_1}\right)\right.\\ &+ \left((\widehat{L}'_3+\widehat{L}'_4+1)\cot\beta_1+2\frac{\partial}{\partial\beta_1})\right)\widehat{L}'_+e^{-i\alpha}\\ &+ e^{i\alpha}\widehat{L}'_+\left((\widehat{L}'_3-\widehat{L}'_4-1)\cot\beta_2+2\frac{\partial}{\partial\beta_2}\right)\\ &+ \left.\left((\widehat{L}'_3-\widehat{L}'_4-1)\cot\beta_2-2\frac{\partial}{\partial\beta_2}\right)\widehat{L}'_-e^{-i\alpha}\right]\end{aligned} \tag{4.71}$$

with $\widehat{L}'_\pm$ denoting $\widehat{L}'_1 \pm i\widehat{L}'_2$

4.1.3 The K-diagonal approximation.

The kinetical couplings obtained in 4.1.2 are quite complicate and even a coupled channel numerical procedure is difficult to be implemented. The problem can be easier handled if we start with that part of the Hamiltonian which conserves the K-quantum number, a case which is appropriate for elongated systems, occuring in fission and fusion. In fact the K-mixing terms in $\widehat{T}^4_{\rm rot}$ with the coefficients μ_{13}, μ_{14}, μ_{23} and μ_{24} are relatively small, because they contain the factor $1/\mu R^2$ which is much smaller than $J_{A(B)}$ in the contact(scission) region. It is then usefull to regroup the kinetic energy operator as follows,

$$\widehat{T} = \widehat{T}' + \widehat{T}'_{\rm C} \tag{4.72}$$

$$\widehat{T}' = \widehat{T}^4_{\rm rot} + \widehat{T}_{\rm vib} \tag{4.73}$$

where $\widehat{T}'_{\rm C}$ was defined in (4.69). In what follows we neglect the effects due to $\widehat{T}'_{\rm C}$.

The K-diagonal part of the rotational energy (4.59), can be splitted in a non-perturbed part and a coupling term

$$\widehat{T}^4_{\rm rot} = \widehat{T}^0_{\rm rot} + \delta\widehat{T}_{\rm rot} \tag{4.74}$$

where

$$\begin{aligned}\widehat{T}^0_{\rm rot} &= \frac{\hbar^2}{2\mu R^2}(\widehat{L}^2-\widehat{L}'^2_3)+\frac{\hbar^2}{2\mu_{33}}\widehat{L}'^2_3+\frac{\hbar^2}{2\mu_{44}}\widehat{L}'^2_4+\frac{\hbar^2}{2\mu_{34}}(\widehat{L}'_3\widehat{L}'_4+\widehat{L}'_4\widehat{L}'_3)\\ &= \frac{\hbar^2}{2\mu R^2}(\widehat{L}^2-\frac{3}{2}\widehat{L}'^2_3-\frac{1}{2}\widehat{L}'^2_4)\\ &+ \frac{\hbar^2}{8}\left[\left(\frac{1}{J_A}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_1}(\widehat{L}'_3+\widehat{L}'_4)^2+\left(\frac{1}{J_B}+\frac{1}{\mu R^2}\right)\frac{1}{\sin^2\beta_2}(\widehat{L}'_3-\widehat{L}'_4)^2\right]\end{aligned} \tag{4.75}$$

and

$$\delta\widehat{T}_{\text{rot}} = \frac{\hbar^2}{4\mu R^2}(\widehat{L}'_3 - \widehat{L}'_4)\cos 2\alpha \cot\beta_1 \cot\beta_2 (\widehat{L}'_3 - \widehat{L}'_4) \tag{4.76}$$

The vibrational operator is splitted as follows

$$\begin{aligned}\widehat{T}^0_{\text{vib}} = & - \frac{\hbar^2}{2}\left[\frac{1}{\mu}\frac{\partial^2}{\partial R^2} + \left(\frac{1}{J_A} + \frac{1}{\mu R^2}\right)\frac{\partial^2}{\partial\beta_1^2} + \left(\frac{1}{J_B} + \frac{1}{\mu R^2}\right)\frac{\partial^2}{\partial\beta_2^2}\right] \\ & - \frac{\hbar^2}{8}\left[\left(\frac{1}{J_A} + \frac{1}{\mu R^2}\right)\left(\frac{1}{\sin^2\beta_1} + 1\right) + \left(\frac{1}{J_B} + \frac{1}{\mu R^2}\right)\left(\frac{1}{\sin^2\beta_2} + 1\right)\right]\end{aligned} \tag{4.77}$$

$$\delta\widehat{T}_{\text{vib}} = -\frac{\hbar^2}{2R^2}(e^{2i\alpha} + e^{-2i\alpha})\left(\frac{\partial^2}{\partial\beta_1\partial\beta_2} - \frac{1}{4}\cot\beta_1\cot\beta_2\right) \tag{4.78}$$

The operator $\widehat{T}'$ is preserving the K-quantum number and therefore the eigenstates are of the rotation-vibration type [1]

$$\phi = D^{I^*}_{MK}(\theta_i)e^{i\nu\alpha}\chi_K(R, \beta_1, \beta_2) \tag{4.79}$$

Once the rotational degrees of freedom are decoupled, we remain with the the internal variables (R, β_1, β_2) which couple with each other through the kinetic energy operator $\widehat{T}'$ and the interaction potential. In this basis the zeroth-order rotational-vibrational kinetic energy reads

$$\begin{aligned}\widehat{T}^0(I, K, \nu) = & -\frac{\hbar^2}{2\mu}\left[\frac{\partial^2}{R^2} - \frac{1}{R^2}\left(I(I+1) - \frac{3}{2}K^2 - \frac{1}{2}\nu^2\right)\right] - \frac{\hbar^2}{8}\left(\frac{1}{J_A} + \frac{1}{J_B} + \frac{2}{\mu R^2}\right) \\ & -\frac{\hbar^2}{2}\left(\frac{1}{J_A} + \frac{1}{\mu R^2}\right)\left(\frac{\partial^2}{\partial\beta_1^2} - \frac{1}{4}\frac{(K+\nu)^2 - 1}{\sin^2\beta_1}\right) \\ & -\frac{\hbar^2}{2}\left(\frac{1}{J_B} + \frac{1}{\mu R^2}\right)\left(\frac{\partial^2}{\partial\beta_2^2} - \frac{1}{4}\frac{(K-\nu)^2 - 1}{\sin^2\beta_2}\right)\end{aligned} \tag{4.80}$$

whereas the perturbed part is given by

$$\begin{aligned}\delta\widehat{T}(K) = & -\frac{\hbar^2}{2\mu R^2}\left\{(e^{2i\alpha} + e^{-2i\alpha})\left(\frac{\partial^2}{\partial\beta_1\partial\beta_2} - \frac{1}{4}(K^2+1)\cot\beta_1\cot\beta_2\right)\right. \\ & + \left.\frac{1}{4}\widehat{L}'_4(e^{2i\alpha} + e^{-2i\alpha})\widehat{L}'_4\cot\beta_1\cot\beta_2\right\}\end{aligned} \tag{4.81}$$

4.1.4 The harmonic approximation

The complicate dynamics of the dinuclear system internal motions can be solved by looking for the normal modes around the equilibrium. The precessional centrifugal terms entering in $\widehat{T}^0(I, K, \nu)$(second and third rows of eq.(4.80), which are produced by $K-$ and $\nu-$ rotational motion around the molecular axis z' will be moved together with the β_i kinetical terms in separated Hamiltonians $\widehat{H}_{\beta_i}$

As will be proved in 6.1.4, the potential $V(R,\alpha,\beta_1,\beta_2)$, written earlier in (3.68), can be expanded in terms of small deviations of the angles β_i, from the equilibrium position $(\beta_1,\beta_2)=(0,0)$:

$$V(R,\alpha,\beta_1,\beta_2)=V(R)+\frac{1}{2}C_1(R)\beta_1^2+\frac{1}{2}C_2(R)\beta_2^2+C_{12}(R)\beta_1\beta_2\cos 2\alpha \tag{4.82}$$

Thus the solution for each β_i-mode is obtained by considering the zeroth-order sub-hamiltonians

$$\widehat{H}^0_{\beta_{1(2)}}=-\frac{\hbar^2}{2\tilde{J}_{A(B)}}\left(\frac{\partial^2}{\partial\beta_{1(2)^2}}-\frac{(K\pm\nu)^2-1}{4\beta^2_{1(2)}}\right)+\frac{1}{2}C_{1(2)}\beta^2_{1(2)} \tag{4.83}$$

where

$$\frac{1}{\tilde{J}_{A(B)}}=\frac{1}{J_{A(B)}}+\frac{1}{\mu R^2}$$

and $\sin\beta_i^2$ was approximated by β_i^2. The solution of the corresponding eigenvalue problem reads [1]

$$\widehat{H}^0_{\beta_i}\varphi_{K\nu n_{\beta_i}}(\beta_i)=E_{K\nu n_{\beta_i}}\varphi_{K\nu n_{\beta_i}}(\beta_i) \tag{4.84}$$

reads

$$\varphi_{K,\nu\,\beta_i}(\varepsilon)=\frac{\left\{\lambda_i^{l_{K\nu}+\frac{3}{2}}\Gamma(l_{K\nu}+\frac{3}{2}+n_{\beta_i})\right\}^{\frac{1}{2}}}{(n_{\beta_i}!)^{\frac{1}{2}}\Gamma(l_{K\nu}+\frac{3}{2})}\beta_i^{l_{K\nu}+1}e^{-\frac{1}{2}\lambda_i\beta_i^2}\;{}_1F_1(-n_{\beta_i},l_{K\nu}+\frac{3}{2};\lambda_i\beta_i^2) \tag{4.85}$$

where $\lambda_i^2=\frac{\tilde{J}_iC_i}{\hbar^2}$, $l_{K,\nu}=\frac{1}{2}\mid K\pm\nu\mid-\frac{1}{2}$ and ${}_1F_1(...)$ is the confluent hypergeometric function. The eigenvalues is given by

$$E_{K\nu n_{\beta_i}}=\left(l_{K\nu}+\frac{3}{2}+2n_{\beta_i}\right)\hbar\omega_{\beta_i},\quad \hbar\omega_{\beta_i}=\sqrt{\frac{C_i}{\tilde{J}_i}} \tag{4.86}$$

The perturbed part of the β_i-mode is derived from (4.81)

$$\begin{aligned}\delta\widehat{H}_{\beta_1\beta_2}=&-\frac{\hbar^2}{2\mu R^2}\left\{\delta_{\nu'\nu+2}\left(\frac{\partial^2}{\partial\beta_1\partial\beta_2}+\frac{1}{4}\frac{1}{\beta_1\beta_2}\right)+\delta_{\nu'\nu-2}\left(\frac{\partial^2}{\partial\beta_1\partial\beta_2}-\frac{3}{4}\frac{1}{\beta_1\beta_2}\right)\right\}\\&+\frac{1}{2}C_{12}(R)(\delta_{\nu'\nu+2}+\delta_{\nu'\nu2-2})\beta_1\beta_2\end{aligned} \tag{4.87}$$

4.1.5 Bound motion in the R-degree of freedom

In this the radial part, $V(R,0,0,0)$, is expanded around the equilibrium position R_{min}, localized on the bottom of the quasi-molecular pocket

$$V(R)=V(R_{\text{min}})+\frac{1}{2}\left.\frac{\partial^2V(R)}{\partial R}\right|_{R=R_{\text{min}}}(R-R_{\text{min}})^2+\mathcal{O}((R-R_{\text{min}})^4) \tag{4.88}$$

Naturally that the R-dependent stiffness coefficients C_1, C_2 and C_{12} must be also expanded around R_{min}.

The centrifugal term in the radial part of the kinetic energy (4.80), is evaluated at R_{min}, and thus the zeroth-order R-mode sub-hamiltonian corresponds to a one dimensional harmonic oscillator

$$\widehat{H}_R^0 = -\frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial R^2} + \frac{1}{2}k_R(R - R_{\text{min}})^2 \tag{4.89}$$

where $k_R = \partial^2 V(R)/\partial R|_{R=R_{\text{min}}}$, the corresponding eigenvalues and eigenvectors being

$$E_{n_R} = \hbar\omega_R\left(n_R + \frac{1}{2}\right), \quad \hbar\omega_{\beta_i} = \hbar\sqrt{\frac{k_R}{\mu}} \tag{4.90}$$

$$f_{n_R}(R) = \sqrt{\frac{1}{2^{n_R} n_R! \sqrt{\pi}}}\frac{\mu\omega}{\hbar}e^{-\frac{\mu\omega}{2\hbar}R^2} H_{n_R}\left(\sqrt{\frac{\mu\omega}{\hbar}}R\right) \tag{4.91}$$

The total unperturbed energy is

$$\begin{aligned} E^I_{K\nu n_R n_{\beta_1} n_{\beta_2}} &= V(R_{\text{min}}) + \hbar\omega_R\left(n_R + \frac{1}{2}\right) \\ &+ \frac{\hbar^2}{2}\left[\frac{1}{\mu R^2}\left(I(I+1) - \frac{1}{2}(3K^2 + \nu^2 + 1)\right) - \frac{1}{4}\left(\frac{1}{J_A} + \frac{1}{J_B}\right)\right] \\ &+ \left(\frac{|K+\nu|}{2} + 1 + 2n_{\beta_1}\right)\hbar\omega_{\beta_1} + \left(\frac{|K-\nu|}{2} + \frac{3}{2} + 2n_{\beta_2}\right)\hbar\omega_{\beta_2} \end{aligned} \tag{4.92}$$

4.1.6 Quasi-bound motion in the R-degree of freedom

In this case only the β_i degrees of freedom are treated as bound and ther frequencies are supposed to depend parametrically on R. The zeroth-order Hamiltonian of the quasi-bound radial mode reads

$$\begin{aligned} \widehat{H}_R^0 &= -\frac{\hbar^2}{2\mu}\left[\frac{\partial^2}{\partial R^2} - \frac{1}{R^2}\left(I(I+1) - \frac{3}{2}K^2 - \frac{1}{2}\nu^2 - \frac{1}{2}\right)\right] \\ &+ \left(\frac{|K+\nu|}{2} + 1 + 2n_{\beta_1}\right)\hbar\omega_{\beta_1}(R) + \left(\frac{|K-\nu|}{2} + \frac{3}{2} + 2n_{\beta_2}\right)\hbar\omega_{\beta_2}(R) \\ &+ V(R) - \frac{\hbar^2}{8}\left(\frac{1}{J_A} + \frac{1}{J_B}\right) \end{aligned} \tag{4.93}$$

Asymptotically he have

$$\widehat{H}_R^0 \longrightarrow -\frac{\hbar^2}{2\mu}\left[\frac{\partial^2}{\partial R^2} - \frac{1}{R^2}\left(I(I+1) - \frac{3}{2}K^2 - \frac{1}{2}\nu^2 - \frac{1}{2}\right) - \frac{2\eta K}{r} + \frac{1}{2}\mu\left(\frac{1}{J_A} + \frac{1}{J_B}\right)\right] \tag{4.94}$$

The corresponding regular Coulomb-like solution is of the form (see App.A.2):

$$F_\ell(\eta, kR) \sim e^{ikR}(kR)^{\ell+1}\ {}_1F_1(\ell + 1 - i\eta, 2\ell + 2, 2ikR) \tag{4.95}$$

where

$$k^2 = \frac{2\mu}{\hbar^2}\left[E + \frac{\hbar^2}{8}\left(\frac{1}{J_A} + \frac{1}{J_B}\right)\right] \tag{4.96}$$

and ℓ, generally non-integer number, reads

$$\ell = \frac{1}{2}\left(\sqrt{4I(I+1) - 6K^2 - 2\nu^2} - 1\right)$$

4.1.7 Symmetries of the Dinuclear System wave function

Boson Symmetry

When the clusters constituting the dinuclear system are identical the wave function should be invariant under their mutual exchange. Exchanging the c.m. positions of two constiuent nuclei, means to reverse the direction of relative vector $\boldsymbol{R}$ and thus, if the clusters are along the z'-axis, to reverse the direction of this axis. Since the intrinsic configurations of two arbitrary oriented objects remain unchanged after the exchane of two nuclei, the orientation of the x'-axis of the molecular frame is kept unchanged. The axes of the transformed molecular frame, i.e. $(x', -y', -z')$ are obtained by rotations with $(\pi+\theta_1, \pi-\theta_2, 0)$. The internal variables will be tranformed according to $(\alpha_1, \beta_1, \alpha_2, \beta_2) \rightarrow (2\pi-\alpha_2, \pi-\beta_2, 2\pi-\alpha_1, \pi-\beta_1)$. Since we introduced earlier the definitions $\alpha = (\alpha_1-\alpha_2)/2$, $\theta_3 = (\alpha_1+\alpha_2)/2$, the exchange operator $\mathcal{P}_{12}$ acts as:

$$\mathcal{P}_{12} : (\theta_1, \theta_2, \theta_3, \alpha, R, \beta_1, \beta_2) \rightarrow (\pi+\theta_1, \pi-\theta_2, -\theta_3, \alpha, R, \beta_1, \beta_2) \tag{4.97}$$

Parity Transformation

The inversion(parity) operation reverses the intrinsic configurations of two deformed nuclei as well as the direction of the relative vector $\boldsymbol{R}$, the x'-axis and z'-axis of the molecular frame. Accordingly the axes of the new molecular frame, $(-x', y', -z')$ is obtained by the rotations $(\pi+\theta_1, \pi-\theta_2, \pi)$. On the intrinsic coordinates, the inversion operator acts as $(\alpha_1, \beta_1, \alpha_2, \beta_2) \rightarrow (2\pi-\alpha_1, \beta_1, 2\pi-\alpha_2, \beta_2)$, where β_i remain unchanged because both the molecular z'-axis and the principal axes z'' are simultaneously reversed. Consequently the inversion operator $\mathcal{P}$ acts as

$$\mathcal{P} : (\theta_1, \theta_2, \theta_3, \alpha, R, \beta_1, \beta_2) \rightarrow (\pi+\theta_1, \pi-\theta_2, \pi-\theta_3, -\alpha, R, \beta_1, \beta_2) \tag{4.98}$$

It should be noted that the definition $\theta_3 = (\alpha_1+\alpha_2)/2$ gives a result $\theta_3 \rightarrow 2\pi-\theta_3$ with $2\pi-\alpha_1$, whose angles are referred to the $-x'$-axis of the new molecular frame, while π of $\pi-\theta_3$ in the above equation is referring to the old x'-axis.

Symmetrized wave functions

Assuming axial symmetry for each deformed cluster, the γ angle will not be affected by any symmery transformation. Also, the density profile of each cluster will be invariant under space inversion due to the positive parity of the ground state. The basis wave function $\phi^0 =$

$D^{I^*}_{MK}(\theta_i)e^{i\nu\alpha}\chi_K(R,\beta_1,\beta_2)$ must be then invariant under action of the inversion operation upon a constituent nucleus,

$$\mathcal{J}_i : (\alpha_i,\beta_i) \longrightarrow (\alpha_i+\pi,\pi-\beta_i) \tag{4.99}$$

Consequently the symmetrization of the wave function is carried out by acting on it with the operator $1+\mathcal{J}_1+\mathcal{J}_2+\mathcal{J}_1\mathcal{J}_2$:

$$\phi^\lambda \equiv (1+\mathcal{J}_1+\mathcal{J}_2+\mathcal{J}_1\mathcal{J}_2)\phi^0 = D^{I^*}_{MK}(\theta_i)e^{i\nu\alpha}\tilde{\chi}_K(R,\beta_1,\beta_2) \tag{4.100}$$

with

$$\begin{aligned}\tilde{\chi}_K(R,\beta_1,\beta_2) &= \chi_K(R,\beta_1,\beta_2)+e^{iK\pi}\chi_K(R,\pi-\beta_1,\pi-\beta_2)\\ &+ e^{i\frac{\pi}{2}(K+\nu)}\chi_K(R,\pi-\beta_1,\beta_2)+e^{i\frac{\pi}{2}(K-\nu)}\chi_K(R,\beta_1,\pi-\beta_2)\end{aligned} \tag{4.101}$$

To the above wave function we apply the boson symmetrization and parity transformation and we use the properties of the Wigner functions, given in App. A.1

$$\begin{aligned}&\frac{1}{2}(1+\mathcal{P}_{12})\frac{1}{2}(1+(-1)^p\mathcal{P})\phi^\lambda\\ =\ & D^{I^*}_{MK}(\theta_i)[e^{i\nu\alpha}\tilde{\chi}_K(R,\beta_1,\beta_2)+(-1)^{p+K}e^{-i\nu\alpha}\tilde{\chi}_K(R,\pi-\beta_2,\pi-\beta_1)]+\\ +\ & (-1)^{p+I-K}D^{I^*}_{M-K}\\ \times\ & [e^{-i\nu\alpha}\tilde{\chi}_{-K}(R,\beta_1,\beta_2)+(-1)^{p+K}e^{i\nu\alpha}\tilde{\chi}_{-K}(R,\pi-\beta_2,\pi-\beta_1)]\end{aligned} \tag{4.102}$$

It should also be noted that due to the periodicity with respect to precessional motions of the constituents nuclei $(K\pm\nu)/2=m$, m being an integer. A remarkable relation is then obtained:

$$\tilde{\chi}_K(R,\pi-\beta_1,\pi-\beta_2) = (-1)^K\tilde{\chi}_K(R,\beta_1,\beta_2) \tag{4.103}$$

and eq.(4.102) can be further simplified

$$\begin{aligned}&\frac{1}{2}(1+\mathcal{P}_{12})\frac{1}{2}(1+(-1)^p\mathcal{P})\phi^\lambda\\ =\ & D^{I^*}_{MK}(\theta_i)[e^{i\nu\alpha}\tilde{\chi}_K(R,\beta_1,\beta_2)+(-1)^{p+}e^{-i\nu\alpha}\tilde{\chi}_K(R,\beta_2,\beta_1)]+\\ +\ & (-1)^{p+I-K}D^{I^*}_{M-K}\\ \times\ & [e^{-i\nu\alpha}\tilde{\chi}_{-K}(R,\beta_1,\beta_2)+(-1)^{p}e^{i\nu\alpha}\tilde{\chi}_{-K}(R,\beta_2,\beta_1)]\end{aligned} \tag{4.104}$$

In particular for $K=0$

$$\phi^\lambda(K=0,-\nu) = (-1)^{p+I}\phi^\lambda(K=0,\nu) \tag{4.105}$$

due to the parity transformation. Thus, $I=even$ corresponds to positive-parity states and $I=odd$ to the negative-parity states.

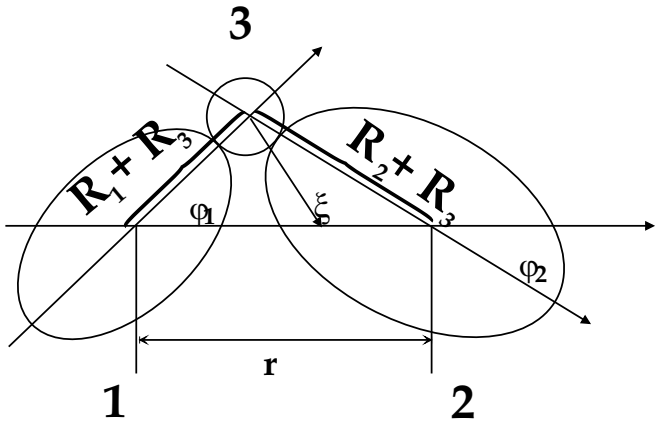

Figure 4.1: Illustration of the main variables of a three cluster molecule. The light cluster is plotted as spherical. In general it will be deformed as the heavy nuclei.

4.2 Geometrical Hamiltonian of Trinuclear Systems

4.2.1 Trinuclear Quasimolecules with Deformed Clusters

In [3] a phenomenological, geometrical model for the system of three clusters, two heavy ones and a light in the middle, was proposed. The model was restricted to butterfly and belly-dancer modes only and the ^{10}Be cluster was assumed to be spherical. However, in general the light cluster can be deformed and its effect must be studied. Also the inclusion of β and γ vibrations must be considered. Because the light cluster in [3] was considered to be spherical, the stiffness of the butterfly motion is mainly determined by the monopole part of the Coulomb repulsion between the heavy fragments [4]. In the case when the light fragment is deformed, this is no longer true and one has to determine explicitely the change of the nuclear and Coulomb interaction between the light and heavy clusters as a function of the inclination angle. In Ref. [4] the nuclear potential was taken into account, including also multipoles higher than the monopole and quadrupole ones.

The model proposed in Ref.[3] is an extension of Ref. [5] presented for two clusters. The molecule exhibits butterfly and belly-dancer modes, β- and γ-vibrations of the two clusters. This picture can be extended straightforwardly to three clusters using the formulas as given in Ref. [5]. It is especially easy for the case when the two big clusters (for example the ^{96}Sr and ^{146}Ba) are connected via a smaller spherical nucleus (^{10}Be). The situation is illustrated in Fig. 4.1, where the main dynamical variables, with a spherical cluster in the middle, are indicated.

In Fig.4.2 possible vibrational modes are indicated, when the light cluster is deformed. The first case corresponds to the above mentioned butterfly mode, whereas the second one is the antibutterfly mode. As will be seen bellow, for the butterfly motion the dominant contribution comes from the movement of the center of masses and, contrary to the two cluster case, deformations play a minor role, except in fixing the length of the axis in Fig. 4.1 and 4.2.

In order to keep the problem tractable, the main assumptions are that the light cluster is

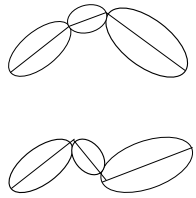

Figure 4.2: In the upper half the butterfly mode is plotted. The lower half presents a more complex case which corresponds to the anti butterfly mode in the two nuclear molecule for the limit of vanishing mass of the light cluster.

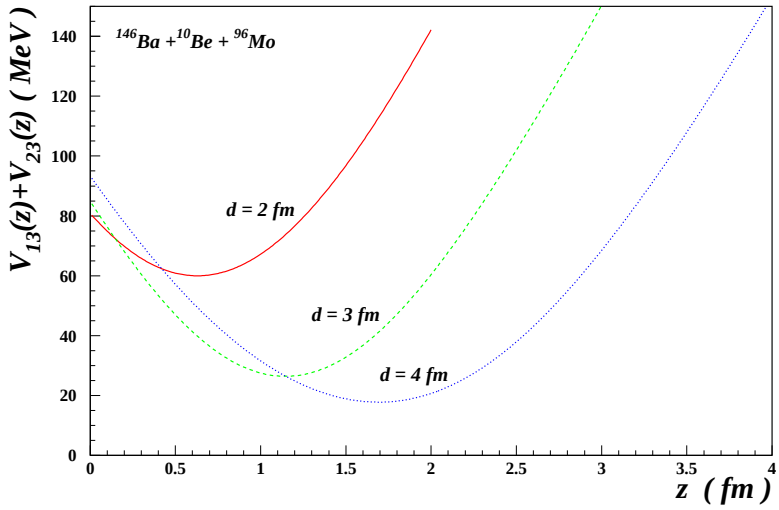

Figure 4.3: The potential of the light cluster 3 in the field of the two heavier fragments along the molecular z axis, for three fixed tip distances : d= 2 (solid lines), 3 (dashed line), 4 (dotted line) FM. The trinuclear molecule comprises ^{96}Sr + ^{10}Be + ^{146}Ba.

sandwiched in-between the heavy nuclei and the inclination angles of the clusters are small with respect to the molecular (fission) z-axis, connecting both heavy clusters. In such a linear chain configuration the total potential of the lighter cluster has an absolute minimum on the axis joining the three fragments. As can be seen in Fig.4.3, for a given distance d between the tips of the two heavier fragments, there is a point on the fission axis z, where the forces exerted by the heavy fragment 1 on the light cluster are canceled by the forces exerted by the heavy fragment 2. This is the so-called electro-nuclear saddle point [6]. Note that the result of Fig.4.3 was obtained using a strong nuclear repulsive force, between the fragments in order to avoid their mutual overlap.

The general formalism

The motion of the three clusters can be divided into the rotations of the individual clusters plus the motion of their center of masses with respect to each other. The part of the Hamiltonian which describes the individual rotations is given in the previous section and can be

also read off from Eqs. (49) and (50) of Ref. [5]. The part which changes originates from the motion of the center of masses. Therefore, the discussion concentrates first on the motion of the three center of masses and is independent of considering either of the two modes described in Fig. 4.2.

In order to separate the center-of-mass motion, the following coordinates are introduced

$$\begin{aligned} \boldsymbol{r} &= \boldsymbol{r_2} - \boldsymbol{r_1} \\ \boldsymbol{\xi} &= \frac{m_1\boldsymbol{r_1} + m_2\boldsymbol{r_2}}{m_1 + m_2} - \boldsymbol{r_3} \end{aligned} \tag{4.106}$$

where $m_k = A_k m$, A_k is the number of nucleons of cluster no. k and m the nucleon mass. The first coordinate ($\boldsymbol{r}$) describes the relative distance of the two heavy fragments while the second one ($\boldsymbol{\xi}$) the distance of the third, lightest cluster to the center of mass of the first two. The kinetic energy, excluding the motion of the total center of mass, acquires the form

$$T = \frac{1}{2}\mu_{12}\dot{\boldsymbol{r}}^2 + \frac{1}{2}\mu_{(12)3}\dot{\boldsymbol{\xi}}^2 + \frac{1}{2}{}^t\boldsymbol{\omega}_1\boldsymbol{\mathcal{J}}_1\boldsymbol{\omega}_1 + \frac{1}{2}{}^t\boldsymbol{\omega}_2\boldsymbol{\mathcal{J}}_2\boldsymbol{\omega}_2 + \frac{1}{2}{}^t\boldsymbol{\omega}_3\boldsymbol{\mathcal{J}}_3\boldsymbol{\omega}_3 \tag{4.107}$$

where $\mu_{12} = \frac{m_1 m_2}{2(m_1+m_2)}$ and $\mu_{(12)3} = \frac{m_3(m_1+m_2)}{2(m_1+m_2+m_3)}$. The first term in (4.107) describes the kinetic energy of the two heavy clusters with respect to each other and the second term the kinetic energy of the third cluster with respect to a mass $(m_1 + m_2)$ at the center of mass of the first two clusters. The mass factors describe the reduced mass for each case. The term proportional to $\dot{\boldsymbol{r}}^2$ has the same form as for the two cluster case and thus is already included in the considerations of the previous section. The last three terms in eq.(4.107) are describing the rotational motion of the three clusters with angular velocities $\boldsymbol{\omega}_{1,2,3}$, referred to the laboratory frame. The inertia tensors $\boldsymbol{\mathcal{J}}_i$ are defined in the intrinsic frame such that in the absence of β and γ vibrations the only non-vanishing components are the first two diagonal terms, $(\boldsymbol{\mathcal{J}}_i)_{11} = (\boldsymbol{\mathcal{J}}_i)_{22} \equiv J_i$, the quantum rotation around the symmetry axis of any of the two heavier fragments being discarded. When β and γ vibrations are included there will be a contribution to $(\boldsymbol{\mathcal{J}}_i)_{33}$ given by a γ dependence [1].

The second term in Eq. (4.107) needs more attention. In Fig. 4.4 the three center of masses are plotted and the relevant coordinates are indicated. The projection of the vector $\boldsymbol{\xi}$ onto the relative distance vector, denoted by ξ_z, and its perpendicular component along the x-axis (ξ_x) are given by

$$\begin{aligned} \xi_z &= \frac{A_2}{(A_1 + A_2)} r - r_{13}\cos\varepsilon \\ \xi_x &= r_{13}\sin\varepsilon \end{aligned} \tag{4.108}$$

where r_{13} is the distance between the cluster no. 1 and 3 and ε is the angle between the axis connecting cluster 1 and 3 to the vector $\boldsymbol{r}$. Note, that this angle is not necessarily the same as the inclination angle of the intrinsic z-axis of cluster no. 1 to the vector $\boldsymbol{r}$. Because the molecular plane is defined by the three center of masses of the clusters the spherical components $\xi_{\pm 1}$ are the same up to a sign. The change in sign in the definition of the spherical components, due to convenience, compared to the usual definition [7] is shown bellow. The $\boldsymbol{\xi}$ contribution of the kinetic energy is obtained by rotating into the molecular

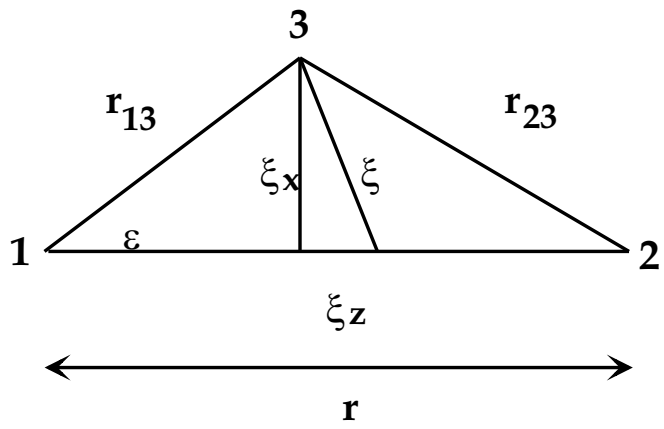

Figure 4.4: Main variables for the discussion of the motion of the clusters with respect to each other. Only the centers of mass of the nuclei are plotted.

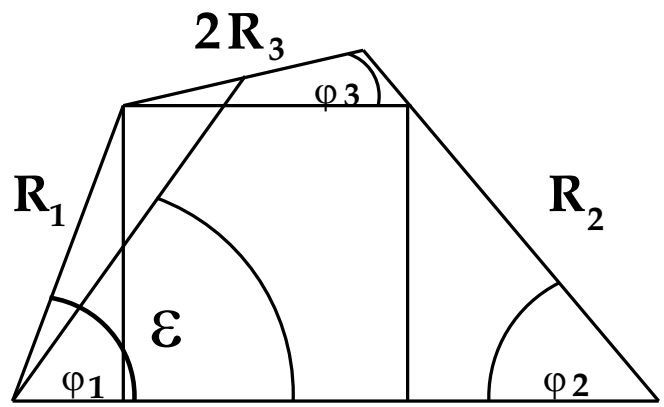

Figure 4.5: In case the third cluster is deformed, the alignment of a heavy nucleus with the light one is not perfect. The relations of the angles are illustrated in this figure.

system and then substituting the expressions for the spherical components of the vector $\boldsymbol{\xi}$. As spherical components we use the expression

$$\begin{aligned} \xi_{\pm 1} &= \pm\frac{1}{\sqrt{2}}(\xi_x \pm \xi_y) \\ \xi_0 &= \xi_z \end{aligned} \tag{4.109}$$

where the definition of $\xi_{\pm 1}$ differs from the usual one [7] for convenience. In the molecular frame the vector $\boldsymbol{\xi}$ is defined to lie in the molecular plane given by the molecular z axis, parallel to the vector $\boldsymbol{r}$. Therefore, the ξ_y component vanishes and the relation of the cartesian to the spherical components is such that the $\xi_{\pm 1}$ components are in their absolute value identical and are given by $\pm\frac{1}{\sqrt{2}}\xi_x$.

In Fig. 4.5 the relation of the angle φ_3 to φ_1 and φ_2 can be read off with the supposition that the three clusters are connected. Assuming small angles, the result is

$$\varphi_3 \approx \sin\varphi_3 = \frac{1}{2R_3}(R_2 \sin\varphi_2 - R_1 \sin\varphi_1) \approx \frac{1}{2R_3}(R_2\varphi_2 - R_1\varphi_1) \tag{4.110}$$

In the above formula we suppose that the heavy cluster 2 has a larger shift on the x-direction than the lighter one. In principle the two angles φ_1 and φ_2 could be treated now as independent with φ_3 constrained by Eq. (4.110). However, the problem would get too complicated, implying coupling terms between the φ_1 and φ_2 motion. Supposing that the cluster in the middle is small, the relation between φ_1 and φ_2 should not differ much from the case when the small cluster is spherical [3]. We use, therefore, the same relation $\varphi_2 \approx \frac{(R_1+R_3)}{(R_2+R_3)}\varphi_1$ as for a spherical cluster and substitute it into Eq. (4.110) resulting in ($\varphi_1 \approx \varepsilon$)

$$\varphi_3 \approx \frac{1}{2}\frac{R_2 - R_1}{R_2 + R_3}\varepsilon \quad . \tag{4.111}$$

One possibility to relax this constraint is to expand the general motion around the point where condition (4.111) holds and diagonalizes the Hamiltonian in this basis.

For the case of connected clusters the distance between center of masses of the third cluster and nucleus no.1, R_{13}, can be approximated by $(R_1 + R_3)$ in the linear, unperturbed configuration. In the perturbed case, when the molecule ceases to be linear, r_{13} is modified by the amount

$$\delta r_{13} \approx -\frac{1}{8}\frac{R_1 R_3}{R_1 + R_3}\left(\frac{R_1 + R_2 + 2R_3}{R_2 + R_3}\right)^2 \varepsilon^2 \tag{4.112}$$

provided ε is small, Similarly we get the variation of r_{23}

$$\delta r_{23} \approx -\frac{1}{8}\frac{R_2 R_3}{R_2 + R_3}\left(\frac{R_1 + R_2 + 2R_3}{R_2 + R_3}\right)^2 \varepsilon^2 \tag{4.113}$$

At equilibrium (ε=0), the distance r, between the center of masses of the two heavy clusters is given by $R_0 \equiv R_1 + R_2 + 2R_3$. When $\varepsilon \neq 0$, and taking into account terms up to second order in ε, r changes to

$$r \approx (R_1 + 2R_3 + R_2)\left(1 - \frac{1}{2}\frac{R_1 + R_3}{R_2 + R_3}\varepsilon^2\right) + \delta r_{13} + \delta r_{23}. \tag{4.114}$$

Therefore, an increase in ε, changes r, r_{13} and r_{23} with a correction in the moment of inertia of the order of ε^2 and higher. At their turn, the components ξ_z and ξ_x acquire the form

$$\begin{aligned}
\xi_z &\approx \left[\frac{A_2}{(A_1 + A_2)}(R_1 + 2R_3 + R_2) - (R_1 + R_3)\right] \\
&+ \frac{A_2}{A_1 + A_2}\delta r_{12} - \delta r_{13} + \frac{(R_1 + R_3)}{2}\varepsilon^2 \\
&= \xi_0 + \delta\xi_z \\
\xi_x &\approx (R_1 + R_3)\varepsilon + \delta\xi_x \quad .
\end{aligned} \tag{4.115}$$

where $\delta\xi_z$ and $\delta\xi_x$ give the contributions due to changes in the relative distance of the clusters 1 to 3 and 2 to 3, i.e. they describe a mode of the stretching vibrations. Again, for simplicity we assume that the relative vibrations are along the molecular z axis, i.e. $\delta\xi_x = 0$. If this assumption is not made, there will be contributions of the type $\delta\dot{\xi}_x\delta\dot{\xi}_z$ in the kinetic energy. One has then to construct the basis first, where this coupling term is absent, and

afterwards to diagonalize the complete Hamiltonian in this basis. Note that the model is quite crude and other coupling terms, like rotation- vibration interactions, have been neglected. Consequently, the spectrum obtained in the model will be a first approximation and the assumptions made above are justified only in this context.

In order to keep the procedure manageable, we assume that the angle ε is small, i.e. the light, third cluster is not far away from the axis connecting the two heavy clusters. Furthermore, we assume that the first cluster, supposed to be the lightest one of the two heavy clusters, touches the third one and the third cluster touches the second one. The situation is illustrated in Fig. 4.5 with a certain exaggeration as concerned to the distance of the third cluster to the axis $(\boldsymbol{r})$ connecting the heavy clusters. These assumptions exclude the anti butterfly mode for which the relation of the angles change. The procedure for that mode, however, is completely analogous.

Neglecting tems of the order $O(\varepsilon^3)$ in Eq.(4.115) we obtain

$$\begin{aligned} \xi_x &= (R_1+R_3)\varepsilon \\ \xi_z &= \xi_0 + \frac{1}{2}\frac{R_1+R_3}{R_2+R_3}\frac{A_1R_2-A_2R_1}{A_1+A_2}\varepsilon^2 \end{aligned} \tag{4.116}$$

where ξ_0 is the value of the ξ-coordinate in the linear chain configuration

$$\xi_0 = \frac{A_2(R_2+R_3)-A_1(R_1+R_3)}{A_1+A_2} \tag{4.117}$$

The time derivative of ξ_m^{lab} is determined using the procedure outlined in Ref. [1]. After some algebra, which implies also the calculation of Wigner D-functions time derivatives we arrive at

$$\begin{aligned} (\dot{\xi}^{lab}\cdot\dot{\xi}^{lab}) \equiv \sum_m (-1)^m \xi_m^{lab}\xi_{-m}^{lab} &\approx (R_1+R_3)^2\dot{\varepsilon}^2 + \xi_0^2(\omega_1'^2+\omega_2'^2) + (R_1+R_3)^2\varepsilon^2\omega_3'^{\,2} \\ &+ 2\xi_0(R_1+R_3)(\dot{\varepsilon}\omega_2' - \varepsilon\omega_1'\omega_2') \quad . \end{aligned} \tag{4.118}$$

The radial mode ξ along the z axis describes the motion of the two heavy clusters in a common direction and of the light cluster in the opposite direction (see Ref. [3]). The other radial mode comes from changes in r and describes a vibration of the two heavy clusters with respect to each other while the small cluster does not move. These types of vibrations will be included in the complete Hamiltonian. Inserting (4.115) into the kinetic energy for the $\boldsymbol{\xi}$ motion, using $\delta\xi_x = 0$, we obtain

$$\begin{aligned} T_\xi &= \frac{1}{2}\mu_{(12)3}\left\{(R_1+R_3)^2\dot{\varepsilon}^2 + \xi_0^2(\omega_1'^{\,2}+\omega_2'^{\,2}) + (R_1+R_3)^2\varepsilon^2\omega_3'^{\,2}\right. \\ &+ \left. 2\xi_0(R_1+R_3)(\dot{\varepsilon}\omega_2' - \varepsilon\omega_1'\omega_2')\right\} \quad , \end{aligned} \tag{4.119}$$

with ω_k' being the angular velocity around the k'th molecular axis ($1=x$, $2=y$ and $3=z$) and ξ is a shorthand notation for $\delta\xi_z$. From here on, when we refer to the total contribution, including the rotational one, we continue to denote it by $\boldsymbol{\xi}$ and the pure motion along the z axis is denoted by ξ.

The kinetic energy of the r and ξ stretching vibrations along the molecular z axis is described by

$$T_r = \frac{1}{2}\mu_{12}\dot{r}^2 + \frac{1}{2}\mu_{(12)3}\dot{\xi}^2 \quad , \tag{4.120}$$

where the last term comes from Eq. (4.119). The coordinates r and ξ are related to the relative distances $r_{13} = z_3 - z_1$ and $r_{23} = z_2 - z_3$ via

$$\begin{aligned} z_2 - z_3 &= \frac{m_1}{m_1 + m_2} r + \xi \\ z_3 - z_1 &= \frac{m_2}{m_1 + m_2} r - \xi \quad . \end{aligned} \tag{4.121}$$

The corresponding potential is given by

$$V_r = \frac{C_{13}}{2}(r_3 - r_1 - r_{13,0})^2 + \frac{C_{23}}{2}(r_2 - r_3 - r_{23,0})^2 \tag{4.122}$$

where $r_{13,0}$ and $r_{23,0}$ are the equilibrium positions $r_{13,0} = (R_1 + R_3)$ and $r_{23,0} = (R_2 + R_3)$ respectively. Note that in our picture cluster no. 1 is to the left and no. 2 to the right and that the coordinates refer to the distance along the intermolecular axis.

Up to know we considered the $\boldsymbol{r}$ and the $\boldsymbol{\xi}$ motion only. The contributions coming from the deformation of the individual clusters can be read off from Ref. [5] and from there the general kinetic energy can be constructed.

Bellow two different cases are considered. In the first one the contributions of β and γ vibrations are excluded and in the second one they are included. In both cases the static deformation in the ground state is assumed to be prolate. This restriction is governed by the necessity to keep the problem solvable otherwise the complicated form of the Hamiltonian would prevent an analytical solution. In case a triaxial nucleus is present the procedure outlined is strictly speaking not valid, but an approximation of the triaxial nucleus by an axial symmetric would do the job.

With the assumption made above, the total kinetic energy is given by

$$\begin{aligned} T &= \frac{1}{2}\Theta_{11}({\omega_1'}^2 + {\omega_2'}^2) + \frac{1}{2}\Theta_{33}{\omega_3'}^2 - \Theta_{13}\varepsilon\omega_1'\omega_3' + \frac{1}{2}\Theta_\varepsilon\dot{\varepsilon}^2 + \Theta_{2\varepsilon}\dot{\varepsilon}\dot{\omega}_2' \\ &+ \frac{1}{2}\mu_{12}\dot{r}^2 + \frac{1}{2}\mu_{(12)3}\dot{\xi}^2 \end{aligned} \tag{4.123}$$

with

$$\begin{aligned} \Theta_{11} &\approx J_1 + J_2 + J_3 + \mu_{(12)3}\xi_0^2 + \mu_{12}R_0^2 \\ \Theta_{33} &\approx \left(J_1 + J_2\frac{(R_1+R_3)^2}{(R_2+R_3)^2} + J_3\frac{(R_1-R_2)^2}{4(R_2+R_3)^2} + \mu_{(12)3}(R_1+R_3)^2 \right)\varepsilon^2 \\ \Theta_{13} &\approx J_1 + J_2\frac{(R_1+R_3)}{(R_2+R_3)} + J_3\frac{(R_1-R_2)}{2(R_2+R_3)} + \mu_{(12)3}\xi_0(R_1+R_3) \\ \Theta_\varepsilon &= \frac{1}{\varepsilon^2}\Theta_{33} \\ \Theta_{2\varepsilon} &= \Theta_{13} \end{aligned} \tag{4.124}$$

For nearly symmetric heavy clusters, not too deformed and a small value of $\frac{m_3}{m_1+m_2}$ the coupling is small compared to the diagonal terms of the moment of inertia and thus can be neglected, as for the other contributions. Also the deformation dependent part in Θ_{11} can be neglected with respect to the last term, as was done also in Ref. [5]. However, for very asymmetric heavy clusters we cannot neglect any more the contribution of $\Theta_{2\varepsilon}$, except for small $\frac{m_3}{m_1+m_2}$. In this case, one has to diagonalize this term in the basis with $\Theta_{2\varepsilon} = 0$, which is in the same spirit as for the coupling terms in the radial vibrations. The basis will be discussed further below.

For the potential we assume a quadratic behaviour in ε, r and ξ, i.e.

$$V = \frac{C_\varepsilon}{2}\varepsilon^2 + \frac{C_r}{2}\bar{r}^2 + \frac{C_\xi}{2}(\xi - \xi_0)^2 \quad , \tag{4.125}$$

where $\bar{r} = (r - r_0)$ with r_0 being the equilibrium position of the nuclear molecule. The parameters C_r and C_ξ are related to C_{13} and C_{23} via

$$\begin{aligned} C_r &= \frac{(m_2^2 C_{13} + m_1^2 C_{23})}{(m_1+m_2)^2} \\ C_\xi &= C_{13} + C_{23} \quad . \end{aligned} \tag{4.126}$$

Other crossing terms of the type $r\xi$, r and ξ also appear, which vanish for a symmetric dinuclear sub-system, formed from clusters 1 and 2. We assume that in general the microscopic interaction is such that also for non-symmetric clusters the coupling terms can be canceled or made small, which is obviously a simplification.

In what follows, we quantize the Hamiltonian with the kinetic energy given in (4.123) taking into account the contribution of the coupling of the ε and ω_2 motion, whose origin is the Coriolis force. The quantization procedure was already explained in 4.1. Then, the Hamiltonian composed by the kinetic energy (4.123) and the potential (4.125) is quantized in this manner, resulting for the kinetic energy in

$$\begin{aligned} \widehat{T} \approx\ & \frac{\hbar^2(\hat{I}^2 - \hat{I}_3'^2)}{2(\Theta_{11} - \frac{\Theta_{13}^2}{\Theta_\varepsilon})} + \frac{\hbar^2 \hat{I}_3'^2}{2(\Theta_\varepsilon - \frac{\Theta_{13}^2}{\Theta_{11}})\varepsilon^2} - \frac{\hbar^2}{2(\Theta_\varepsilon - \frac{\Theta_{13}^2}{\Theta_{11}})}\left(\frac{\partial^2}{\partial\varepsilon^2} + \frac{1}{4\varepsilon^2}\right) \\ & - \frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\bar{r}^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\xi^2} + \frac{\Theta_{13}}{(\Theta_{11}\Theta_\varepsilon - \Theta_{13}^2)}\left[\frac{\hbar^2}{\varepsilon}\hat{I}_1'\hat{I}_3' - \hbar\hat{I}_2'\left(\frac{\hbar}{i}\frac{\partial}{\partial\varepsilon}\right)\right] \end{aligned} \tag{4.127}$$

In the following discussion we will skip the term in the parenthesis $\{...\}$ which has to be treated as a perturbative interaction and the corrections given by $\frac{\Theta_{13}^2}{\Theta_\varepsilon\Theta_{11}}$, which is small compared to one for the molecular systems studied in the present case. In very assymmetric systems, however, both terms have to be included and diagonalized in the basis which will be constructed in what follows. The terms which do not contain a derivative come from the additional potential of Eq. (4.63)

Neglecting the term with the parenthesis $\{...\}$ in Eq. (4.127) and corrections of the order of $\frac{\Theta_{13}^2}{\Theta_{11}}$ and $\frac{\Theta_{13}^2}{\Theta_\varepsilon}$, the corresponding static Schrödinger equation can be solved with the ansatz

$$\phi = D_{MK}^{I^*}(\vartheta)\chi_{K,n_\varepsilon}(\varepsilon)g_{n_r}(r) \tag{4.128}$$

were $D^{I^*}_{MK}(\vartheta)$ is the Wigner D-matrix, $g_{n_r}(r)$ is the one dimensional harmonic oscillator for the relative motion and χ_{K,n_ε} is the solution of the differential equation

$$\begin{aligned} & \left[-\frac{\hbar^2}{2\Theta_\varepsilon}\frac{\partial}{\partial\varepsilon^2} + \frac{1}{\Theta_\varepsilon}(K^2 - \frac{1}{4})\frac{\hbar^2}{2\varepsilon^2} + \frac{C_\varepsilon}{2}\varepsilon^2 \right. \\ + & \left. \left(\frac{\hbar^2}{2\Theta_{11}}[I(I+1) - K^2] + \hbar\omega_r(n_r + \frac{1}{2}) + \hbar\omega_\xi(n_\xi + \frac{1}{2}) - E \right) \right] \phi = 0 \end{aligned} \quad (4.129)$$

where I is the total spin, K its projection to the molecular z axis, $\hbar\omega_r = \hbar\sqrt{\frac{C_r}{\mu_{(12)}}}$, $\hbar\omega_\xi = \hbar\sqrt{\frac{C_\xi}{\mu_{3(12)}}}$ and E the total energy of the state ϕ.

This equation can be solved with the solution given by (see also Ref. [1])

$$\chi_{K,n\varepsilon}(\varepsilon) = \frac{\left\{ \lambda^{l_k+\frac{3}{2}}\Gamma(l_K + \frac{3}{2} + n_\varepsilon) \right\}^{\frac{1}{2}}}{(n_\varepsilon!)^{\frac{1}{2}}\Gamma(l_K + \frac{3}{2})} \varepsilon^{l_K+1} e^{-\frac{1}{2}\lambda\varepsilon^2}\ {}_1F_1(-n_\varepsilon, l_K + \frac{3}{2}; \lambda\varepsilon^2) \quad , \quad (4.130)$$

where $\lambda^2 = \frac{\Theta_\varepsilon C_\varepsilon}{\hbar^2}$, $l_K = \mid K \mid - \frac{1}{2}$ and ${}_1F_1(...)$ is the confluent hypergeometric function.

The total energy is given by

$$E = \frac{\hbar^2}{2\Theta_{11}}\left[I(I+1) - K^2\right] + \hbar\omega_r(n_r + \frac{1}{2}) + \hbar\omega_\varepsilon(\mid K \mid + 2n_\varepsilon + \frac{3}{2}) \quad (4.131)$$

and $\hbar\omega_\varepsilon = \hbar\sqrt{\frac{C_\varepsilon}{\Theta_\varepsilon}}$.

When there are two identical clusters the wavefunction has still to be symmetrized with respect the interchange of cluster no. 1 and 2. For the light cluster the variables transform in the same way as indicated by the second cluster except for the change of indices from 2 to 3. With respect to the variable ε the transformation is $\varepsilon \to -\varepsilon$.

The extension to β and γ vibrations is straightforward. Including also the rotation around the intrinsic z axis of axial symmetric nuclei, given by Φ_k ($k = 1, 2, 3$), leads to a very complicated form. Additionally we have to assume that the clusters are prolately deformed. Otherwise a complex coupling between the rotations around the x, y and z will appear. The β and γ vibrational variables of the k'th cluster are defined by

$$\begin{aligned} \bar{\beta}_k &= a^k_0 - \beta_{0k} \\ \eta_k &= a^k_2 \quad , \end{aligned} \quad (4.132)$$

where a^k_0 and a^k_2 are the components of the quadrupole deformation variable of the k'th cluster with respect to the principal axes. Using Eq. (50) of the second paper in Ref. [5] for the moments of inertia plus the corrections discussed further up, the Hamiltonian acquires

the form

$$
\begin{aligned}
\widehat{T} \;=\; & \frac{\hbar^2(\hat{I}^2-\hat{I}_3'^2)}{2(\Theta_{11}-\frac{\Theta_{13}^2}{\Theta_\varepsilon})}+\frac{\hbar^2\hat{I}_3'^2}{2(\Theta_\varepsilon-\frac{\Theta_{13}^2}{\Theta_{11}})\varepsilon^2}-\frac{\hbar^2}{2(\Theta_\varepsilon-\frac{\Theta_{13}^2}{\Theta_{11}})}\left(\frac{\partial^2}{\partial\varepsilon^2}-\frac{1}{4\varepsilon^2}\right)\\
& -\frac{\hbar I_3'}{\Theta_\varepsilon\varepsilon^2}\left(\frac{\hbar}{i}\frac{\partial}{\partial\Phi_1}+\frac{\hbar}{i}\frac{\partial}{\partial\Phi_2}+\frac{\hbar}{i}\frac{\partial}{\partial\Phi_3}\right)\\
& -\frac{\hbar^2}{2\Theta_\varepsilon\varepsilon^2}\left(\frac{\partial^2}{\partial\Phi_1^2}+\frac{\partial^2}{\partial\Phi_2^2}+\frac{\partial^2}{\partial\Phi_3^2}\right)\\
& -\frac{\hbar^2}{\Theta_\varepsilon\varepsilon^2}\left(\frac{\partial^2}{\partial\Phi_1\partial\Phi_2}+\frac{\partial^2}{\partial\Phi_1\partial\Phi_3}+\frac{\partial^2}{\partial\Phi_2\partial\Phi_3}\right)\\
& -\frac{\hbar^2}{2B_1}\frac{\partial^2}{\partial^2\bar{\beta}_1^2}-\frac{\hbar^2}{2B_2}\frac{\partial^2}{\partial^2\bar{\beta}_2^2}-\frac{\hbar^2}{2B_3}\frac{\partial^2}{\partial^2\bar{\beta}_3^2}\\
& -\frac{\hbar^2}{16B_1\eta_1^2}-\frac{\hbar^2}{16B_3\eta_3^2}-\frac{\hbar^2}{16B_3\eta_3^2}\\
& -\frac{\hbar^2}{4B_1}\frac{\partial^2}{\partial^2\eta_1^2}-\frac{\hbar^2}{4B_2}\frac{\partial^2}{\partial^2\eta_2^2}-\frac{\hbar^2}{4B_3}\frac{\partial^2}{\partial^2\eta_3^2}\\
& -\frac{\hbar^2}{16B_1\eta_1^2}\frac{\partial^2}{\partial\Phi_1^2}-\frac{\hbar^2}{16B_2\eta_2^2}\frac{\partial^2}{\partial\Phi_2^2}-\frac{\hbar^2}{16B_3\eta_3^2}\frac{\partial^2}{\partial\Phi_3^2}\\
& -\frac{\hbar^2}{2\mu_{(12)}}\frac{\partial^2}{\partial\bar{r}^2}-\frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\xi^2}
\end{aligned}
\tag{4.133}
$$

where the variables $\bar{\beta}_k$ and η_k ($k=1,2,3$) describe the β and γ degree of freedom as defined in Ref. [1].

The complete potential is given by

$$
\begin{aligned}
V \;=\; & \frac{C_\varepsilon}{2}\varepsilon^2+\frac{C_r}{2}\bar{r}^2+\frac{C_\xi}{2}(\xi-\xi_0)^2\\
& +C_{\eta_1}\eta_1^2+C_{\eta_2}\eta_2^2+C_{\eta_3}\eta_3^2\\
& +\frac{C_{\beta_1}}{2}\bar{\beta}_1^2+\frac{C_{\beta_2}}{2}\bar{\beta}_2^2+\frac{C_{\beta_3}}{2}\bar{\beta}_3^2
\end{aligned}
\tag{4.134}
$$

and the determinant g is

$$
g \;=\; 8^3 2^3\Theta_{11}^2\Theta_\varepsilon^2 B_1^3B_2^3B_3^3\eta_1^2\eta_2^2\eta_3^2\varepsilon^2 \quad . \tag{4.135}
$$

where $\Theta_{22}\approx\Theta_{11}$ was used. The factor $2^3B_1^2B_2^2B_3^2=(2B_1)(2B_2)(2B_3)\;B_1B_2B_3$ comes from the β and γ part of the metric tensor $g_{\mu\nu}$.

Neglecting, as in the case without β and γ vibrations,corrections of the order of $\frac{\Theta_{13}^2}{\Theta_\varepsilon\Theta_{11}}$ the static Schrödinger equation can be solved with the ansatz

$$
\begin{aligned}
& \phi(\vartheta_1,\vartheta_2,\vartheta_3,\varepsilon,\bar{r},\bar{\beta}_1,\bar{\beta}_2,\bar{\beta}_3,\eta_1,\eta_2,\eta_3)=e^{i(K_1\Phi_1+K_2\Phi_2+K_3\Phi_3)}D_{MK}^{I^*}(\vartheta)\chi_{\tilde{K},n\varepsilon}(\varepsilon)g_{n_r}(\bar{r})\\
& g_{n_\xi}(\xi)g_{n_{\beta_1}}(\bar{\beta}_1)g_{n_{\beta_2}}(\bar{\beta}_2)g_{n_{\beta_3}}(\bar{\beta}_3)\chi_{K_1,n_{\eta_1}}(\eta_1)\chi_{K_2,n_{\eta_2}}(\eta_2)\chi_{K_2,n_{\eta_2}}(\eta_2)
\end{aligned}
\tag{4.136}
$$

where K_k is the eigenvalue of the operator $\frac{1}{i}\frac{\partial}{\partial\Phi_k}$ and $\tilde{K}$ stands for $\mid K-K_1-K_2-K_3\mid$. To obtain the final form of the wave function, we have to apply the transformations of the coordinate symmetries for a molecule composed of prolate nuclei.

The transformations of the molecular frame consists of the operators $\hat{R}_{1,m}$ and $\hat{R}^2_{2,m}$, the m refers to the molecular frame, and their action is given by

$$\begin{aligned} \hat{R}_{1,m}(x,y,z) &= (x,-y,-z) \\ \hat{R}_{2,m}(x,y,z) &= (-x,-y,z) \quad . \end{aligned} \tag{4.137}$$

These operators act on the Euler angles ϑ_k ($k = 1,2,3$), the other coordinates χ_i, ϕ_i, Φ_i ($i = 1,2,3$) and on $\bar{r}$ and ξ. The result is given in Table 4.1. The angles χ_i, ϕ_i and Φ_i correspond to the Euler angles describing the rotation of the nucleus from the molecular frame to the principal axis of the i-th cluster.

Because the three clusters are supposed to lie in a plane the angles χ_i are put to zero. Also a small butterfly angle is assumed and all angles φ_i are proportional to it. A coordinate symmetry transformation consists of those actions where the angels χ_i and φ_i are changed at most by a sign. Inspecting Table 4.1, the only combinations allowed are $\hat{R}_{1,m}\hat{R}_{1,p_i}$ and $\hat{R}_{2p_i}$ whose action is given in Table 4.2.

These operators have to be applied to the solution

$$\begin{aligned} &\phi(\vartheta_1,\vartheta_2,\vartheta_3,\varepsilon,\bar{r},\bar{\beta}_1,\bar{\beta}_2,\bar{\beta}_3,\eta_1,\eta_2,\eta_3) = e^{i(K_1\Phi_1+K_2\Phi_2+K_3\Phi_3)} D^{I*}_{MK}(\vartheta)\chi_{\tilde{K},n\varepsilon}(\varepsilon) g_{n_r}(\bar{r}) \\ &\times g_{n_\xi}(\xi) g_{n_{\beta_1}}(\bar{\beta}_1) g_{n_{\beta_2}}(\bar{\beta}_2) g_{n_{\beta_3}}(\bar{\beta}_3)\chi_{K_1,n_{\eta_1}}(\eta_1)\chi_{K_2,n_{\eta_2}}(\eta_2)\chi_{K_2,n_{\eta_2}}(\eta_2) \end{aligned} \tag{4.138}$$

of the Schrödinger equation. The action of $\hat{R}_{2,p_i}$ ($i = 1,2,3$) on this state changes Φ_i to $\Phi_i \pm \frac{\pi}{2}$ and η_i to $-\eta_i$, i.e. it acts only on $e^{iK_i\Phi_i}$ and $\chi_{K_i,n_{\eta_i}}(\eta_i)$. The result is a phase $(-1)^{K_i}$ implying only even values of K_i.

The action of the operator $\hat{R}_{1,m}\ \hat{R}_{1,p_i}$ is more involved. The result is

$$\begin{aligned} &\phi(\vartheta_1,\vartheta_2\vartheta_3,\varepsilon,\bar{r},\bar{\beta}_1,\bar{\beta}_2,\bar{\beta}_3,\eta_1,\eta_2,\eta_3) = \\ &\mathcal{N}\left\{D^{I\ *}_{M\ K}(\vartheta) f(\Phi_1,\Phi_2,\Phi_3) + (-1)^{I-K} D^{I\ *}_{M\ -K}(\vartheta) f(-\Phi_1,-\Phi_2,-\Phi_3)\right\} \\ &\times\chi_{K,n\varepsilon}(\varepsilon) g_{n_r}(\bar{r}) g_{n_\xi}(\xi) g_{n_{\beta_1}}(\bar{\beta}_1) g_{n_{\beta_2}}(\bar{\beta}_2) g_{n_{\beta_3}}(\bar{\beta}_3)\chi_{K_1,n_{\eta_1}}(\eta_1)\chi_{K_2,n_{\eta_2}}(\eta_2)\chi_{K_2,n_{\eta_2}}(\eta_2) \end{aligned} \tag{4.139}$$

where $\mathcal{N}$ is a normalization factor and

$$\begin{aligned} f(\Phi_1,\Phi_2,\Phi_3) &= e^{i(K_1\Phi_1+K_2\Phi_2+K_3\Phi_3)} + e^{-i(K_1\Phi_1+K_2\Phi_2-K_3\Phi_3)} \\ &\quad\ e^{i(K_1\Phi_1-K_2\Phi_2-K_3\Phi_3)} + e^{-i(K_1\Phi_1-K_2\Phi_2+K_3\Phi_3)} \quad . \end{aligned} \tag{4.140}$$

The quantum numbers acquire the possible values

$$\begin{aligned} K_i &= 0,2,4,... \\ K &= 0,1,2,3,4,... \\ L &= K, K+1, K+2,... \\ n_r,\ n_\xi,\ n_\varepsilon,\ n_{\beta_i},\ n_{\eta_i} &= 0,1,2,3,4,... \quad . \end{aligned} \tag{4.141}$$

variable	$\hat{R}_{1,m}$	$\hat{R}^2_{2,m}$	$\hat{R}_{1,p_i}$	$\hat{R}_{2,p_i}$
ϑ_1	$\vartheta_1+\pi$	ϑ_1	ϑ_1	ϑ_1
ϑ_2	$\pi-\vartheta_2$	ϑ_2	ϑ_2	ϑ_2
ϑ_3	$-\vartheta_3$	$\vartheta_3+\pi$	ϑ_3	ϑ_3
χ_i	$-\chi_i$	$\chi_i+\pi$	χ_i	χ_i
φ_i	$\pi-\varphi_i$	φ_i	$\varphi_i+\pi$	φ_i
Φ_i	$\Phi_i+\pi$	Φ_i	$\pi-\Phi_i$	$\Phi_i+\frac{\pi}{2}$
ξ_i	ξ_i	ξ_i	ξ_i	ξ_i
η_i	η_i	η_i	η_i	$-\eta_i$
$\bar{r}$	$\bar{r}$	$\bar{r}$	$\bar{r}$	$\bar{r}$
ξ	ξ	ξ	ξ	ξ

Table 4.1: The action of the basic coordinate symmetry operators on the collective variables

variable	ϑ_1	ϑ_2	ϑ_3	χ_i	φ_i	Φ_i	ξ_i	η_i	$\bar{r}$	ξ
$\hat{R}_{1,m}\hat{R}_{1,p_i}$	$\vartheta_1+\pi$	$\pi-\vartheta_2$	$-\vartheta_3$	$-\chi_i$	$-\varphi_i$	$-\Phi_i$	ξ_i	η_i	$\bar{r}$	ξ
$\hat{R}_{2,p_i}$	ϑ_1	ϑ_2	ϑ_3	χ_i	φ_i	$\Phi_i+\frac{\pi}{2}$	ξ_i	η_i	$\bar{r}$	ξ

Table 4.2: The action of the allowed combinations of symmetry operators which satisfy the condition that after their application χ_i is still zero and φ_i is maintained near zero.

The energy is

$$\begin{aligned} E &= \frac{\hbar^2}{2\Theta_{11}}[I(I+1)-K^2]+\hbar\omega_\varepsilon(\mid K-K_1-K_2-K_3\mid +2n_\varepsilon+1) \\ &+\hbar\omega_{\beta_1}(n_{\beta_1}+\frac{1}{2})+\hbar\omega_{\beta_2}(n_{\beta_2}+\frac{1}{2})+\hbar\omega_{\beta_3}(n_{\beta_3}+\frac{1}{2}) \\ &+\hbar\omega_{\eta_1}(\frac{1}{2}\mid K_1\mid +2n_{\eta_1}+1)+\hbar\omega_{\eta_2}(\frac{1}{2}\mid K_2\mid +2n_{\eta_2}+1) \\ &+\hbar\omega_{\eta_2}(\frac{1}{2}\mid K_3\mid +2n_{\eta_3}+1)+\hbar\omega_r(n_r+\frac{1}{2})+\hbar\omega_\xi(n_\xi+\frac{1}{2}) \quad . \end{aligned} \tag{4.142}$$

The frequencies are

$$\begin{aligned} \hbar\omega_\varepsilon=\hbar\sqrt{\tfrac{C_\varepsilon}{\Theta_\varepsilon}},\quad \hbar\omega_{\eta_k}=\hbar\sqrt{\tfrac{C_{\eta_k}}{B_k}},\quad \hbar\omega_{\beta_k}=\hbar\sqrt{\tfrac{C_{\beta_k}}{B_{\beta_k}}} \\ \hbar\omega_r=\hbar\sqrt{\tfrac{C_r}{\mu_{(12)}}},\quad \hbar\omega_\xi=\hbar\sqrt{\tfrac{C_\xi}{\mu_{(12)3}}} \quad . \end{aligned} \tag{4.143}$$

Before we apply the outlined procedure to the cases ^{96}Sr + ^{10}Be + ^{146}Be, ^{112}Ru + ^{10}Be + ^{130}Ru and ^{108}Mo + ^{10}Be + ^{134}Te, which are all non-symmetric systems, the derivation of the potential parameters is outlined. The ones related to the β and γ vibrations are obtained through the spectrum of the individual clusters.

Derivation of relative potentials

We derive now the expressions for the stiffness coefficients appearing in Eq.(4.125). For that we need to calculate the interactions between the nuclei composing the trinuclear molecule. Since the interaction potential between the clusters should depend not only on their reciprocal distances but also on orientations we choose again the double folding-model potential written in (3.49).

The ground state one-body nuclear densities of the fragments are taken as Fermi distributions in the intrinsic frame

$$\rho(\boldsymbol{r}) = \rho_0 \left[1 + \exp \frac{1}{a} \left(r - R_0 \left(1 + \sum_\lambda \beta_\lambda Y_{\lambda 0}(\theta, 0) \right) \right) \right]^{-1} . \tag{4.144}$$

The constant ρ_0 is fixed by normalizing the proton and neutron density to the Z proton and N neutron numbers, respectively. This condition ensures the volume conservation. The radius R_0 and diffusivity parameters were taken from liquid drop model calculations [8] for the heavy fragments, whereas for the light cluster we consider the prescription $R_0 = 1.04 \cdot A_3^{1/3}$ for the radius and $a = 0.35$ for the diffusivity. As static deformations, β_λ, we considered quadrupole, octupole and hexadecupole deformations.

In the region of nuclear-density overlap we introduce a phenomenological repulsive potential which originates from the compression effects of the overlapping density as explained in 3.3.3

$$V_{\text{rep}}(\boldsymbol{R}) = V_p \int d\boldsymbol{r}_1 \int d\boldsymbol{r}_2 \, \widetilde{\rho}_1(\boldsymbol{r}_1) \widetilde{\rho}_2(\boldsymbol{r}_2) \delta(\boldsymbol{s}) \tag{4.145}$$

where the tildes on the densities signify a distribution of the same shape as (4.144) but possessing an almost sharp surface. The strength of the compression term V_p was determined from the nuclear equation of state [1] by requiring for total overlap of two nuclei a double normal density of nuclear matter. For a given dinuclear subsystem ($A_{\text{light}} + A_{\text{heavy}}$) we take the value of the nuclear compression modulus K according to the receipt proposed in [9], and compute V_p from the equation giving the binding energy loss for total overlap [10], i.e. $R = 0$:

$$V_{\text{M3Y}}(0) + V_{\text{rep}}(0) \approx \frac{1}{9} K A_{\text{light}} \tag{4.146}$$

The double orientation of the double folded potential is computed by using (3.68).

In [4] it was assumed that when the nuclear molecule is bent the reciprocal distances between the heavy fragments and the light cluster are preserved, i.e. the trinuclear molecule is allowed to perform only vibrations which result in the increase of the angle between the two valence bonds. In this way possible bond stretching vibrations were excluded. To overcome this restriction we take into account such a degree of freedom and by $\delta r_{13(23)}$ we denote the change in the distance between clusters 1(2) and 3. The geometry of the system, presented

in Fig.4.5 for a deformed light cluster, provides the values of these quantities as a function of the deflection angles $\varphi_{1,2,3}$:

$$\begin{aligned}\delta r_{13} &= -\frac{1}{2}\frac{R_1R_3}{(R_1+R_3)}(\varphi_1+\varphi_3)^2\\ \delta r_{23} &= -\frac{1}{2}\frac{R_2R_3}{(R_2+R_3)}(\varphi_2-\varphi_3)^2\end{aligned} \tag{4.147}$$

On the other hand, from the inspection of Fig.4.5, the interaction between the deformed light cluster 3 and the heavier deformed nucleus 1, reads :

$$V(\boldsymbol{r}_{13}) = \sum_{\lambda_1,\lambda_2,\lambda_3} V^{\mu\ -\mu\ 0}_{\lambda_1\ \lambda_2\ \lambda_3}(R_1+R_3+\delta r_{13})d^{\lambda_1}_{\mu 0}(\varphi_1-\varepsilon)d^{\lambda_2}_{-\mu 0}(\varepsilon+\varphi_3) \tag{4.148}$$

Since we made earlier the approximation $\varphi_1 \approx \varepsilon$, the variation of the valence bond length (4.147) reads

$$\delta r_{13} = -\frac{1}{8}\frac{R_1R_3}{R_1+R_3}\left(\frac{R_1+R_2+2R_3}{R_1+R_3}\right)^2\varepsilon^2 \tag{4.149}$$

Expanding the potential (4.148) with respect to the small angle ε we have that

$$V(\boldsymbol{r}_{13}) = V(R_1+R_3) + \frac{1}{2}C^{13}_{\varepsilon}\varepsilon^2 \tag{4.150}$$

where

$$\begin{aligned}C^{13}_{\varepsilon} &= -\frac{1}{4}\left(\frac{R_0}{R_1+R_3}\right)^2\\ &\times \sum_{\lambda_i}\left(\frac{R_1R_3}{R_2+R_3}\frac{\partial V^{0\ 0\ 0}_{\lambda_1\lambda_2\lambda_3}(r_{13})}{\partial r_{13}} - \frac{1}{2}\lambda_2(\lambda_2+1)V^{0\ 0\ 0}_{\lambda_1\lambda_2\lambda_3}(r_{13})\right)_{r_{13}=R_1+R_3}\end{aligned} \tag{4.151}$$

Using similar arguments we get the expression for the stiffness of the butterfly mode, coming from the interaction between clusters 2 and 3

$$\begin{aligned}C^{23}_{\varepsilon} &= -\frac{1}{4}\left(\frac{R_0}{R_2+R_3}\right)^2\\ &\times \sum_{\lambda_i}\left(\frac{R_2R_3}{R_2+R_3}\frac{\partial V^{0\ 0\ 0}_{\lambda_1\lambda_2\lambda_3}(r_{23})}{\partial r_{23}} - \frac{1}{2}\lambda_2(\lambda_2+1)V^{0\ 0\ 0}_{\lambda_1\lambda_2\lambda_3}(r_{23})\right)_{r_{23}=R_2+R_3}\end{aligned} \tag{4.152}$$

The last contribution to the stiffness coefficient of the butterfly mode comes from the interaction of the heavier fragments 1 and 2. Using again the geometry of Fig.4.5 we write in multipolar form the interaction between these nuclei

$$V(\boldsymbol{r}) = \sum_{\lambda_1,\lambda_2,\lambda_3} V^{\mu\ -\mu\ 0}_{\lambda_1\ \lambda_2\ \lambda_3}(R_0+\delta r)d^{\lambda_1}_{\mu 0}(\varphi_1)d^{\lambda_2}_{-\mu 0}(\varphi_2) \tag{4.153}$$

According to eqs.(4.112-4.114) the shift in the interfragment distance reads

$$\delta r = -\frac{1}{2}R_0\frac{R_1+R_3}{R_2+R_3}\varepsilon^2 + \delta r_{13} + \delta r_{23} \tag{4.154}$$

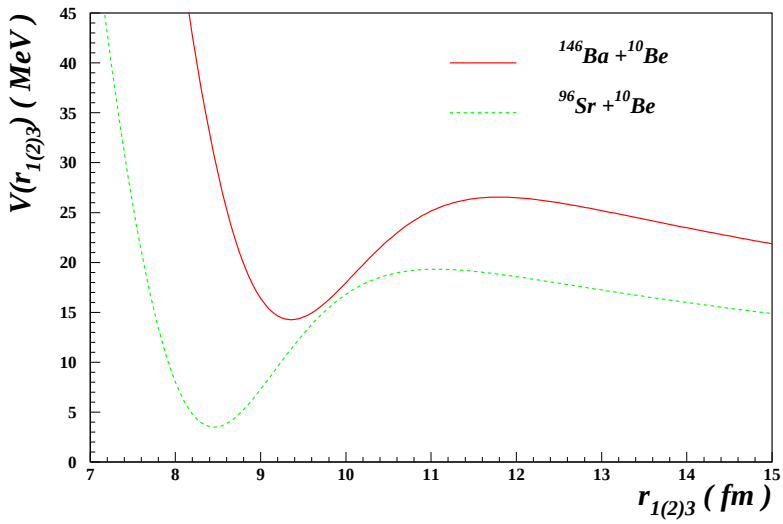

Figure 4.6: The potential between the heavy fragment ^{146}Ba and ^{10}Be (solid line) and between ^{96}Sr and ^{10}Be (dashed line).

Consequently expanding in Taylor series this potential too, we obtain the stiffness coefficient

$$\begin{aligned} C_\varepsilon &= \frac{1}{2}\sum_{\lambda_1\lambda_2\lambda_3}\left(\lambda_1(\lambda_1+1)\frac{R_1-R_2}{R_2+R_3}-\lambda_2(\lambda_2+1)\frac{(R_1-R_2)(R_1+R_3)}{(R_2+R_3)^2}\right. \\ &\qquad \left.-\lambda_3(\lambda_3+1)\frac{R_1+R_3}{R_2+R_3}\right)V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(R_0) \\ &- R_0\sum_{\lambda_1\lambda_2\lambda_3}\left(\frac{R_1+R_3}{R_2+R_3}+\frac{R_0R_3(R_1R_3+R_2R_3+2R_1R_2)}{4(R_1+R_3)(R_2+R_3)^3}\right)\frac{\partial V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(R_0)}{\partial r} \end{aligned} \tag{4.155}$$

In order to obtain the stiffness coefficients of the bond streching vibrations C_{13} and C_{23}, we expand the potentials $V(\boldsymbol{r}_{13})$ and $V(\boldsymbol{r}_{23})$, up to second power of δr_{13} and δr_{23}. Such an expansion is possible in view of the relative minimum in the potential with respect to the inter-cluster distance. As an example we give in Fig. 4.6 the potential between the heavy cluster1(2) and the light cluster 3. After some algebra we obtain for the dinuclear subensemble (13)

$$V(\boldsymbol{r}_{13}) = V(R_1+R_3) + A_{13}\delta r_{13} + \frac{1}{2}C_{13}\delta r_{13}^2 \tag{4.156}$$

where

$$A_{13} = \sum_{\lambda_1,\lambda_2,\lambda_3}\left(\frac{\lambda_2(\lambda_2+1)}{2}\frac{R_1R_3}{R_1+R_3}V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(r_{13})+\frac{\partial V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(r_{13})}{\partial r_{13}}\right)_{r_{13}=R_1+R_3} \tag{4.157}$$

$$C_{13} = \sum_{\lambda_1,\lambda_2,\lambda_3} \left(\frac{\lambda_2(\lambda_2+1)(3\lambda_2^2+3\lambda_2+1)}{24} \left(\frac{R_1+R_3}{R_1R_3}\right)^2 V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(r_{13}) \right.$$
$$\left. + \lambda_2(\lambda_2+1)\frac{R_1+R_3}{R_1R_3}\frac{\partial V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(r_{13})}{\partial r_{13}} + \frac{\partial^2 V^{0\,0\,0}_{\lambda_1\lambda_2\lambda_3}(r_{13})}{\partial r_{13}^2} \right)_{r_{13}=R_1+R_3} \quad (4.158)$$

Since the linear term in δr_{13} has only the effect to shift the origin of the harmonic oscillator, the stiffness coefficient of the bond-stretching vibrations is specified by C_{13}. In the same manner we derive the coefficient C_{23} and next using the relations from eq.(4.126) we derive the stiffness coefficients of the r and ξ-modes.

Applications to ^{10}Be-like molecules

The model developed above is applied next to the systems ^{96}Sr + ^{10}Be + ^{146}Ba, ^{112}Ru + ^{10}Be + ^{130}Sn and ^{108}Mo + ^{10}Be + ^{134}Te. For the computation of the numbers we use mc^2 = 938 MeV for the nuclear mass, $\hbar c$ = 197.33 MeV fm^2, 1.2 $A^{1/3}$ for the spherical equivalent radius of a heavy cluster and $1.3A^{1/3}$ for the light one. In order to obtain $\frac{3\hbar^2}{J_k}$ we set it equal to the energy of the 2_1^+ state of the individual clusters.

In case we consider a spherical nucleus, the $E(2_1^+)$ value is taken as the vibrational energy. This has to be used with caution because for a rotor the 2_1^+ state is a rotational one. In a nuclear molecule the rotation of individual clusters is constrained due to the link to other clusters. Its rotation is converted into the butterfly motion. This is not the case for a vibrational state. We assume a deformed ^{10}Be where the deformation is *not taken* from Ref.[12] because the assumptions used there are no longer valid for light deformed nuclei. We rather use the $SU(3)$ model of the nucleus and deduced from there a deformation of 0.175 (see Ref. [3]). It will be seen that the results do not sensitively depend on that. Interpreting the 2_1^+ state at 3.368 MeV in ^{10}Be as rotational, it is absorbed into the butterfly motion. Instead the first vibrational state is a γ-mode at 5.958 MeV, indicating a very stiff system. The fact that the 3.368 MeV transition (minus the 6 keV shift) is seen in experiment speaks in favour of a vibrational ^{10}Be nucleus. We carried out computations in frame of the Hartree-Fock method, with pairing correlations taken into account and using Skyrme III forces. The result was that the deformation energy curve of ^{10}Be has a spherical minimium and it is symmetric for small deformations. Nevertheless, we would like to see the effects of a possibly deformed light cluster. All this has to be taken into account when it comes to the interpretation of the theoretical results.

After having described the three systems mentioned above, the nuclear structure of the participants of the system ^{90}Y + ^{10}Be + ^{142}Cs will be discussed shortly. ^{90}Y and ^{142}Cs are odd-even nuclei with an odd number of protons. Their treatment requires the inclusion of the spins of the extra protons.

^{96}Sr + ^{10}Be + ^{146}Be

The nuclear structure of this system was already discussed in Refs. [3, 4]. The new contribution here is the inclusion of the β and γ and the relative vibrational modes.

The Sr and the Ba nuclei are prolately deformed and the corresponding deformation values are given in Table 4.3. The parameters of the Hamiltonian are deduced and listed in Table 4.1. The spectrum can then be determined from the Eq. (4.142). The radii along the

system	^{96}Sr + ^{10}Be +^{146}Ba	^{112}Ru +^{10}Be + ^{130}Sn	^{108}Mo + ^{10}Be + ^{134}Te
β_{01}	0.338	0.237	0.333
β_{02}	0.199	0.	0.
β_{03}	0.175	0.175	0.175
$\frac{\hbar^2}{2\Theta_{11}}$	8.62 10^{-4}	9.10 10^{-4}	9.20 10^{-4}
$\hbar\omega_r$	4.270	3.911	3.850
$\hbar\omega_\xi$	19.417	19.035	18.511
$\hbar\omega_\varepsilon$	2.427	2.880	2.681
$\hbar\omega_{\beta_1}$	1.229	-	-
$\hbar\omega_{\beta_2}$	1.053	1.220	1.280
$\hbar\omega_{\beta_3}$	6.179	6.179	6.179
$\hbar\omega_{\eta_1}$	1.507	0.524	0.586
$\hbar\omega_{\eta_2}$	1.566	-	-
$\hbar\omega_{\eta_3}$	5.958	5.958	5.958
R_1	6.66	6.88	6.92
R_2	7.11	6.08	6.14
R_3	3.11	3.11	3.11

Table 4.3: Parameters of the three systems. In case the *deformation* β_k, for one particular nucleus k, is *zero* the $\hbar\omega_{\beta_k}$ has to be interpreted as the energy $\hbar\omega_k$ of the five dimensional harmonic oscillator. For the case of an oscillator the corresponding $\hbar\omega_{\eta_k}$ is put to zero because it is not relevant. The deformation parameters have no units. The one of the $\hbar\omega_k$ are in MeV. The units of the radii are is in fm, where we used for the spherical equivalent radius the formula $r_0 A^{1/3}$ with $r_0 = 1.2$ for a heavy and $r_0 = 1.3$ for the light cluster. An "-" indicates that either no information is available, very insecure or β_{0k} is zero.

prolate symmetry axis are also listed in Table 4.3. For the Be nucleus it was shown in Ref. [3] that according to the $SU(3)$ model it can be taken as triaxial and the deformation is 0.175. This consideration does exclude any mixing with other $SU(3)$ representations due to the $SU(3)$ mixing terms like pairing and spin-orbit interaction. There are many indications that the ^{10}Be nucleus can be assumed to be spherical [3]. However, in order to see the influence of a deformed light cluster we will assume a deformed ^{10}Be nucleus.

Using the parameters of this system listed in Table 4.3, the spectrum of the molecular states is given by (units are in MeV)

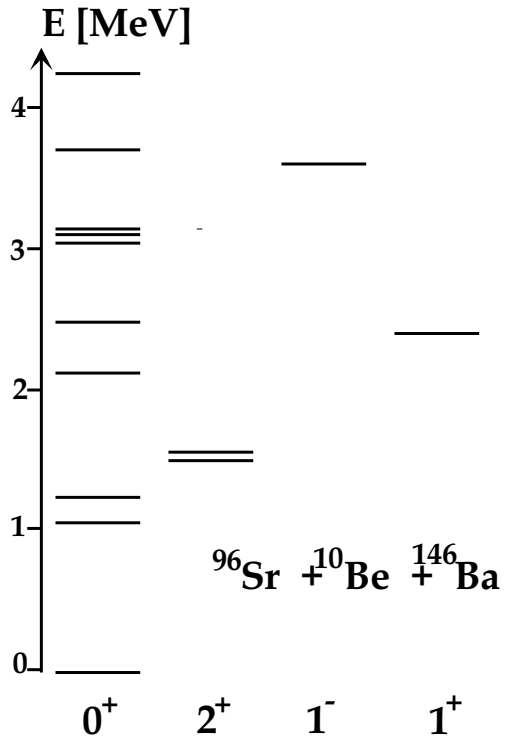

Figure 4.7: Spectrum of the system ^{96}Sr + ^{10}Be + ^{146}Ba. For detailed explanations, see the text. Only band heads are shown. On top of each band head there is a rotational band with the characteristics explained in the text. The butterfly mode is the 1^+ state to the right and the first relative vibration is given by the 1^- state. The 2^+ states belong to the γ vibrational states with either K_1 or K_2 equal to 2. The 0^+ band heads consist of the ground state, β and γ vibrational (with $n_{\eta_i} = 1$) band heads of the heavy clusters.

$$
\begin{aligned}
E \;=\; & 0.000862[I(I+1) - K^2] + 2.406(\mid K - K_1 - K_2 - K_3 \mid + 2n_\varepsilon) \\
& +1.229 n_{\beta_1} + 1.053 n_{\beta_2} + 6.179 n_{\beta_3} \\
& +1.507(\frac{1}{2} \mid K_1 \mid + 2n_{\eta_1}) + 1.566(\frac{1}{2} \mid K_2 \mid + 2n_{\eta_2}) + 5.958(\frac{1}{2} \mid K_3 \mid + 2n_{\eta_3}) \\
& +3.61 n_r + 17.59 n_\xi \quad , \qquad (4.159)
\end{aligned}
$$

where we have skipped the zero point energy contribution, i.e. E gives the difference in energy to the ground state. All deformation vibrational states are lying above 1 MeV. The same holds for the butterfly frequency. In conclusion, below 1 MeV only the rotational states belonging to the ground state band appear. The relative ξ motion is at such a large energy that it does not play any practical role. Interesting to note is that in the calculation of $\frac{2\Theta_{11}}{\hbar^2}$ the dominant contribution comes from the last term of Θ_{11} as given in Eq. (4.124). The other terms contribute at most three percent. Even the rotational contribution from the light Be cluster, given by the term before the last one of Θ_{11}, can be neglected due to a small ξ_0. This is similar to the two cluster case where the corresponding term is dominating all others.

In Fig.4.7 the expected structure of the spectrum is plotted. Only band heads contained in

Eq.(4.159) are shown and not those belonging to other degrees of freedom, like the rotational octupole band head state 1^- in ^{146}Ba and the extra 0^+ state at 1.465 MeV in ^{96}Sr. As can be seen in Fig. 4.7 , there are no excited band head states below 1 MeV suggesting a stable behavior against the butterfly motion. Nearly all states below 1 MeV are rotational one belonging to the ground state band. Note that in Ref. [3] The $\hbar\omega_\varepsilon$ was estimated assuming a spherical ^{10}Be nucleus. 4.3).

The energy values of the first 2_1^+ and 4_1^+ states are 5.2 keV and 17 keV respectively. The deformation of ^{10}Be results in an increase of the separation of the heavy clusters which raises the moment of inertia. The expected lowering in the energies of the rotational states is small compared to the results of Ref. [3], which are 6 keV and 20 keV for the 2_1^+ and 4_1^+ states, indicating a small influence of the supposed deformation of the ^{10}Be nucleus.

^{112}*Ru* + ^{10}*Be* + ^{130}*Sn*

This system is the most symmetric one we could get for which experimental information about the structure of the individual clusters are available and not just the ground state only. This is important for deducing the deformation of the nuclei. This system has not been seen yet but should exist.

The heavy fragments are again even-even nuclei. Using the tables of Ref. [12] the corrected quadrupole deformation of ^{112}Ru is given by 0.237 corresponding to a large β_2. For ^{130}Sn no information in these tables are available. However, The Sn isotopes are known to be an excellent example for the seniority scheme [14]. The proton shell is closed and the neutron shell is open. Because the seniority scheme is realized a zero deformation can be assumed. This is also confirmed by the $(E(4_1^+)/E(2_1^+))$ ratio [15] which is 1.63 for ^{130}Sn. For ^{112}Ru the ratio is 2.72 indicating a rotational structure.

The parameters of the nuclei and the system are listed in Table 4.3, including the radii of the clusters along the line of contacts. The excitation energy is given by (units are in MeV)

$$\begin{aligned} E &= 0.00094[I(I+1)-K^2] + 2.215(\mid K-K_1-K_2-K_3 \mid +2n_\varepsilon) \\ &\quad +0.524(\frac{1}{2} \mid K_1 \mid +2n_{\eta_1}) + 5.958(\frac{1}{2} \mid K_3 \mid +2n_{\eta_3}) \\ &\quad +3.911n_r + 19.035n_\xi \\ &\quad +1.220N_{\text{Sn}} \quad , \end{aligned} \tag{4.160}$$

where the last term describes the five-dimensional harmonic oscillator for the Sn nucleus. The spectrum is presented in Fig. 4.8, with the same characteristics as in ^{96}Sr + ^{10}Be + ^{146}Ba. Not plotted are the band heads belonging to different degrees of freedom than those described by the model. Also suppressed are the states of the five dimensional harmonic oscillator of ^{130}Sn with $\hbar\omega_\beta$ = 1.220 MeV. There is a γ vibrational state at approximately half a MeV though. One should observe above this band head a rotational structure similar to the ground state band with the difference that also a 3^+ state exists at about 5.5 keV and a 4^+ state at 12.7 keV above the 2^+ band head state. The ξ relative vibrational term can also be neglected. In this system the rotational part is dominated to almost 100% by the last term of Θ_{11} of Eq. (4.124). The influence of the assumed deformation of ^{10}Be is again small.

^{108}*Mo* + ^{10}*Be* + ^{134}*Te*

The heavy fragments are even-even nuclei. Using the tables of Ref. [12] the deformation

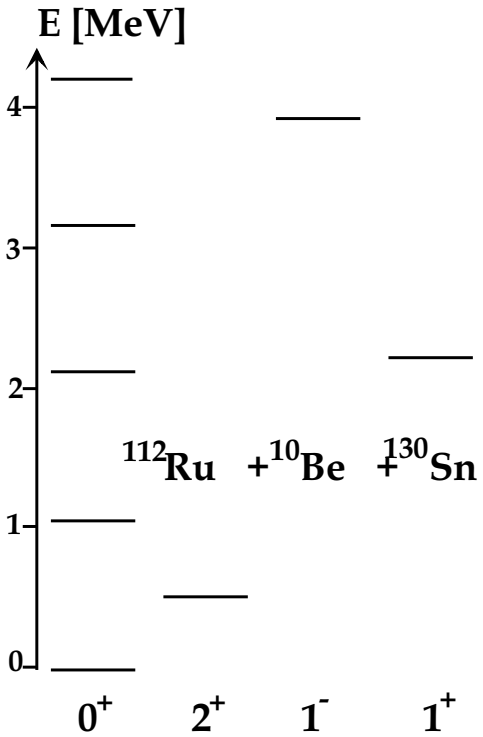

Figure 4.8: Spectrum of the system ^{112}Ru + ^{10}Be + ^{130}Sn. On top of each band head there is a rotational band with the characteristics explained in the text. The butterfly mode is the 1^+ state to the right and the first relative vibration is given by the 1^- state. The 2^+ state belongs to the γ vibrational state of the first cluster with $K_1 = 2$. The 0^+ band heads consist of the ground state and the γ vibrational (with $n_{\eta_i} = 1, ..., 4$) band heads of ^{112}Ru. No β vibrational states could be identified in the three clusters at energies below 5 MeV.

of ^{108}Mo is given by 0.354 which we corrected to 0.333 using the additional deformation dependent terms in the $B(E2; 0_1^+ \rightarrow 2_1^+)$ as given in Ref. [1]. The experimental information of the spectrum is taken from Ref. [13]. For ^{134}Te no information in these tables are available, however, the tendency observed coming from the lighter isotopes indicates a small β. When we look at the ratio $(E(4_1^+)/E(2_1^+))$ we obtain 2.92 and 1.23 for ^{108}Mo and ^{134}Te respectively. This experimental observation supports a deformed nucleus for ^{108}Mo and possibly a spherical deformation for ^{134}Te.

The ^{108}Mo nucleus is particularly difficult to treat. Within the geometrical model [1] the Potential-Energy-Surface (PES) of the neigbouring nucleus ^{108}Ru has a spherical absolute minimum and a triaxial local minimum at large deformation [16]. The energy of the ground state, however, lies above the saddle point and the ground state is a strong mixture between both deformations. A large β in average is also indicated by the results of $\beta\beta$ decay for ^{108}Mo where with great success a model for strongly deformed nuclei was applied [17].

Under the assumption that the Mo isotope is deformed while the Te nucleus is a vibrator

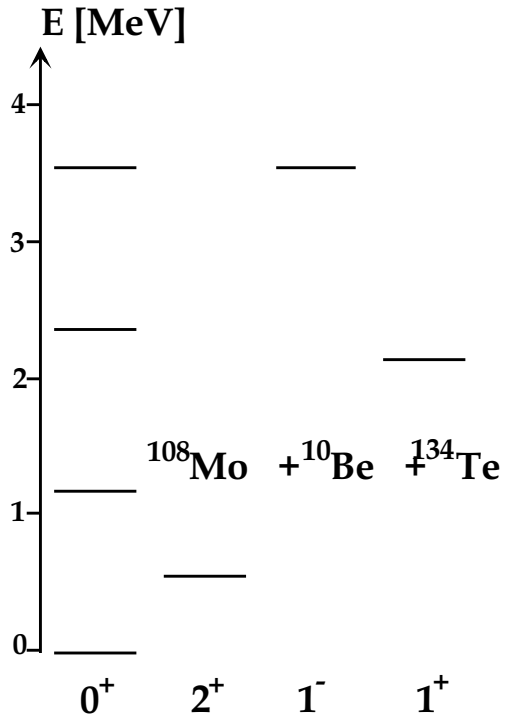

Figure 4.9: Spectrum of the system ^{108}Mo + ^{10}Be + ^{134}Te. On top of each band head there is a rotational band with the characteristics explained in the text. The butterfly mode is the 1^+ state to the right and the first relative vibration is given by the 1^- state. The 2^+ state belongs to the γ vibrational state of the first cluster with $K_1 = 2$. The 0^+ band heads consist of the ground state and the γ vibrational (with $n_{\eta_i} = 1, ..., 3$) band heads of ^{108}Mo. No β vibrational states could be identified in the three clusters at energies below 5 MeV.

and using the parameters listed in Table 4.3, the excitation energy is given by

$$\begin{aligned} E &= 0.00092[I(I+1) - K^2] + 2.127(\mid K - K_1 - K_2 - K_3 \mid + 2n_\varepsilon) \\ &\quad +0.586(\frac{1}{2} \mid K_1 \mid + 2n_{\eta_1}) + 5.958(\frac{1}{2} \mid K_3 \mid + 2n_{\eta_3}) \\ &\quad +3.54n_r + 17.30n_\xi \\ &\quad +1.280N_{Te} \quad , \end{aligned} \tag{4.161}$$

where the last term describes the five-dimensional harmonic oscillator for the Te nucleus. The spectrum is presented in Fig.4.9, again with the same characteristics as in ^{96}Sr + ^{10}Be + ^{146}Ba. Not plotted are the band heads belonging to degrees of freedom different from those of the model. Also excluded are the states of the five dimensional harmonic oscillator of ^{134}Te with $\hbar\omega_{\text{Te}} = 1.280$ MeV. There is a γ vibrational state at approximately half a MeV though. One should observe above this band head a rotational structure similar to the ground state band with the difference that also a 3^+ state exists at about 5.5 keV and a 4^+ state at 12.8 keV above the 2^+ band head state. The ξ relative vibrational term can also be neglected. In this system the rotational part is dominated to almost 100% by the last term of Θ_{11} of Eq. (4.124). Again the interpretation with respect to the influence of the ^{10}Be deformations similar as in the two former cases.

Structure of the participants in $^{90}Y + {}^{10}Be + {}^{142}Cs$

This is the third system possibly identified in the experiment of Ref. [11]. The heavy clusters are odd-odd nuclei. The influence of the extra odd protons should be included in our consideration. The way to do it for individual nuclei is given in Ref. [1]. Such nuclei can be described by an even-even core and an odd proton around it. The deformation can be deduced via the one of neighboring nuclei. Take $^{90}_{38}\mathrm{Sr}_{52}$, $^{90}_{40}\mathrm{Zr}_{50}$ as neighboring nuclei and also $^{88}_{38}\mathrm{Sr}_{50}$ as the core. The deformation of these nuclei are listed in Ref. [12], except for $^{90}_{38}\mathrm{Sr}_{52}$. The deformation are 0.09 and 0.12 for the last two nuclei respectively. However, one must take some care in using these tables. The formula used to deduce the deformation from the experimental $B(E2, 0_1^+ \rightarrow 2_1^+)$ transition is theoretically biased, i.e. a strongly axial symmetric deformation is assumed. In this case the deformation value is proportional to the square root of the transition. For spherical and triaxial nuclei the formula is incorrect and in general the deformations deduced are too high due to the fact that higher orders [1] in deformation are also neglected. Nevertheless, the tables in Ref. [12] give a good idea about the trends. Another possibility is to use the tables of M. Sakai [15] where the ratio $(E(4_1^+)/E(2_1^+))$ is investigated. For rotational nuclei the ratio is 3.33 and for vibrational nuclei it is 2.0. Most nuclei lie in between these two values. For the three nuclei mentioned above this ratio is 1.99, 1.41 and 3.15 respectively. Except for the last value, which is near to a rotator, the data indicate a spherical deformation, in agreement with the data listed in Ref. [12].

For $^{142}_{55}\mathrm{Cs}_{87}$ as neighboring nuclei we took $^{142}_{54}\mathrm{Xe}_{88}$, $^{142}_{56}\mathrm{Ba}_{86}$ and for the core $^{140}_{54}\mathrm{Xe}_{86}$. The deformation values listed in Ref. [12] are 0.157 and 0.114 for the last two systems. No information is listed for the first nucleus. Using the tables of Sakai [15], except for the first nucleus were we used the ISOTOPE EXPLORER [13], the ratio of $(E(4_1^+)/E(2_1^+))$ are respectively 2.41, 2.32 and 2.22. They hint to a spherical nucleus while the β values are in between. In Ref. [11] a deformed nucleus was assumed, which are not confirmed by data.

4.2.2 Trinuclear Quasimolecules with Spherical Clusters

In the subsection 4.2 we treated the ^{10}Be-like Giant Trinuclear Molecule (GTM) with all three clusters deformed and sitting in the equilibrium configuration with their symmetry axes aligned. The light cluster was sandwiched in-between the two heavier fragments. A large variety of collective modes are showing-up such as the butterfly, belly-dancer which are molecular vibrations, rotations of the whole system and also β and γ vibrations of each clusters. Only a few vibrational states are showing-up below 1 MeV, the rotational states of the ground state band being strongly squeezed. The first 2^+ state is at approximately 5-6 keV and the 4^+ state at approximately 17-19 keV, whereas the butterfly motion lies at energies around 2.5 MeV.

Fundamentally in our treatment is the existence of a minimum in the total heavy-ion potential which arises as a consequence of the interplay between the repulsive-attractive nuclear forces and the purely repulsive Coulomb interaction. When this is valid for the case of ^{10}Be-accompanied ternary fission then quasi-molecular configurations could develope also in the α-accompanied ternary cold fission. For the time being there is no evidence of such a molecule because of the ”inert” nature of the LCP, whose first excited lays at 20 MeV. In the present paper we study the molecular spectrum of an α-like GTM whose heavy partners

are also spherical clusters with the aim to understand the differences between the triangular (oblate) configuration, which is the best candidate of a GTM in our view, and the linear (prolate) one.

The Three Spherical Clusters Hamiltonian

The Hamiltonian for three clusters with masses m_1, m_2 and m_3 interacting by means of two-body forces has the form

$$H = \frac{\boldsymbol{p}_1}{2m_1} + \frac{\boldsymbol{p}_2}{2m_2} + \frac{\boldsymbol{p}_3}{2m_3} + V_{12} + V_{23} + V_{13} \tag{4.162}$$

In [3, 4, 18] this problem was handled by separating out the centre-of-mass motion and introducing Jacobi coordinates. The cartesian space coordinates being denoted by $\boldsymbol{r}_1, \boldsymbol{r}_2$ and $\boldsymbol{r}_3$, the Jacobi coordinates, for which the two heavier clusters 1 and 2 appear explicitely as a subsystem, are introduced by means of the following transformations:

$$\begin{aligned} \boldsymbol{\rho} &= \boldsymbol{r}_2 - \boldsymbol{r}_1 \\ \boldsymbol{\lambda} &= \frac{m_1\boldsymbol{r}_1 + m_2\boldsymbol{r}_2}{m_1 + m_2} - \boldsymbol{r}_3 \\ \boldsymbol{R}_{\text{c.m.}} &= \frac{m_1\boldsymbol{r}_1 + m_2\boldsymbol{r}_2 + m_3\boldsymbol{r}_3}{m_1 + m_2 + m_3} \end{aligned} \tag{4.163}$$

The corresponding transformation for momentum coordinates becomes:

$$\begin{aligned} \boldsymbol{p}_\rho &= \frac{m_1\boldsymbol{p}_2 - m_2\boldsymbol{p}_1}{m_1 + m_2} \\ \boldsymbol{p}_\lambda &= \frac{m_3(\boldsymbol{p}_1 + \boldsymbol{p}_2) - (m_1 + m_2)\boldsymbol{p}_3}{m_1 + m_2 + m_3} \end{aligned} \tag{4.164}$$

$$\boldsymbol{P}_{\text{c.m.}} = \boldsymbol{p}_1 + \boldsymbol{p}_2 + \boldsymbol{p}_3 \tag{4.165}$$

The momentum $\boldsymbol{p}_\rho$, which is canonically conjugate to $\boldsymbol{\rho}$, is the relative momentum of the particles 1 and 2, and $\boldsymbol{p}_\lambda$, which is canonically conjugate to $\boldsymbol{\lambda}$, is the relative momentum of cluster 3 relative to a mass $(m_1 + m_2)$ at the center of mass of cluster 1 and 2.

In Jacobi coordinates, the Hamiltonian has the form

$$H = \frac{1}{2(m_1 + m_2 + m_3)}\boldsymbol{P}^2 + \frac{1}{2\mu_{12}}\boldsymbol{p}_\rho^2 + \frac{1}{2\mu_{(12)3}}\boldsymbol{p}_\lambda^2 + \sum_{i\neq j=1}^{3} V_{ij} \tag{4.166}$$

with the reduced masses

$$\mu_{12} = \frac{m_1 m_2}{(m_1 + m_2)}, \qquad \mu_{12} = \frac{m_3(m_1 + m_2)}{m_1 + m_2 + m_3}$$

Next we define the components of the vectors $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ according to the geometry adopted in Fig.4.10.

$$\boldsymbol{\rho} = (\rho \sin a\gamma, 0, \rho \cos a\gamma), \qquad \boldsymbol{\lambda} = (-\lambda \sin(1-a)\gamma, 0, \lambda \cos(1-a)\gamma) \tag{4.167}$$

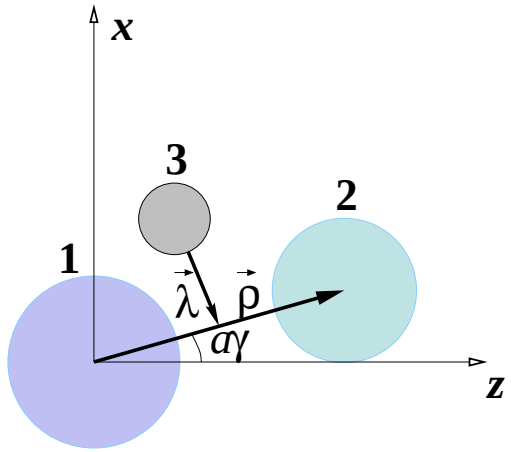

Figure 4.10: The three-body problem : the vector $\boldsymbol{\rho}$ joining the heavier clusters 1 and 2 makes an angle $a\gamma$ with the z-axis.

This corresponds to define the intermediate z-axis in between the two vectors (see Fig. 4.10). For a=0 the vector $\boldsymbol{\rho}$ lies along the z-axis. For reasons that will become transparent below we are not choosing $\boldsymbol{\rho}$ along the z-axis.

The kinetic energy

The components of $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ in the laboratory system are defined in eq.(4.167). We rotate these vectors in the x-z plane of the molecular system (see Fig.4.10)

$$\bar{\varrho}_i = \sum_k D^1_{ki}(\theta)\varrho_k \qquad \bar{\lambda}_i = \sum_k D^1_{ki}(\theta)\lambda_k. \tag{4.168}$$

For the time-derivative of the rotation matrix acting on the cartesian components of $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ we use:

$$\dot{D}_{ij} = \sum_k \Omega_{ik} D_{kj} \quad . \tag{4.169}$$

with $\Omega_{ij} = -\Omega_{ji}$ as an antisymmetric matrix whose components are the angular velocity in the plane (ij) :

$$\Omega = \begin{pmatrix} 0 & \omega'_3 & -\omega'_2 \\ -\omega'_3 & 0 & \omega'_1 \\ \omega'_2 & -\omega'_1 & 0 \end{pmatrix}$$

After substituting in (4.168) the ansatz (4.167) we get the new forms of $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ which are to be used in the classical expression of the kinetic energy (4.172)

$$\begin{aligned} \dot{\boldsymbol{\rho}}^2 &= \dot{\rho}^2 + a^2\rho^2\dot{\gamma}^2 + \rho^2\left[\cos^2(a\gamma)\omega_1'^2 + \omega_2'^2 + \sin^2(a\gamma)\omega_3'^2\right] \\ &- \rho^2\sin(2a\gamma)\omega'_1\omega'_3 + 2a\rho^2\omega'_2\dot{\gamma} \end{aligned} \tag{4.170}$$

$$\begin{aligned} \dot{\boldsymbol{\lambda}}^2 &= \dot{\lambda}^2 + (1-a)^2\lambda^2\dot{\gamma}^2 + \lambda^2\left[\cos^2((1-a)\gamma)\omega_1'^2 + \omega_2'^2 + \sin^2((1-a)\gamma)\omega_3'^2\right] \\ &+ \lambda^2\sin(2(1-a)\gamma)\omega'_1\omega'_3 - 2a\lambda^2\omega'_2\dot{\gamma} \end{aligned} \tag{4.171}$$

After inserting the ansatz (4.167) for $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ we obtain the following expression for the classical kinetic energy (with C.M. part excluded)

$$\begin{aligned}
T_{\rm cl} &= \frac{1}{2}\mu_{12}\dot{\boldsymbol{\rho}}^2 + \frac{1}{2}\mu_{(12)3}\dot{\boldsymbol{\lambda}}^2 \\
&= \frac{1}{2}\mu_{12}\dot{\rho}^2 + \frac{1}{2}\mu_{(12)3}\dot{\lambda}^2 + \frac{1}{2}\left[\mu_{12}a^2\rho^2 + \mu_{(12)3}(1-a)^2\lambda^2\right]\dot{\gamma}^2 \\
&+ \frac{1}{2}\left\{\mu_{12}\rho^2\cos^2(a\gamma) + \mu_{(12)3}\lambda^2\cos^2[(1-a)\gamma]\right\}\omega_1'^2 \\
&+ \frac{1}{2}\left(\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2\right)\omega_2'^2 \\
&+ \frac{1}{2}\left\{\mu_{12}\rho^2\sin^2(a\gamma) + \mu_{(12)3}\lambda^2\sin^2[(1-a)\gamma]\right\}\omega_3'^2 \\
&- \frac{1}{2}\left\{\mu_{12}\rho^2\sin(2a\gamma) - \mu_{(12)3}\lambda^2\sin[2(1-a)\gamma]\right\}\omega_1'\omega_3' \\
&+ \left[\mu_{12}a\rho^2 - \mu_{(12)3}(1-a)\lambda^2\right]\omega_2'\dot{\gamma} \quad . \qquad (4.172)
\end{aligned}$$

where $\boldsymbol{\omega}' \equiv (\omega_1', \omega_2', \omega_3')$ is the angular velocity of the molecular frame.

The above expression looks at first glance rather involved, but by a convenient choice of the parameter a, depending also on which configuration we are interested in, the unwanted non-diagonal terms can be removed.

The potential

The interaction between the nuclei composing the quasi-molecule is taken as in previously to result from a heavy-ion double folding integral.

The total ternary potential which occurs in the Hamiltonian (4.162) is plotted in Fig.4.11 for two different inter-fragment distances between the heavier clusters, namely ^{132}Sn and ^{116}Pd. As can be noticed on this figure the potential displays a quasi-molecular pattern with two minima in the equatorial region and two at the poles of the system. Due to the axial symmetry, the minima in the equatorial region are equivalent, and actually one may speak about a ring which represents the geometrical locus of the points where the three-body potential attains its absolute minimum. This is the case for $R = 11$ fm, i.e. when the two heavier clusters are in the touching configuration. When the distance R is increased up to $R = 15.35$ fm the above mentioned ring shrinks to a point on the symmetry axis. In order to better understand this circumstance we displayed in Fig.4.12 the minimum value of the total ternary potential for different values of R. For R=11 fm, the system has a triangular configuration and a stable minimum is obtained whereas for R=15.35 fm, the clusters are aligned and the minimum is unstable. As a matter of fact, multidimensional tunneling calculations reported in [19] showed that the dynamical trajectory of the α particle will always be repelled from the symmetry axis and the system preserves its triangular geometry during the three-body break-up. It is also worthwhile to mention that preformation calculations carried out recently [20] are leading to the conclusion that when the two heavier fragments are at scission (touching point) the α-preformation amplitudes are showing-up a pronounced maximum off the fission axis, that is the third lighter cluster is preformed from the mother nucleus off the symmetry axis.

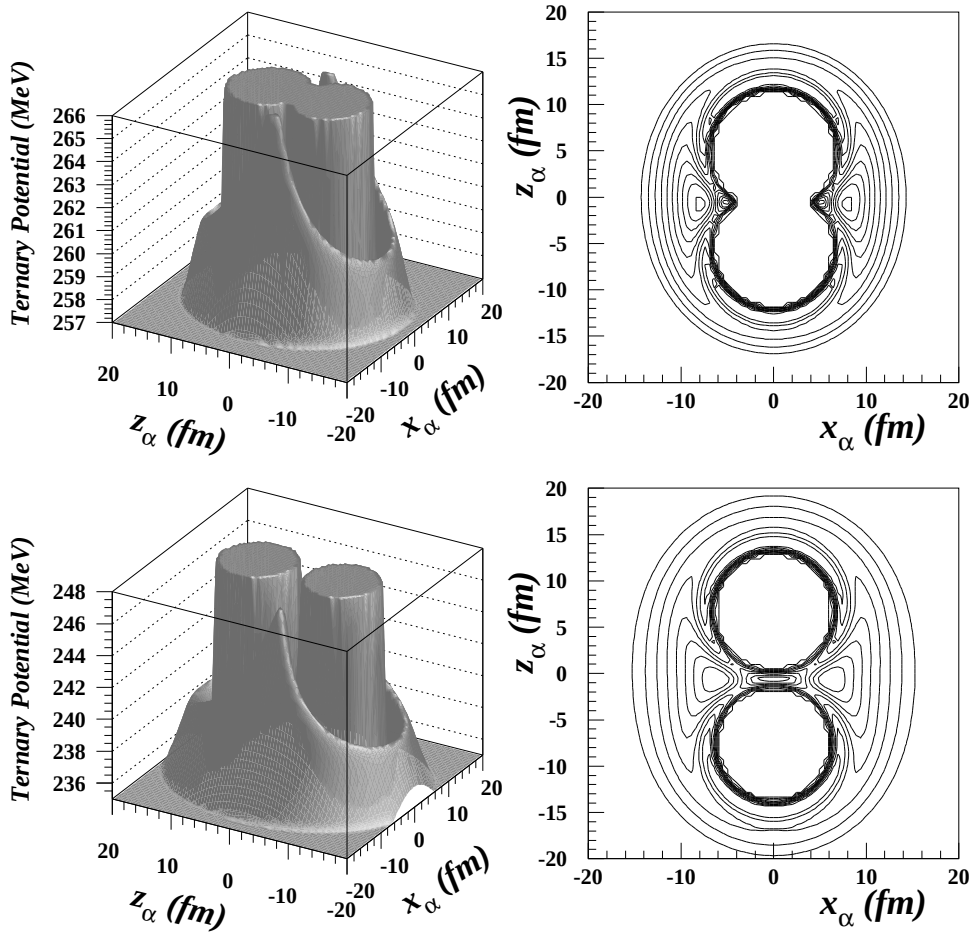

Figure 4.11: The total ternary potential in a three-dimensional plot (left panel) and a contour plot (right panel) for the GTM ^{132}Sn $+ \alpha +^{116}$Pd. The upper pannels are corresponding to an interfragment distance R=11 fm whereas the lower to 15.35 fm

Next, the potential is expanded up to quadratic terms around the molecular minima :

$$V_{\rm cl} = \frac{1}{2}\sum_{i>j=1}^{3} C_{ij}(\boldsymbol{r}_j - \boldsymbol{r}_i)^2 = \frac{1}{2}C_\rho \delta\boldsymbol{\rho}^2 + \frac{1}{2}C_\lambda \delta\boldsymbol{\lambda}^2 + C_{\rho\lambda}\delta\boldsymbol{\rho}\delta\boldsymbol{\lambda} \tag{4.173}$$

where the relation between the two sets of stiffness parameters is given by

$$\begin{aligned} C_\rho &= C_{21} + \frac{C_{32}m_1^2 + C_{31}m_2^2}{(m_1+m_2)^2} \\ C_\lambda &= C_{32} + C_{31} \\ C_{\rho\lambda} &= \frac{C_{32}m_1 - C_{31}m_2}{m_1+m_2} \end{aligned} \tag{4.174}$$

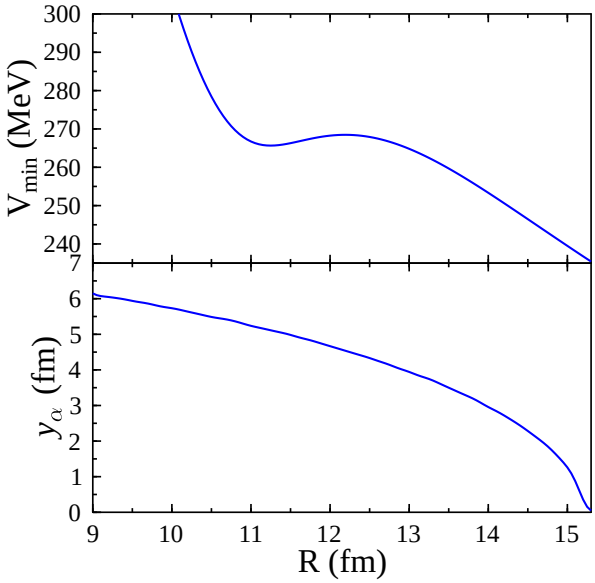

Figure 4.12: The minimum value of the total ternary potential (upper panel) and the position on the y-axis of this minimum (lower panel) as a function of the inter-fragment distance R.

In eq.(4.173) the value of the potential energy at the minimum configuration was omitted since it will contribute as a constant term.

Substituting (4.167) in (4.173), and neglecting the terms proportional to a and a^2, which are small as one shall see later, we get the following expression for the potential energy

$$\begin{aligned} V_{\rm cl} &\approx \frac{1}{2}C_\rho\delta\rho^2 + \frac{1}{2}C_\lambda\delta\lambda^2 + \frac{1}{2}C_\lambda\lambda^2\delta\gamma^2 \\ &+ C_{\rho\lambda}(\delta\rho\delta\lambda\cos\gamma - \lambda\delta\rho\delta\gamma\sin\gamma) \quad , \end{aligned} \tag{4.175}$$

The Collective Spectrum

The Triangular Configuration

The kinetic energy (4.172) contains two types of couplings : a Coriolis coupling and a rotation-vibration interaction. In the case of a triangular configuration it is worthwhile to remove the last non-diagonal contribution by choosing

$$a = \frac{\mu_{(12)3}\lambda^2}{\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2} \quad . \tag{4.176}$$

With this eq.(4.172) is rewritten as

$$
\begin{aligned}
T_{\rm cl} &= \frac{1}{2}\mu_{12}\dot{\rho}^2 + \frac{1}{2}\mu_{(12)3}\dot{\lambda}^2 + \frac{1}{2}\frac{\mu_{12}\mu_{(12)3}\rho^2\lambda^2}{\mu_{12}\rho^2+\mu_{(12)3}\lambda^2}\dot{\gamma}^2 \\
&+ \frac{1}{2}\left\{\mu_{12}\rho^2\cos^2(a\gamma)+\mu_{(12)3}\lambda^2\cos^2[(1-a)\gamma]\right\}\omega_1'^2 \\
&+ \frac{1}{2}\left\{\mu_{12}\rho^2+\mu_{(12)3}\lambda^2\right\}\omega_2'^2 \\
&+ \frac{1}{2}\left\{\mu_{12}\rho^2\sin^2(a\gamma)+\mu_{(12)3}\lambda^2\sin^2[(1-a)\gamma]\right\}\omega_3'^2 \\
&- \frac{1}{2}\left\{\mu_{12}\rho^2\sin(2a\gamma)-\mu_{(12)3}\lambda^2\sin[2(1-a)\gamma]\right\}\omega_1'\omega_3' \qquad (4.177)
\end{aligned}
$$

We next specialize our considerations to the case of the GTM ^{132}Sn + α + ^{116}Pd which presumably occurs in the α-accompanied ternary cold fission of ^{252}Cf. Picking-up for ρ and λ their values at the minimum configuration, we obtain a $\approx$1/300. Therefore a $\ll$1 and several terms of the above expression can be safely left out, as have been done above with some potential terms. Moreover we can approximate that γ consists of small deviations from $\pi/2$, i.e. we perform the change of variable $\gamma = \pi/2 - \varepsilon$ (this is so because we are not far from the totally symmetric case with $A_1 = A_2$). Under this circumstance one get the new expression

$$
\begin{aligned}
T_{\rm cl} &\approx \frac{1}{2}\mu_{12}\dot{\rho}^2 + \frac{1}{2}\mu_{(12)3}\dot{\lambda}^2 + \frac{1}{2}\mu_{(12)3}\lambda^2\dot{\varepsilon}^2 \\
&+ \frac{1}{2}\mu_{12}\rho^2(\omega_1'^2+\omega_2'^2) + \frac{1}{2}\mu_{(12)3}\lambda^2\omega_3'^2 + \mu_{(12)3}\lambda^2\varepsilon\omega_1'\omega_3' \qquad (4.178)
\end{aligned}
$$

Applying the Pauli-Podolsky quantization method [1] we obtain

$$
\begin{aligned}
\widehat{T} &= -\frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\rho^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} - \frac{\hbar^2}{2\mu_{(12)3}\lambda^2}\frac{\partial^2}{\partial\varepsilon^2} \\
&+ \frac{\hbar^2}{2\mu_{12}\rho^2}(\boldsymbol{L}^2 - L_3'^2) + \frac{\hbar^2}{2\mu_{(12)3}\lambda^2}L_3'^2 - \frac{\hbar^2\varepsilon}{\mu_{12}\rho^2}L_1'L_3' \qquad (4.179)
\end{aligned}
$$

As one can see this form contains couplings between the different vibrational (specified by the observables ρ, λ and ε) and rotational (specified by the angular momentum) modes in a non-trivial way. At this stage we assume that near the minimum's position the displacements $\delta\rho, \delta\lambda$ and ε are not large with respect to the equilibrium values $y_0 = \rho_0, \lambda_0$ and $\varepsilon_0 = 0$, i.e. $y = y_0 + \delta y$, with $\delta y \ll 1$. Under this assumption one can expand in Taylor series all coordinate functions of the kinetic energy and potential energy operators. Formally one can write :

$$
\widehat{H} = \widehat{H}^{(0)} + \sum_i \delta y_i \widehat{H}_i^{(1)} + \frac{1}{2}\sum_{ij}\delta y_i\delta y_j\widehat{H}_{ij}^{(2)} + \ldots \qquad (4.180)
$$

For the moment we restrict ourselves to the zeroth-order approximation and we get

$$\begin{aligned}\widehat{H}^{(0)} &= -\frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\rho^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} - \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^2}\frac{\partial^2}{\partial\varepsilon^2} \\ &+ \frac{\hbar^2}{2\mu_{12}\rho^2}\boldsymbol{L}^2 + \frac{\hbar^2}{2}\left(\frac{1}{\mu_{(12)3}\lambda_0^2} - \frac{1}{\mu_{12}\rho_0^2}\right)L_3'^2 \\ &+ \frac{1}{2}C_\rho\delta\rho^2 + \frac{1}{2}C_\lambda\delta\lambda^2 + \frac{1}{2}C_\lambda\lambda_0^2\delta\varepsilon^2 - C_{\rho\lambda}\lambda_0\delta\rho\varepsilon \end{aligned} \tag{4.181}$$

Note that in the zeroth-order approximation the kinetic non-diagonal term $\propto L_1'L_3'$ and the potential non-diagonal term $\propto \delta\rho\delta\lambda$ are not present.

Due to the degeneracy of the problem the eigenfunctions of the Hamiltonian must be constructed as linear combinations of the rotation matrices D^I_{MK}. Since $\boldsymbol{L}^2$ and L_z' are commuting with $\widehat{H}_0$ only combinations of different K-values for the same I and M are occuring

$$\Psi = \sum_{K=-I}^{+I} F_K^I(\rho,\lambda,\gamma)D^I_{MK}(\theta) \tag{4.182}$$

and the Schrödinger equation corresponding to the eigenvalue E reads:

$$\left\{\frac{1}{\mu_{12}}\frac{\partial^2}{\partial\rho^2} + \frac{1}{\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} + \frac{\hbar^2}{\mu_{(12)3}\lambda_0^2}\frac{\partial^2}{\partial\varepsilon^2}\right. \tag{4.183}$$

$$\begin{aligned} &- C_\rho(\rho-\rho_0)^2 - C_\lambda(\lambda-\lambda_0)^2 - C_\lambda\lambda_0^2\varepsilon^2 + 2C_{\rho\lambda}\lambda_0\varepsilon(\rho-\rho_0) \\ &\left.+\frac{1}{\mu_{12}\rho_0^2}I(I+1) + \left(\frac{1}{\mu_{(12)3}\lambda_0^2} - \frac{1}{\mu_{12}\rho_0^2}\right)K^2 + 2E\right\}F_K^I = 0 \end{aligned} \tag{4.184}$$

We therefore have an equation which describes three one-dimensional oscillators, two of them being coupled through a coordinate-coordinate term, i.e. there is a potential coupling between the modes ρ and ε. The decoupling of these two modes can be easily done by means of an unitary transformation [21]

$$\widehat{U} = \exp\left\{i\eta\left(\sqrt{\frac{\mu_{12}}{\mu_{(12)3}}}\frac{\rho}{\lambda_0}\frac{\partial}{\partial\varepsilon} - \sqrt{\frac{\mu_{(12)3}}{\mu_{12}}}\lambda_0\varepsilon\frac{\partial}{\partial\rho}\right)\right\} \tag{4.185}$$

where η is obtained from the following implicit equation

$$\tan 2\eta = -\frac{2C_{\rho\lambda}}{\sqrt{\mu_{12}\mu_{(12)3}}(\omega_\rho^2 - \omega_\varepsilon^2)} \tag{4.186}$$

such that the non-perturbed spectrum reads

$$\begin{aligned} E^{(0)}_{IKn_\rho n\varepsilon n_\lambda} &= \hbar\tilde{\omega}_\rho\left(n_\rho + \frac{1}{2}\right) + \hbar\tilde{\omega}_\varepsilon\left(n_\varepsilon + \frac{1}{2}\right) + \hbar\omega_\lambda\left(n_\lambda + \frac{1}{2}\right) \\ &+ \frac{\hbar^2}{2\mu_{12}\rho_0^2}\left[I(I+1) - K^2\right] + \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^2}K^2 \end{aligned} \tag{4.187}$$

the frequencies being defined as follows

$$\omega_\rho = \sqrt{\frac{C_\rho}{\mu_{12}}}, \qquad \omega_\lambda = \omega_\varepsilon = \sqrt{\frac{C_\lambda}{\mu_{(12)3}}} \tag{4.188}$$

$$\tilde{\omega}^2_{\rho(\varepsilon)} = \frac{1}{2}\left(\omega^2_\rho + \omega^2_\varepsilon \pm (\omega^2_\rho - \omega^2_\varepsilon)\sec 2\eta\right) \tag{4.189}$$

provided $\tilde{\omega}_\rho > \tilde{\omega}_\varepsilon$.

We have yet to consider the action of the parity transformation on the three cluster system. This will tell us the parity of a given state and the sequence of positive and negative parity states within rotational bands. To start with, note that all three clusters have different masses. We therefore can define the intrinsic system, the molecular plane in a definite manner. According to Fig. 4.10 cluster number 1 is left to cluster number 2, as seen with respect to the molecular z-axis. The third cluster is in the upper plane with a positive x value. This defines the equilibrium position around which small oscillations are considered. After the parity transformation is applied, the cluster 2 is to the left of cluster 1 and cluster 3 is in the lower half plane. In order to fulfill the same conditions of the molecular intrinsic system, as defined above, we have to apply an additional rotation in order to come back to the situation where cluster 2 is to the right of cluster 1 and cluster 3 is again in the upper plane. This rotation is given by

$$\hat{R}(x, y, z) = (-x, y, -z) \quad . \tag{4.190}$$

Note that the relative angles do not change, i.e. γ stays the same. This is easy to understand, because the triangular equilibrium position is well defined and the relative angles do not change. Therefore, the parity transformation implies only a redefinition of the rotational angles with respect to the laboratory. The effect of this rotation is obtained by applying $\hat{R}$ to the rotation matrices in Eq. (4.182). This rotation is a subsequent application of changing first (x, y, z) to $(x, -y, -z)$ and an additional one by 180^0 around the new z-axis. The action is described in Ref. [1] and the result is

$$\hat{R}D^I_{MK}(\theta) = (-1)^{I+K} D^I_{M-K}(\theta) \quad . \tag{4.191}$$

Therefore the rotational part of the solution in Eq. (4.182) has to be modified such that the states have a definite parity. This is achieved by

$$(D^I_{MK} \pm (-1)^{I+K} D^I_{M-K}) \quad , \tag{4.192}$$

where the positive parity states are given by the plus and the negative ones by the minus sign. Also K has to be positive in order to avoid double counting. The Eq. (4.192) immediately gives us a selection rule for the case $K = 0$. The sequence of states is

$$K = 0: \quad 0^+,\ 1^-,\ 2^+,\ 3^-,\ 4^+,\ ..., \tag{4.193}$$

i.e. for $K = 0$ bands positive and negative parity states are alternating. Like in the case of octupole deformed nuclei [1] the symmetry related to the reflexion $z \longrightarrow -z$ is broken in the GTM due to the fact that the clusters 1 and 2 are not identical although in the case considered

in this paper they have close masses. Consequently the condition that K is restricted to even integers is not valid in this case. For $K \neq 0$ there is no selection rule and for a given angular momentum there is a *parity doublet*:

$$K \neq 0: \quad K^{\pm},\ (K+1)^{\pm},\ (K+2)^{\pm},\ ..., \tag{4.194}$$

Including also the quadratic terms from the expansion (4.180) we must add to the zeroth-order Hamiltonian (4.181) the below listed perturbation.

$$\begin{aligned}
\delta\widehat{H}^{(2)} &= \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^3}\left(2-3\frac{\delta\lambda}{\lambda_0}\right)\delta\lambda\left(\frac{\partial^2}{\partial\varepsilon^2}-L_3'^2\right) \\
&- \frac{\hbar^2}{2\mu_{12}\rho_0^2}\left[\left(2-3\frac{\delta\rho}{\rho_0}\right)\frac{\delta\rho}{\rho_0}(\boldsymbol{L}^2-L_3'^2-1)-2\left(1-2\frac{\delta\rho}{\rho_0}\right)\varepsilon L_1'L_3'\right] \\
&+ \left[C_\lambda\lambda_0\delta\lambda-\frac{3\hbar^2}{4\mu_{12}\rho_0^2}\left(1+2\frac{\mu_{(12)3}\lambda_0^2}{\mu_{12}\rho_0^2}\right)\right]\varepsilon^2,
\end{aligned} \tag{4.195}$$

and second-order corrections to (4.187) can be computed according to the stationary perturbations theory.

In Fig.4.13 we represented the rotational states with energy smaller than 1 MeV for the g.s. band and the band with K= 1 and 2 of the GTM ^{132}Sn + α +^{116}Pd. The excited rotational state 1_1^- is at 5.4 keV, and the 2_1^+ state at 16.8 keV. The first state of the $K=1$ band is at 213 keV whereas the 2^+ state of the $K=2$ band is at 836 keV. The heads of the vibrational bands are laying at much higher energy, and were not plotted on this figure. For example the band head ($n_\rho=1, n_\lambda=n_\varepsilon=0$) is located at 3.85 MeV, a state which however could be reached in cold fission. We also compared on the same figure the results obtained in the nonperturbed case and taking into account the perturbation. It is easy to infer from here that

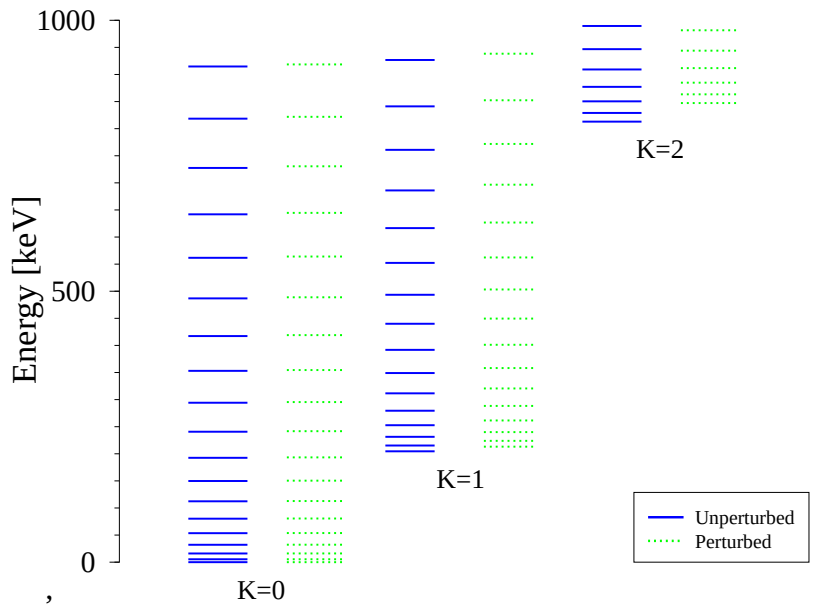

Figure 4.13: The first rotational bands $K=0,1,2$ in the zeroth-order approximation (full lines) and with the account of the first order perturbation (dashed lines).

taking only the zeroth-order Hamiltonian is a satisfactory approximation.

The Linear Configuration

In this case the center-of-masses of the three clusters are sitting on the same axis in the equilibrium position and the bending of this configuration is described by the angle $\gamma \equiv \varepsilon$. In order to cope with the non-diagonal terms we first equate to zero the factor multiplying $\omega_1'\omega_3'$ which provides the following equation for the parameter a

$$\tan 2a\gamma = \frac{\mu_{(12)3}\lambda^2 \sin 2\gamma}{\mu_{12}\rho^2 + \mu_{(12)3}\cos 2\gamma}$$

which for very small values of γ coincides with (4.176). This makes that also the coupling $\omega_3'\dot{\gamma}$ disappears and the classical kinetic energy of the linear GTM reads :

$$\begin{aligned} T_{\rm cl} &\approx \frac{1}{2}\mu_{12}\dot{\rho}^2 + \frac{1}{2}\mu_{(12)3}\dot{\lambda}^2 + \frac{1}{2}\frac{\mu_{12}\rho^2\mu_{(12)3}\lambda^2}{\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2}\dot{\varepsilon}^2 \\ &+ \frac{1}{2}(\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2)(\omega_1'^2 + \omega_2'^2) + \frac{1}{2}\frac{\mu_{12}\rho^2\mu_{(12)3}\lambda^2}{\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2}\varepsilon^2\omega_3'^2 \end{aligned} \tag{4.196}$$

Upon quantization one gets

$$\begin{aligned} \widehat{T} &= -\frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\rho^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} - \frac{\hbar^2}{2}\left(\frac{1}{\mu_{(12)3}\lambda^2} + \frac{1}{\mu_{12}\rho^2}\right)\frac{\partial^2}{\partial\varepsilon^2} \\ &+ \frac{\hbar^2}{2(\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2)}(\boldsymbol{L}^2 - L_3'^2) + \frac{\hbar^2}{2\varepsilon^2}\left(\frac{1}{\mu_{(12)3}\lambda^2} + \frac{1}{\mu_{12}\rho^2}\right)\left(L_3'^2 - \frac{1}{4}\right) \end{aligned} \tag{4.197}$$

Using the same approximations as in the previous case we obtain for the fluctuating part of the potential in the zeroth-order approximation

$$\widehat{V} = \frac{1}{2}C_\rho\delta\rho^2 + \frac{1}{2}C_\lambda\delta\lambda^2 + \frac{1}{2}C_\lambda\lambda_0^2\varepsilon^2 + \frac{1}{2}C_{\rho\lambda}\delta\rho\delta\lambda \tag{4.198}$$

Thus in the linear case we deal with a ρ-λ coupling instead of a ρ-ε one. Undertaking the same steps as previously, the zeroth-order spectrum is readily computed :

$$\begin{aligned} E^{(0)}_{IKn_\rho n\varepsilon n_\lambda} &= \hbar\tilde{\omega}_\rho\left(n_\rho + \frac{1}{2}\right) + \hbar\tilde{\omega}_\lambda\left(n_\lambda + \frac{1}{2}\right) + \hbar\omega_\varepsilon\left(|K| + n_\varepsilon + \frac{3}{2}\right) \\ &+ \frac{\hbar^2}{2(\mu_{12}\rho_0^2 + \mu_{(12)3}\lambda_0^2)}\left[I(I+1) - K^2\right] \end{aligned} \tag{4.199}$$

where the frequencies are defined as follows

$$\tilde{\omega}^2_{\rho(\lambda)} = \frac{1}{2}\left(\omega_\rho^2 + \omega_\lambda^2 \pm (\omega_\rho^2 - \omega_\lambda^2)\sec 2\eta\right) \tag{4.200}$$

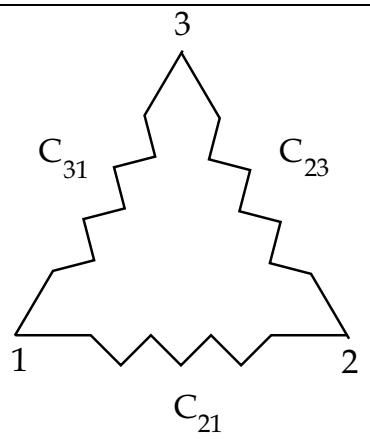

Figure 4.14: The three cluster molecule is presented via three mass points. Between each two mass points a spring is plotted, representing the approximate harmonic interaction between these mass points. The numbers 1 to 3 enumerate the clusters.

in case $\tilde{\omega}_\rho > \tilde{\omega}_\lambda$ and

$$\omega_\varepsilon = \lambda_0 \sqrt{C_\lambda \left(\frac{1}{\mu_{(12)3}\lambda^2} + \frac{1}{\mu_{12}\rho^2} \right)} \tag{4.201}$$

The parameter η is determined from a relation similar to (4.186) with the difference that ε is traded for λ.

$$\tan 2\eta = -\frac{2C_{\rho\lambda}}{\sqrt{\mu_{12}\mu_{(12)3}}(\omega_\rho^2 - \omega_\lambda^2)}$$

The rotational spectrum of the linear molecule is approximately two times more compressed than the one corresponding to the triangular configuration. This time the excited rotational 1_1^- state is at 2.8 keV and the 2_1^+ state at 8.33 keV. Since the molecule is linear there is a pronounced hindrance to $K \neq 0$ rotations, the state $1^\pm$ of the $K = 1$ band being located at 14.5 MeV. The first vibrational band heads are located at almost the same energies as in the triangular case.

4.3 Algebraic Models

4.3.1 U(7) Model of Trinuclear Quasimolecules

In the subsection 4.2 the GTM was treated by assuming that : i) the system is in a linear configuration, ii) the inclination angles of the nuclear symmetry axis to the axis which defines the linear orientation should be very small and iii) the light cluster has to be sandwiched between the two heavy ones. An algebraic model, inspired by previous work on baryon structure [22], might overcome these restrictions. The advantage of this model is the easiness in how one can describe complicated systems, which would require complex procedures in the geometrical model. Also it permits to discuss possible dynamical symmetry limits, allowing for an analytical description of the spectrum. This latter part will not be discussed here. The

main idea of the algebraic model were presented in [23]. The main problem is how to obtain the model parameters starting from known stiffness parameters, describing the pairwise interaction between the clusters. On one side we will have the picture of three masses connected via a spring (see Fig. 4.14) and on the other side there is the algebraic model. The mapping can not be one-to-one but it should reproduce at low energies similar frequencies of the *vibrational* modes. For the rotational part we will assume a simple expression. The model does not contain yet the vibrational contributions of the individual clusters, i.e. no β and γ vibrations. In other words, the clusters are treated without structure yet. Without any inner structure, a geometrical model can still be carried out, as shown in this paper. However, the introduction of structure to the clusters will be straight forward within the algebraic model and without the problems involved in the geometrical treatment. As one possibility, one can introduce the IBA-I Hamiltonian [24] for the individual clusters, using a dynamical symmetry limit for simplicity. As it turned out in Ref. [18], the dominant contribution to the energy of a linear configuration comes from the relative motion of the nuclei and not from the deformation of the clusters. The deformation plays a role in the vibrational modes of the individual clusters.

In what follows we will briefly present the U(7) model and the Hamiltonian.

In [22] an algebraic model for three particles with identical mass was proposed, in the context of the orbital excitations of quarks. In [25] the model was extended to three particles with, in general, different masses, intended to be applied to atomic molecules. The U(7) model, whose name will become more transparent further below, can be applied to any kind of three particle systems, as the three-cluster molecule [18], discussed in the introduction.

The number of degrees of freedom are six and for each relative coordinate we can introduce boson creation and annihilation operators, carrying negative parity. The basic concept of the U(7) model is to introduce a cutoff through the addition of a scalar boson of positive parity. With this the spherical components of the creation operators are given by

$$p^\dagger_{\rho,m}, \quad p^\dagger_{\lambda,m}, \quad s^\dagger \quad (m=-1,0,1). \tag{4.202}$$

The total number of bosons $N = n_\rho + n_\lambda + n_s$ is conserved, which implies that the total number of p-bosons is restricted between zero and N.

Taking all possible double bilinear products of a creation with an annihilation operator, we obtain the algebra u(7) with its 49 generators. A convenient form of the generators in terms of tensors with definite angular momentum is given in Eq. (5) of Ref. [25]. The classification and the structure of possible dynamical subgroups is given in Ref. [22]. The most convenient basis, with respect to which the model Hamilton operator will be diagonalized, is the one given by Eq. (4.2) of Ref. [22]. The basis states are given by

$$\mid N, (n_\rho, L_\rho), (n_\lambda, L_\lambda); LM_L > \quad , \tag{4.203}$$

with n_ρ and n_λ the number operator of the ρ- and λ- oscillation quanta respectively. The L_ρ and L_λ are the angular momenta of the ρ and λ part, L is the total angular momentum and M_L its projection.

As the model Hamiltonian we use the one given by Eq. (12) of Ref. [25] plus a rotational energy contribution. We will resume it here for completeness:

$$\begin{aligned} H &= a_L \boldsymbol{L}^2 + A P_1^\dagger P_1 + C P_2^\dagger P_2 + C P_3^\dagger P_3 \\ &+ D(P_1^\dagger P_2 + P_2^\dagger P_1) + E(P_1^\dagger P_3 + P_3^\dagger P_1) + F(P_2^\dagger P_3 + P_3^\dagger P_2) \quad . \end{aligned} \tag{4.204}$$

The operators P_i are defined via

$$\begin{aligned} P_1^\dagger &= p_\rho^\dagger \cdot p_\rho + p_\lambda^\dagger \cdot p_\lambda - R_0^2 s^\dagger s^\dagger \\ P_2^\dagger &= \sin^2\beta_0\, p_\lambda^\dagger \cdot p_\lambda - \cos^2\beta_0\, p_\rho^\dagger \cdot p_\rho \\ P_3^\dagger &= \sin(2\beta_0) p_\rho^\dagger \cdot p_\lambda - \cos\gamma_0(\sin^2\beta_0\, p_\lambda^\dagger \cdot p_\lambda + \cos^2\beta_0\, p_\rho^\dagger \cdot p_\rho) \quad , \end{aligned} \tag{4.205}$$

where the parameter $R_0 = \sqrt{\boldsymbol{\rho}_0 \cdot \boldsymbol{\rho}_0 + \boldsymbol{\lambda}_0 \cdot \boldsymbol{\lambda}_0}$ describes the extension of the system. The β_0 is defined by the relative size of $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$, i.e. $\lambda_0 = R_0 \cos\beta_0$ and $\rho_0 = R_0 \sin\beta_0$, and γ_0 gives the angle between the two vectors. The index zero indicates the equilibrium position of the system around which the oscillations take place. The parameters R_0, β_0 and γ_0 can be related to the definition of a coherent state, which at these values describes the approximate eigenstate of the system [25]. The coherent state is defined via

$$| NR_0, \beta_0\gamma_0 >= \frac{1}{\sqrt{N!}}(b_c^\dagger)^N \mid 0 > \quad , \tag{4.206}$$

where

$$b_c^\dagger = \frac{\left[s^\dagger + R_0\cos\beta_0 p_{\lambda,x}^\dagger + R_0 \sin\beta_0(\cos\gamma_0 p_{\rho,x}^\dagger + \sin\gamma_0 p_{\rho,y}^\dagger)\right]}{\sqrt{1+R_0^2}}$$

is called the *condensate boson*. For more details, see Ref. [25] and references therein.

In order to describe the motion around the equilibrium position, fluctuation bosons are introduced in [25], which are orthogonal to $b_c^\dagger$. These are $b_u^\dagger$, describing the breathing mode, $b_v^\dagger$, the butterfly mode, and $b_w^\dagger$ is the mode where the angle γ between the vectors $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ is changing (shearing mode).

A Bogoliubov treatment is applied, where the $b_c^\dagger$ and b_c are substituted by their expectation value $\sqrt{N}$ and only leading terms in N are taken into account. The Hamiltonian obtained has the form

$$H_B = \sum_{\alpha_1,\alpha_2} \varepsilon_{\alpha_1\alpha_2} b_{\alpha_1}^\dagger b_{\alpha_2} \tag{4.207}$$

with $i, j = u$,v, w. The frequencies are given by [25]

$$\begin{aligned} \varepsilon_u &= 4ANR_0^2 \\ \varepsilon_v &= BNR_0^2 \sin^2(2\beta_0)/(1+R_0^2) \\ \varepsilon_w &= CNR_0^2 \sin^2(2\beta_0)\sin^2\gamma_0/(1+R_0^2) \\ \varepsilon_{uv} &= 2DNR_0^2 \sin(2\beta_0)/\sqrt{1+R_0^2} \\ \varepsilon_{uw} &= 2ENR_0^2 \sin(2\beta_0)\sin\gamma_0/\sqrt{1+R_0^2} \\ \varepsilon_{vw} &= FNR_0^2 \sin^2(2\beta_0)\sin\gamma_0/(1+R_0^2) \quad . \end{aligned} \tag{4.208}$$

These are the estimates of the lowest frequencies, which we will need later in order to relate them to those calculated in a nuclear interaction model.

In order to determine the parameters of the U(7) model, in a first step the stiffness of oscillations between each two of the three clusters is determined. For illustration, see Fig.4.14

where the interaction between two clusters is presented via a spring. Afterwards we will apply several steps until we can compare the energies at low values to those of the Hamilton of eq. (4.207). Because the Hamiltonian, in a geometrical picture, is not identical to the one of the U(7) model, we will require that the lowest frequencies are similar. The geometrical potential, we start with, is itself an approximation to the one calculated between the nuclei, i.e. a mapping which reproduces the lowest frequencies is more than sufficient. The mapping is necessary because in the algebraic model there is no direct relation to the stiffnesses which are calculated in a nuclear interaction model.

The stiffness coefficients are again calculated using the double folding potential and the resulting potential can exhibit at some point a minimum. A quadratic expansion around this minimum is made, giving the stiffness C_{ij} of the potential between cluster i and j.

The relevant degrees of freedom of the three particle system, excluding the center of mass motion, can be cast into Jacobi coordinates, and defined in (4.163). This definition deviates by some factors to the one given in [25]. It is not relevant for the further steps. Doing the same for the coordinates at the equilibrium position, we can define $\delta\boldsymbol{\rho} = \boldsymbol{\rho} - \boldsymbol{\rho}_0$ and $\delta\boldsymbol{\lambda} = \boldsymbol{\lambda} - \boldsymbol{\lambda}_0$. This allows us to relate the spatial differences between two clusters to the difference vectors $\delta\boldsymbol{\rho}$ and $\delta\boldsymbol{\lambda}$. Further we have

$$\begin{aligned} \boldsymbol{r_2} - \boldsymbol{r_1} &= \boldsymbol{\rho} \\ \boldsymbol{r_3} - \boldsymbol{r_1} &= \frac{m_2}{m_1+m_2}\boldsymbol{\rho} - \boldsymbol{\lambda} \\ \boldsymbol{r_2} - \boldsymbol{r_3} &= \frac{m_1}{m_1+m_2}\boldsymbol{\rho} + \boldsymbol{\lambda} \quad , \end{aligned} \tag{4.209}$$

where m_k refer to the masses involved.

The classical Hamiltonian, from which we start, and where the center of mass motion is excluded already, is given by

$$H_{cl} = \frac{\mu_{12}}{2}\delta\dot{\boldsymbol{\rho}}^2 + \frac{\mu_{(12)3}}{2}\delta\dot{\boldsymbol{\lambda}}^2 + \frac{C_\rho}{2}\delta\boldsymbol{\rho}^2 + \frac{C_\lambda}{2}\delta\boldsymbol{\lambda}^2 + C_{\rho\lambda}\delta\boldsymbol{\rho}\cdot\delta\boldsymbol{\lambda} \tag{4.210}$$

with

$$\begin{aligned} C_\rho &= C_{21} + \frac{C_{23}m_1^2 + C_{31}m_2^2}{(m_1+m_2)^2} \\ C_\lambda &= C_{23} + C_{31} \\ C_{\rho\lambda} &= \frac{C_{23}m_1 - C_{31}m_2}{m_1+m_2} \end{aligned} \tag{4.211}$$

and μ_{12} is the reduced mass between cluster 1 and 2, while $\mu_{(12)3}$ is the reduced mass between cluster 3 and the combined mass $(m_1 + m_2)$ at the position of the center of mass of the first two clusters.

When the vector $\boldsymbol{\lambda}$ is defined along the molecular x axis and the vector $\boldsymbol{\rho}$ in the molecular (xz)-plane (note that the molecular y-axis is defined perpendicular to the plane of the molecule) the potential acquires the form

$$V_{\text{cl}} = \frac{C_\rho}{2}\delta\rho^2 + \frac{C_\rho}{2}\rho_0^2\delta\gamma^2 + \frac{C_\lambda}{2}\delta\lambda^2 + C_{\rho\lambda}(\cos\gamma_0\delta\rho\delta\lambda - \rho_0\sin\gamma_0\delta\gamma\delta\lambda) \tag{4.212}$$

where γ_0 and ρ_0 are the equilibrium values of γ and ρ at the potential minimum and an expansion up to quadratic terms was made. We are interested in the pure vibrational part only. Excluding the rotational part and possible mixings the kinetic energy is given by

$$\hat{T}_{\text{vib}} = \frac{\mu_{12}}{2}\delta\dot{\rho}^2 + \frac{\mu_{(12)3}}{2}\delta\dot{\lambda}^2 + \frac{\mu_{12}\rho_0^2}{2}\delta\dot{\gamma}^2 \quad . \tag{4.213}$$

The ρ and λ give the length of the corresponding vectors.

The classical Hamiltonian is quantized using the Pauli-Podolsky procedure [1]. The expression is given by

$$\widehat{T}_{\text{vib}} = -\frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\rho^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} - \frac{\hbar^2}{2}\left(\frac{1}{\mu_{12}\rho_0^2} + \frac{1}{\mu_{(12)3}\lambda_0^2}\right)\frac{\partial^2}{\partial\gamma^2} \quad . \tag{4.214}$$

In order to *estimate* the vibrational energies we set, for a brief moment, $C_{\rho\lambda} = 0$. This is the case where the coupling terms of the potential between λ, ρ and γ vanish. The vibrational part of the Hamiltonian can be solved analytically with the frequencies

$$\begin{aligned}
\varepsilon_\rho &= \hbar\sqrt{\frac{C_\rho}{\mu_{12}}} \\
\varepsilon_\lambda &= \hbar\sqrt{\frac{C_\lambda}{\mu_{(12)3}}} \\
\varepsilon_\gamma &= \hbar\sqrt{C_\rho\rho_0^2\left(\frac{1}{\mu_{12}\rho_0^2} + \frac{1}{\mu_{(12)3}\lambda_0^2}\right)} \quad .
\end{aligned} \tag{4.215}$$

They refer to the *fluctuations* in ρ, λ and γ respectively.

For the rotational part we assume a simple form $a\boldsymbol{L}^2$. More terms can be introduced, like $\boldsymbol{L}_\rho^2$ or $\boldsymbol{L}_\lambda^2$ (see ref. [25]). As for the value a we take $\frac{1}{2\Theta_0}$, with $\Theta_0 = (\mu_{12}\rho_0^2 + \mu_{(12)3}\lambda_0^2)$ which is the largest moment of inertia possible. For the rotational kinetic energy we finally have

$$\widehat{T}_{\text{rot}} = \frac{\hbar^2}{2(\mu_{12}\rho_0^2 + \mu_{(12)3}\lambda_0^2)}\boldsymbol{L}^2 \quad , \tag{4.216}$$

which is still too simple because in general mixing terms like $\boldsymbol{L}_{k_1}\boldsymbol{L}_{k_2}$ occur. These terms have to be simulated by some combination of operators of the U(7) model, which has not been done yet.

A problem is still related to the interaction part. If we substitute the fluctuation coordinates $\delta\rho$, $\delta\lambda$ and $\delta\gamma$ in terms of their boson creation and annihilation operators, terms of the type $p^\dagger p^\dagger$ appear, not conserving the number of bosons. One possible proposal is to substitute it by

$$\begin{aligned}
\delta\rho\delta\lambda &\rightarrow \frac{1}{2}(\delta\rho\delta\lambda + \pi_{\delta\rho}\pi_{\delta\lambda}) \\
\delta\gamma\delta\lambda &\rightarrow \frac{1}{2}(\delta\gamma\delta\lambda + \pi_{\delta\gamma}\pi_{\delta\lambda}) \quad ,
\end{aligned} \tag{4.217}$$

where $\pi_{\delta\rho}$, $\pi_{\delta\lambda}$ and $\pi_{\delta\gamma}$ are the conjugate momenta to the corresponding coordinates. Here we really change the original Hamiltonian. The change is justified by three arguments: i) the frequencies in (4.215) are of the order of several MeV, which implies that the mixing between the oscillations, given by the terms $p^\dagger p^\dagger$, is probably small ($2\hbar\omega_\alpha$ excitations) and ii) the parameter in front of this interaction turns out to be small for the system discussed. iii) The Hamiltonian, we started with, is itself an approximation and changing it to the more advantageous form of the U(7) model might not change much. Nevertheless, in other situations the procedure might not be valid.

As the next step we relate the fluctuations in ρ, λ and γ to the breathing (u), the butterfly (v) and the shearing mode (w). Inspired by [25] we take

$$\begin{aligned} p^\dagger_{\delta\rho} &= \sin\beta_0 b^\dagger_u - \cos\beta_0 b^\dagger_v \\ p^\dagger_{\delta\lambda} &= \cos\beta_0 b^\dagger_u + \sin\beta_0 b^\dagger_v \\ p^\dagger_{\delta\gamma} &= p^\dagger_w \quad . \end{aligned} \tag{4.218}$$

With this we find as the final vibrational Hamiltonian

$$\begin{aligned} \widehat{H}_{\text{vib}} &= \varepsilon_u b^\dagger_u b_u + \varepsilon_v b^\dagger_v b_v + \varepsilon_w b^\dagger_w b_w \\ &+ \varepsilon_{uv}(b^\dagger_u b_v + b^\dagger_u b_v) + \varepsilon_{uw}(b^\dagger_u b_w + b^\dagger_u b_w) \\ &+ \varepsilon_{vw}(b^\dagger_v b_w + b^\dagger_w b_v) \quad , \end{aligned} \tag{4.219}$$

with

$$\begin{aligned} \varepsilon_u &= \varepsilon_\rho + \varepsilon_\lambda + \frac{C_{\rho\lambda}\hbar}{2[\mu_{12}C_\rho\mu_{(12)3}C_\lambda]^{\frac{1}{4}}}\sin\beta_0\cos\beta_0\cos\gamma_0 \\ \varepsilon_v &= \varepsilon_\rho + \varepsilon_\lambda - \frac{C_{\rho\lambda}\hbar}{2[\mu_{12}C_\rho\mu_{(12)3}C_\lambda]^{\frac{1}{4}}}\sin\beta_0\cos\beta_0\cos\gamma_0 \\ \varepsilon_w &= \varepsilon_\gamma \\ \varepsilon_{uv} &= (\varepsilon_\lambda - \varepsilon_\rho)\sin\beta_0\cos\beta_0 - \frac{C_{\rho\lambda}\hbar}{2[\mu_{12}C_\rho\mu_{(12)3}C_\lambda]^{\frac{1}{4}}}\cos\gamma_0\cos(2\beta_0) \\ \varepsilon_{uw} &= -\frac{C_{\rho\lambda}\hbar}{2[\mu_{12}C_\rho\mu_{(12)3}C_\lambda]^{\frac{1}{4}}}\sin\gamma_0\cos\beta_0 \\ \varepsilon_{uv} &= -\frac{C_{\rho\lambda}\hbar}{2[\mu_{12}C_\rho\mu_{(12)3}C_\lambda]^{\frac{1}{4}}}\sin\gamma_0\sin\beta_0 \quad . \end{aligned} \tag{4.220}$$

These values have to be compared to the ones given in (4.208).

Application to ^{132}Sn $+ \alpha +$ ^{116}Pd

The double folding potential provides us the values of the stiffness coefficients in the quadratic approximation as discussed in 4.2.2 Since the present model copes with spherical clusters only the monopolar term is retained in the expansion of the potential.

For the α particle we use a gaussian-like density with a width parameter $\beta = 0.69\text{fm}^{-1}$. For the strength of the compression term $V_{\text{comp},0}$ we used for exemplification 2 choices: 1)

V_{comp12}	V_{comp23}	V_{comp13}	K_{12}	K_{23}	K_{13}
187.5	228.7	225.5	214.9	213.2	218.8
300.	300.	300.	383.	327.	324.
$R_{12\text{min}}(\rho_0)$	$R_{23\text{min}}$	$R_{13\text{min}}$	λ_0	β_0	γ_0
11.0	7.37	7.65	5.14	65°	84°
11.1	7.53	7.82	5.34	64^o17'	84°
C_{12}	C_{23}	C_{13}	C_ρ	C_λ	$C_{\rho\lambda}$
97.3	17.77	10.29	104.59	28.06	-4.64
163.8	25.43	12.63	173.8	38.06	-7.63

Table 4.4: Relevant parameters of the theory, including the values for the stiffness of the potential parameters.

We seek to reproduce the nuclear compression modulus K in accordance to the nuclear EOS (nuclear equation of state) [26] by varying $V_{\text{comp,0}}$ separately for all the three pairs (see first line of Table 4.4 where all the values are in MeV). 2) A unique value of $V_{\text{comp,0}}$ = 300 MeV is used, as have been done in a calculation of α-like nuclear molecules life-times [27](see second rows of Table 4.4). This choice obviously leads to sensitively larger nuclear compressibilities, which according to experiments on giant monopole resonance should range between 180 and 240 MeV [28].

Along with the stiffness parameters we obtained also the location of the absolute minima of the total ternary potential [29] which we chose to be the equilibrium position of our triangular molecule. In Fig. 4.15 the situation is illustrated, where at the position 1 there is the ^{132}Sn, at 2 the ^{116}Pd and at 3 the α particle. We obtained for the equilibrium positions ρ_0 and

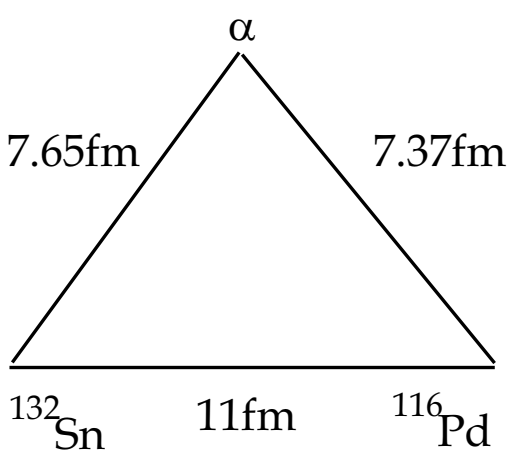

Figure 4.15: The geometry of the system $^{132}\text{Sn} + \alpha + {}^{116}\text{Pd}$.

A[MeV]	B[MeV]	C[MeV]	D[MeV]
0.0043428	4.3748	1.4507	0.018401
E[MeV]	F[MeV]	a_L[MeV]	-
-0.018401	-0.012917	-0.87808	-

Table 4.5: Parameter values of the U(7) Hamiltonian.

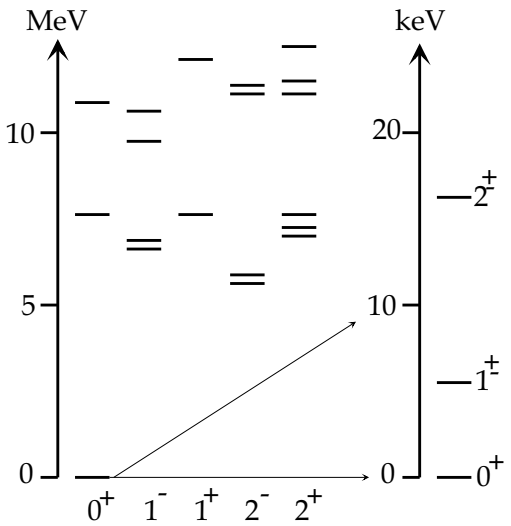

Figure 4.16: The spectrum of ^{132}Sn $+ \alpha +$ ^{116}Pd as obtained in the U(7) model. On the left hand side all states up to spin 2 are shown, except for the excited states of the ground state band. The latter are given on the right hand side. There is a degeneracy of positive and negative spin states for spin 1 and 2 in the ground state band.

λ_0 the values 11 fm and 5.14 fm respectively. For the angle γ_0 we obtain $\approx 84^0$.

Taking as $N = 10$ and using Eqs. (4.208), (4.211) and (4.220), we obtain the parameters A to F and a_L which are listed in Table 4.5.

Setting the $C_{\rho\lambda}$ equal to 0 does change A, B, C and D only at the last digits shown, however E and F are 0. This situation is the same as in the geometric picture when we extracted the eigenfrequencies ε_ρ, ε_λ and $\varepsilon_\gamma = \varepsilon_\rho$ for $C_{\rho\lambda} = 0$. The values are $\varepsilon_\rho = 8.386$MeV, $\varepsilon_\lambda = 17.20$MeV and $\varepsilon_\gamma = 71.57$MeV. The latter vibrational state is too high in order to be believed to exist. After the mapping, this division into ε_ρ and ε_λ, which are nearly separated, gets lost and there is a strong mixing between the breathing (u) and the butterfly (v) mode, which manifests itself in a large ε_{uv}. In order to compare the numerical evaluation to the estimates of the frequencies as given in Eq. (4.215) and using the parameter values as given in Table 4.4, we have to put $C_{\varrho\lambda} = 0$ in order that the mixing between the ρ- and λ-modes does not alter the position of the vibrational states. We obtain as the lowest frequency for the (u) and (γ) motion the values 9.1851MeV and 17.7067 MeV, while for the (v) mode we have $\approx$73MeV. These values are sufficiently close to the ones given above.

When the mixing due to $C_{\rho\lambda}$ is taken into account, the spectrum changes by shifting

ε_u	ε_v	ε_w	ε_{uv}	ε_{uw}	ε_{vw}
25.6083	25.5671	8.3857	3.4159	-2.3847	-5.1035

Table 4.6: Values of the parameters of Eq. (4.220), in MeV.

the vibrational modes to lower energies. In Figure 4.16 we present the spectrum obtained. According to this, the ground state band is severely squeezed as for the case of the linear molecule [18]. However, for the excited vibrational states there is no clear band structure apparent. The reason is the following: In the zero order approximation of our mapping, obtaining the values ε_α's of Eq. (4.220), no Coriolis force is present in an apparent form. However, in the $\frac{1}{N}$ corrections the $U(7)$ model introduces Coriolis couplings leading to the effect seen. Due to a strong Coriolis force, the band structure gets distorted.

The values of the vibrational parameters, as given in (4.220) are given in Table 4.6 where it can be noticed that the mixing between the butterfly (v) and the breathing (u) mode is very strong, i.e. it will be difficult to obtain states with a pure motion related to one of the two modes. As it seems, in triaxial nuclear molecules, with two heavy and one light cluster, there is no clear separation between the u- and the v-mode. In other configurations this might be possible, when the value of ε_{uv} is small compared to ε_u and ε_v.

One signature in the γ-spectrum for the existence of a three-cluster molecule would be a transition from a vibrational state to levels in the ground state rotational band. Because the ground state band is strongly squeezed in energy, one should observe a splitting of the transition line into several others with only a few keV apart. Selection rules in angular momentum have to be taken into account and will limit the amount of splitted levels. For example, an $E1$-transition from a 1^+ vibrational state is only allowed to decay to the 1^- and 2^- levels of the ground state band (see Fig.4.16).

Bibliography

[1] J.Eisenberg and W.Greiner, Nuclear Theory, vol.I, Phenomenological Models, North-Holland, Amsterdam 1988.

[2] E.Uegaki and Y.Abe, Progr.Theor.Phys.**90**, (1993) 615.

[3] P.O.Hess, W.Scheid, W.Greiner and J.H.Hamilton, Jour. Phys. G **25**, L139 (1999).

[4] Ş. Mişicu, P.O.Hess, A. Sandulescu and W. Greiner, J.Phys.**G25** L133 (1999).

[5] P.O.Hess, W.Greiner and W.T.Pinkston, Phys.Rev.Lett.**53**, 1535 (1984);
P.O.Hess and W.Greiner, *Il Nuovo Cimento* **83** 76 (1984).

[6] Ş.Mişicu, A.Sandulescu, F.Carstoiu, M.Rizea and W.Greiner, Il Nuovo Cimento **A112** 313 (1999).

[7] A.R.Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton, New Jersey, 1974).

[8] P.Möller, J.R.Nix, W.D.Myers and W.J.Swiatecki, At. Data and Nucl. Data Tables **59**, 185 (1995).

[9] W.D.Myers and W.J.Świątecki, *Phys.Rev.C* **57** 3020 (1998).

[10] V.N.Bragin and M.V.Zhukov, Sov.J.Part.Nucl. **15**, 325 (1984).

[11] J.H.Hamilton *et al.*, in *Proc. of the Symposium Fundamental Issuses in Elementary Matter, Bad Honnef, Germany, September 25-29, 2000*, edited by W.Greiner (EP Systems Bt., Debrecen, 2001), p.151.

[12] S.Raman et al., At. Data and Nucl. Data Tables **36**, 1 (1987).

[13] C.Y.Cha, N.Nordberg, R.B.Firestone and L.P.Ekström, ISOTOPE EXPLORER 2.23, Jan. 28 (1999).

[14] P.Ring and P.Schuck, *The Nuclear Many Body Problem* (Heidelberg, Springer-Verlag, 1980).

[15] M.Sakai, At. Data and Nucl. Data Tables **31**, 400 (1984).

[16] D.Troltenier, J.A.Maruhn, W.Greiner, V.Velazquez, P.O.Hess and J.H.Hamilton, Zeit.Phys. A **338**, 261 (1991).

[17] J.G.Hirsch, O.Castaños, P.O.Hess and O.Civitarese, Phys. Rev. C **51**, 2252 (1995).

[18] P.O. Hess, Ş. Mişicu, W. Greiner and W. Scheid, Jour. Phys. G **26** , 1 (2000);
P. O. Hess, Ş. Mişicu and W. Greiner, Rev. Mex. Fís. **46 S1**, 73 (2000).

[19] F.Carstoiu, I.Bulboaca, A.Săndulescu and W.Greiner, Phys.Rev C **61**, 044606-1 (2000).

[20] A.Florescu, A.Săndulescu, D.S.Delion, J.H.Hamilton, A.V.Ramayya and W.Greiner, Phys.Rev C **61**, 051602-1 (2000).

[21] J.-P.Blaizot and G.Ripka, *Quantum Theory of Finite Systems* (MIT Press, Cambridge-Massachusetts, 1986), p.58.

[22] R. Bijker, F. Iachello and A. Leviatan, Ann. Phys. (N.Y.) **236**, 69 (1994); Ann. Phys. (N.Y.) **284**, 89 (2000).

[23] R. Bijker, P. O. Hess and Ş Mişicu, Heavy Ion Physics **13**, 89 (2001); *Rev.Mex.Fis.* **47**, Supp.2, 52 (2001).

[24] F. Iachello and A. Arima, *The Interacting Boson Model*, (Cambridge Monographs on Mathematical Physics, Cambridge, 1987).

[25] R. Bijker and L. Leviatan, Few-Body Systems **25**, 89 (1998).

[26] W.D.Myers and W.J.Swiatecki, Phys. Rev. **C57**, 3020 (1998).

[27] F.Carstoiu, I.Bulboaca, A.Săndulescu and W.Greiner, Phys. Rev.**61** (2000) 044606

[28] J.P.Blaizot, Phys.Rep. **64C** (1980), 171.

[29] A.Sandulescu, Ş.Mişicu and W.Greiner , Symposium *Advances in Nuclear Physics*, eds.D.Poenaru and S.Stoica, Bucharest, pp.83-91, World Scientific 2000.