Notes of CH_3F-H_2 Project

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I. INTRODUCTION OF HAMILTONIAN AND BASIS

Within the electronic Born-Oppenheimer approximation, the potential energy for a system of weakly bond dimer depends only on the intermolecular distance and the relative orientations not the frame chosen to describe it. However, one have to give the Hamiltonian and the basis a space usually a frame to represent them. Once the frame is chosen, the Hamiltonian, the basis have actual mathematical formalization. While, there are two kinds of frame: Space-Fixed(SFF) and Body-Fixed Frame(BFF) used in bound state calculating in general. We use the latter in this system and the Hamiltonian reads

$$\hat{H}(R,\alpha,\beta,\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) = -\frac{\hbar^2}{2\mu}R^{-1}\frac{\partial^2}{\partial R^2}R + \frac{(\hat{\vec{J}}-\hat{\vec{j_1}}-\hat{\vec{j_2}})^2}{2\mu R^2} + \hat{H}_{\mathrm{CH_3F}} + \hat{H}_{\mathrm{H_2}} + V(R,\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \tag{1}$$

where R specifies the distance from the center of mass of CH_3F molecule to the center of mass of H_2 molecule, α and β denote the vector $\vec{R}$ respect to the space-fixed frame. The α_1,β_1,γ describe the rotation of CH_3F in Body-fixed frame and the α_2, β_2 describe the rotation of H_2 molecule. $\hat{J}$ is the total angular momentum operator for the whole complex in the space-fixed frame, $\hat{j}_1$ and $\hat{j}_2$ the angular momentum operator of CH_3F and H_2 in the body-fixed frame,respectively. μ is the reduced mass of the whole system: $\frac{1}{\mu} = \frac{1}{\mathrm{m_{CH_3F}}} + \frac{1}{\mathrm{m_{H_2}}}$.

The total basis is constructed by two parts: radial basis part and angular basis part. We use the $\sin R$ function as the radial basis and the Discrete Variable Representation (DVR) method to calculate the radial matrix elements. In general, the angular basis are of two kinds. One kind is coupled. It's main idea is using the total angular basis that coupled from the molecular angular basis and the end-over-end angular basis to describe the rotation of the system. The other kind is uncoupled which looks like a hartree product but only multiplying the molecular angular basis and end-over-end angular basis. Of course, if remove the coupled term (the $3j$ or Clesch-Gordan or vector coupling coefficient) of the coupled basis, you can get the uncoupled basis. However, both of the two are extensively used in the rovibrational spectra calculation and we use the uncoupled basis in this system. It's defined as $\boldsymbol{|j_1k_1m\rangle|j_2k_2\rangle|JKM\rangle}$. In which the $|j_1k_1m\rangle$ describe the CH_3F rotation, j_1 is the angular quantum number of CH_3F, k_1 is the projection of j_1 on the z-axis of body-fixed frame, m is the projection on the molecular rotational axis. $|j_2k_2\rangle$ is the H_2 molecule rotation basis and j_2 is the angular quantum number of H_2 and k_2 is the projection of j_2 on the z-axis of body-fixed frame. $|JKM\rangle$ describes the total rotation and J is the total rotational angular quantum number and K is also the projection of J on body-fixed frame, where

$$\langle\alpha,\beta,0|JKM\rangle = \sqrt{\frac{2J+1}{4\pi}}D^J_{MK}(\alpha,\beta,0)^* = \sqrt{\frac{2J+1}{4\pi}}e^{iM\alpha}d^J_{MK}(\beta) \tag{2}$$

$$\langle\alpha_1,\beta_1,\gamma|j_1k_1m\rangle = \sqrt{\frac{2j_1+1}{8\pi^2}}D^{j_1}_{k_1m}(\alpha_1,\beta_1,\gamma)^* = \sqrt{\frac{2j_1+1}{8\pi^2}}e^{ik_1\alpha}d^{j_1}_{k_1m}(\beta_1)e^{im\gamma} \tag{3}$$

$$\langle\alpha_2,\beta_2 0|j_2 0k_2\rangle = \sqrt{\frac{2j_2+1}{4\pi}}D^{j_2}_{k_20}(\alpha_2,\beta_2,0)^* = \sqrt{\frac{2j_2+1}{4\pi}}e^{ik_2\alpha}d^{j_2}_{k_20}(\beta_2) \tag{4}$$

and,

$$d^j_{Kk}(\beta) = \sum_v(-1)^v\frac{[(l+K)!(l-K)!(l+k)!(l-k)!]^{\frac{1}{2}}}{(l-k-v)!(l+K-v)!(v+k-K)!v!}$$
$$\times(\cos\frac{\beta}{2})^{2j+K-k-2v}(-\sin\frac{\beta}{2})^{k-K+2v} \tag{5}$$

The D^J_{MK} and d^j_{Kk} etc. are rotational matrix and Wigner d-matrix elements.

We can yields the parity adapted basis using molecular symmetry theory to reduce the calculation quantity as we have done in the CH_3F-He system. The basic idea is after vibrational averaging and separating the v_3 vibration of CH_3F molecule from the total vibration of the system, the CH_3F molecule can be treated as a rigid molecule. The molecular symmetry group (MS) or permutation-inversion (PI) of CH_3F-H_2 system is $D_{3h}(M)$ or G_{12}. Of course, it's isotopic with the D_{3h} point group. However, the MS or PI of CH_3F-He system is $C_{3v}(M)$ or G_6 and isotopic with the C_{3v} point group. $D_{3h}(M) = C_{3v}(M)\otimes C_s(M)$. From the view of symmetry group, the whole permutation operations $C_{3v}(M)$ come from the symmetry of CH_3F molecule in the CH_3F-He system. The permutation operations come from both of the symmetry of CH_3F and H_2 molecules in CH_3F-H_2. So, the $C_s(M)$ got in cause H_2 isn't a point like the helium atom. It's a linear rotor whose rotation splits the energy level into two that respect to the *para*-H_2 (restrict j_2 is even) and *ortho*-H_2 (restrict j_2 is odd), respectively.

The angular basis function for the irreducible representations of PI(C_{3v}) is shown below:

irreps	wavrfunction	restricted
A_1	$\|j00JM\rangle$	for even J
	$[\|jkKJM\rangle+(-1)^{J+k}\ \|j-k-KJM\rangle]/\sqrt{2}$	for $k \equiv 0$ (mod 3)
A_2	$\|j00JM\rangle$	for odd J
	$[\|jkKJM\rangle$-$(-1)^{J+k}\ \|j-k-KJM\rangle]/\sqrt{2}$	for $k \equiv 0$ (mod 3)
E	$\|jkKJM\rangle$	for $k \neq 0(mod\ 3)$
	$\|j-k-KJM\rangle$	

For conciseness, the angular basis can be classify the wave function into two categories in the calculation and our parity adapted basis read:

1.) $|\psi_n\rangle|j_1k_1m\rangle|j_2k_2\rangle|JKM\rangle$ $(|\psi_n\rangle|j_1-k_1-m|j_2-k_2\rangle|J-KM\rangle)$ for $m \neq 0$ (mod 3);

2.) $|\psi_n\rangle\left[|j_1k_1m\rangle|j_2k_2\rangle|JKM\rangle+(-1)^{J+m+P}|j-k_1-m\rangle|j_2-k_2\rangle|J-KM\rangle\right]/\sqrt{2(1+\delta_{k_1,0}\delta_{m,0}\delta_{K,0})}$ for $m \equiv 0$ (mod 3);

Of cause, it's looks like the parity adapted basis of CH_3F-He system cause the same permutation-inversion operator G_6 or $C_{3v}(M)$. $|j_2,k_2\rangle$ is introduced to represent the rotation of H_2 molecule and we restrict j_2 even or odd to describe the energy level split. As discussed before, we can divide these basis into four categories that respected to deferent molecule species. For example, the first basis can be divide into two categorise and correspond to *para*-CH_3F-*para*-H_2 which restricts $m \neq 0$ (mod 3), j_2 is even and *para*-CH_3F-*ortho*-H_2 which restricts $m \neq 0$ (mod 3), j_2 is odd, respectively.

The second basis can also be divide into two categorise and correspond to *ortho*-CH_3F-*para*-H_2 that restricts $m \equiv 0$ (mod 3), j_2 is even and *ortho*-CH_3F-*ortho*-H_2 that restricts $m \equiv 0$ (mod 3), j_2 is odd, respectively. At last, we'd better restrict the quantum numbers in order to remove the linear and null basis here. Those are $K \geq 0$; $k_1 \geq 0$ (if $K = 0$); $m \geq 0$ (if $k_1 = K = 0$).

In all of above equations, J and M are conserved because of the spatial isotropy in the absence of external magnetic or electric filed.

II. MATRIX ELEMENTS

A. The First Term of the Hamiltonian

1. use the first basis

The first term of the Hamiltonian is only correspond with the Radial basis and the angular basis is diagonal. So the matrix elements reads

$$\begin{aligned}&\langle JK'M|\langle j_1'k_1'm'|\langle j_2'k_2'|\langle\psi_{n'}|-\frac{\hbar^2}{2\mu}R^{-1}\frac{\partial^2}{\partial R^2}R|\psi_n\rangle|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\ &=\langle\psi_n'|-\frac{\hbar^2}{2\mu}R^{-1}\frac{\partial^2}{\partial R^2}R|\psi_n\rangle\delta_{K'K}\delta_{j_1'j_1}\delta_{k_1'k_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{k_2'k_2}\end{aligned} \tag{6}$$

2. use the second basis

$$\begin{aligned}&\langle\Theta^{JMPK'}_{j_1'k_1'm'j_2'k_2'n'}|-\frac{\hbar^2}{2\mu}R^{-1}\frac{\partial^2}{\partial R^2}R|\Theta^{JMPK}_{j_1k_1mj_2k_2n}\rangle \\ =&\frac{1}{2\sqrt{(1+\delta_{k_1',0}\delta_{m',0}\delta_{K',0})(1+\delta_{k_1,0}\delta_{m,0}\delta_{K,0})}}\Big[\langle JK'M|\langle j_1'k_1'm'|\langle j_2'k_2'| \\ &+(-1)^{J+m'+P}\langle J-K'M|\langle j_1'-k_1'-m'|\langle j_2'-k_2'|\Big]-\frac{\hbar^2}{2\mu}R^{-1}\frac{\partial^2}{\partial R^2}R \\ &\Big[|j_1k_1m\rangle|j_2k_2\rangle|JKM\rangle+(-1)^{J+m+P}|j-k_1m\rangle|j_2-k_2\rangle|J-KM\rangle\Big] \\ =&-\frac{\hbar^2}{2\mu}\langle\psi_{n'}|R^{-1}\frac{\partial^2}{\partial R^2}|\psi_n\rangle\frac{\delta_{j_1'j_1}\delta_{j_2'j_2}}{\sqrt{(1+\delta_{K'0}\delta_{k_1'0}\delta_{m'0})(1+\delta_{K0}\delta_{k_10}\delta_{m0})}}\times \\ &\left[\delta_{KK'}\delta_{k_1'k_1}\delta_{m'm}\delta_{k_2'k_2}+(-1)^{J+m+P}\delta_{K'-K}\delta_{k_1'-k_1}\delta_{m'-m}\delta_{k_2'-k_2}\right] \\ =&-\frac{\hbar^2}{2\mu}\langle\psi_{n'}|R^{-1}\frac{\partial^2}{\partial R^2}R|\psi_n\rangle\frac{\delta_{j_1'j_1}\delta_{j_2'j_2}\delta_{k_1'k_1}\delta_{K'K}\delta_{m'm}\delta_{k_2'k_2}}{1+\delta_{K0}\delta_{k_10}\delta_{m0}}\left(1+(-1)^{J+m+P}\delta_{K0}\delta_{k_10}\delta_{k_20}\delta_{m0}\right)\end{aligned} \tag{7}$$

$$\tag{8}$$

ATTENTION!!! The last equal exist unless $K\geq 0; k_1\geq 0; m\geq 0; k_2\geq 0$.

For $-\frac{\hbar^2}{2\mu}\langle\psi_{n'}|R^{-1}\frac{\partial^2}{\partial R^2}R|\psi_n\rangle$ is easy to calculate using DVR equation. It's matrix elements are:

$$T_{ii'}=\frac{\hbar^2}{2\mu}\frac{(-1)^{i-i'}}{(b-a)^2}\frac{\pi^2}{2}\{\frac{1}{\sin^2\frac{\pi(i-i')}{2N}}-\frac{1}{\sin^2\frac{\pi(i+i')}{2N}}\}\text{ for }(i\neq i'). \tag{9}$$

$$T_{ii} = \frac{\hbar^2}{2\mu}\frac{(-1)}{(b-a)^2}\frac{\pi^2}{2}[\frac{(2N^2+1)}{3} - \frac{1}{\sin\frac{\pi i}{N}}] \text{ for } (i = i^{'}). \tag{10}$$

I will not give more detail expressions deduction of DVR equation here. One can consult (JCP-82-1401(1985)).

B. The Second Term of the Hamiltonian

The second term of the Hamiltonian can be expanded to be:

$$\frac{1}{2\mu R^2}[\hat{J}^2 + (\hat{j}_1 + \hat{j}_2)^2 - 2\hat{J}(\hat{j}_1 + \hat{J}_2)] \tag{11}$$

where

$$(\hat{j}_1 + \hat{j}_2)^2 = j_1^2 + j_2^2 + 2j_{1z}j_{2z} + j_1^+ j_2^- + j_1^- j_2^+ \tag{12}$$

$$2\hat{J}\hat{j}_1 = 2J_z j_{1z} + j_1^+ J^+ + j_1^- J^- \tag{13}$$

$$2\hat{J}\hat{j}_2 = 2J_z j_{2z} + j_2^+ J^+ + J^- j_2^- \tag{14}$$

and

$$\hat{j}_{1\pm} = \hat{j}_{1x} \pm i\hat{j}_{1y}; \hat{j}_{2\pm} = \hat{j}_{2x} \pm i\hat{j}_{2y}; \hat{J}_\pm = \hat{J}_x \mp i\hat{J}_y \tag{15}$$

The ladder operator have the normal actions on the operands, i.e.,

$$\hat{j}_{1\pm}|j_1 k_1 m\rangle = \sqrt{j_1(j_1+1) - k_1(k_1 \pm 1)}|j_1 k_1 \pm 1 m\rangle \tag{16}$$

$$\hat{j}_{2\pm}|j_2 k_2\rangle = \sqrt{j_2(j_2+1) - k_2(k_2 \pm 1)}|j_2 k_2 \pm 1\rangle \tag{17}$$

$$\hat{J}_\pm|JKM\rangle = \sqrt{J(J+1) - K(K \pm 1)}|JK \pm 1M\rangle. \tag{18}$$

We set $C_{1\pm} = \sqrt{j_1(j_1+1) - k_1(k_1 \pm 1)};\ C_{2\pm} = \sqrt{j_2(j_2+1) - k_2(k_2 \pm 1)}$ and $C_{J\pm} = \sqrt{J(J+1) - K(K \pm 1)}$.

1. use the first basis

With all these relations above, we can get the matrix elements after some algebra change and they read:

$$
\begin{aligned}
&\langle JK'M|\langle j_1'k_1'm|j_2'k_2'|\langle\psi_{n'}|\frac{(J-j_1-j_2)^2}{2\mu R^2}|\psi_n\rangle|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\
= &\ \frac{\delta_{n'n}}{2\mu R_n^2}\Big[\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{K'K}\delta_{j_1'j_1}\delta_{k_1'k_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{k_2'k_2} \\
&+C_{1+}C_{2-}\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{K'K}\delta_{k_1'k_1+1}\delta_{k_2'k_2-1}+C_{1-}C_{2+}\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{K'K}\delta_{k_1'k_1-1}\delta_{k_2'k_2+1} \\
&-C_{1+}C_{J+}\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{K'K+1}\delta_{k_1'k_1+1}\delta_{k_2'k_2}-C_{1-}C_{J-}\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{K'K-1}\delta_{k_1'k_1-1}\delta_{k_2'k_2} \\
&-C_{2+}C_{J+}\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{K'K+1}\delta_{k_1'k_1}\delta_{k_2'k_2+1}-C_{2-}C_{J-}\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{K'K-1}\delta_{k_1'k_1}\delta_{k_2'k_2-1}\Big]
\end{aligned}
\tag{19}
$$

2. use the second basis

$$
\begin{aligned}
&\langle\Theta^{JK'PMn'}_{j_1'k_1'm_1j_2'k_2}|\frac{(J-j_1-j_2)^2}{2\mu R^2}|\Theta^{JKPMn}_{j_1k_1m_1j_2k_2}\rangle
\end{aligned}
\tag{20}
$$

$$
\begin{aligned}
= &\ \frac{1}{2\mu R_n^2}\frac{\delta_{n'n}}{2\sqrt{(1+\delta_{k_1',0}\delta_{m',0}\delta_{K',0})(1+\delta_{k_1,0}\delta_{m,0}\delta_{K,0})}}\Big[\langle JK'M|\langle j_1'k_1'm'|\langle j_2'k_2'| \\
&+(-1)^{J+m'+P}\langle J-K'M|\langle j_1'-k_1'-m|\langle j_2'-k_2'|\Big](J-j_1-j_2)^2\Big[|j_1k_1m\rangle|j_2k_2\rangle|JKM\rangle+ \\
&(-1)^{J+m+P}|j-k_1m\rangle|j_2-k_2\rangle|J-KM\rangle\Big]
\end{aligned}
$$

$$
\begin{aligned}
= &\ \frac{1}{4\mu R_n^2}\frac{\delta_{n'n}}{\sqrt{(1+\delta_{k_1',0}\delta_{m',0}\delta_{K',0})(1+\delta_{k_1,0}\delta_{m,0}\delta_{K,0})}} \\
&\Big[\langle JK'M|\langle j_1'k_1'm|j_2'k_2'|(J-j_1-j_2)^2|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\
&+(-1)^{J+m+P}\langle JK'M|\langle j_1'k_1'm|j_2'k_2'|(J-j_1-j_2)^2|j_2-k_2\rangle|j_1-k_1-m\rangle|J-KM\rangle \\
&+(-1)^{J+m'+P}\langle J-K'M|\langle j_1'-k_1'-m|\langle j_2'-k_2'|(J-j_1-j_2)^2|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\
&+(-1)^{m'+m}\langle J-K'M|\langle j_1'-k_1'-m|\langle j_2'-k_2'|(J-j_1-j_2)^2|j_2-k_2\rangle|j_1-k_1-m\rangle|J-KM\rangle\Big]
\end{aligned}
\tag{21}
$$

The matrix element of the first term in bracket is same as eq (19)., the second third and forth term given below.

$$(-1)^{J+m+P}\langle JK'M|\langle j_1'k_1'm'|j_2'k_2'|(J-j_1-j_2)^2|j_2-k_2\rangle|j_1-k_1-m\rangle|J-KM\rangle \tag{22}$$
$$\begin{aligned}
= \Big[&\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{K'-K}\delta_{j_1'j_1}\delta_{k_1'-k_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{k_2'-k_2} \\
&+C_{1-}C_{2+}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{K'-K}\delta_{k_1'-k_1+1}\delta_{k_2'-k_2-1}+C_{1+}C_{2-}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{K'-K}\delta_{k_1'-k_1-1}\delta_{k_2'-k_2+1} \\
&-C_{1-}C_{J-}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{K'-K+1}\delta_{k_1'-k_1+1}\delta_{k_2'-k_2}-C_{1+}C_{J+}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{K'-K-1}\delta_{k_1'-k_1-1}\delta_{k_2'k_2} \\
&-C_{2-}C_{J-}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{K'-K+1}\delta_{k_1'-k_1}\delta_{k_2'-k_2+1}-C_{2+}C_{J+}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{K'-K-1}\delta_{k_1'-k_1}\delta_{k_2'-k_2-1}\Big]
\end{aligned}$$

$$(-1)^{J+m'+P}\langle J-K'M|\langle j_1'-k_1'-m'|\langle j_2'-k_2'|(J-j_1-j_2)^2|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \tag{23}$$
$$\begin{aligned}
= \Big[&\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{-K'K}\delta_{j_1'j_1}\delta_{-k_1'k_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-k_2'k_2} \\
&+C_{1+}C_{2-}\delta_{j_1'j_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-K'K}\delta_{-k_1'k_1+1}\delta_{-k_2'k_2-1}+C_{1-}C_{2+}\delta_{j_1'j_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-K'K}\delta_{-k_1'k_1-1}\delta_{-k_2'k_2+1} \\
&-C_{1+}C_{J+}\delta_{j_1'j_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-K'K+1}\delta_{-k_1'k_1+1}\delta_{-k_2'k_2}-C_{1-}C_{J-}\delta_{j_1'j_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-K'K-1}\delta_{-k_1'k_1-1}\delta_{-k_2'k_2} \\
&-C_{2+}C_{J+}\delta_{j_1'j_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-K'K+1}\delta_{-k_1'k_1}\delta_{-k_2'k_2+1}-C_{2-}C_{J-}\delta_{j_1'j_1}\delta_{-m'm}\delta_{j_2'j_2}\delta_{-K'K-1}\delta_{-k_1'k_1}\delta_{-k_2'k_2-1}\Big]
\end{aligned}$$

$$(-1)^{m'+m}\langle J-K'M|\langle j_1'-k_1'-m'|\langle j_2'-k_2'|(J-j_1-j_2)^2||j_2-k_2\rangle|j_1-k_1-m\rangle|J-KM\rangle \tag{24}$$
$$\begin{aligned}
= \Big[&\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{-K'-K}\delta_{j_1'j_1}\delta_{-k_1'-k_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{-k_2'-k_2} \\
&+C_{1-}C_{2+}\delta_{j_1'j_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{-K'-K}\delta_{-k_1'-k_1+1}\delta_{-k_2'-k_2-1}+C_{1+}C_{2-}\delta_{j_1'j_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{-K'-K}\delta_{-k_1'-k_1-1}\delta_{-k_2'-k_2+1} \\
&-C_{1-}C_{J-}\delta_{j_1'j_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{K'-K+1}\delta_{-k_1'-k_1+1}\delta_{-k_2'-k_2}-C_{1+}C_{J+}\delta_{j_1'j_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{-K'-K-1}\delta_{-k_1'-k_1-1}\delta_{-k_2'-k_2} \\
&-C_{2-}C_{J-}\delta_{j_1'j_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{-K'-K+1}\delta_{-k_1'-k_1}\delta_{-k_2'-k_2+1}-C_{2+}C_{J+}\delta_{j_1'j_1}\delta_{-m'-m}\delta_{j_2'j_2}\delta_{-K'-K-1}\delta_{-k_1'-k_1}\delta_{-k_2'-k_2-1}\Big]
\end{aligned}$$

Sum the first and forth term, the diagonal elements reads:

$$2\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{K'K}\delta_{j_1'j_1}\delta_{k_1'k_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{k_2'k_2}$$

we can get the sum of m' and m is even from the function $\delta_{m'm}$. So $(1+(-1)^{m'+m})=2$. At last, the diagonal elements reduce as

$$2\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{K'K}\delta_{j_1'j_1}\delta_{k_1'k_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{k_2'k_2}$$

The nondiagonal elements reads:

$$
2\delta_{j_1'j_1}\delta_{m'm}\delta_{j_2'j_2}\Big[C_{1+}C_{2-}\delta_{K'K}\delta_{k_1'k_1+1}\delta_{k_2'k_2-1}+C_{1-}C_{2+}\delta_{K'K}\delta_{k_1'k_1-1}\delta_{k_2'k_2+1}-C_{1+}C_{J+}\delta_{K'K+1}\delta_{k_1'k_1+1}\delta_{k_2'k_2}
$$
$$
-C_{1-}C_{J-}\delta_{K'K-1}\delta_{k_1'k_1-1}\delta_{k_2'k_2}-C_{2+}C_{J+}\delta_{K'K+1}\delta_{k_1'k_1}\delta_{k_2'k_2+1}-C_{2-}C_{J-}\delta_{K'K-1}\delta_{k_1'k_1}\delta_{k_2'k_2-1}\Big] \tag{25}
$$

The second and third term can also be summed and after some algebraic reduction, we can get the diagonal elements

$$
2(-1)^{J+m+P}\big[J(J+1)+j_1(j_1+1)+j_2(j_2+1)+2k_1k_2-2Kk_1-2Kk_2\big]\delta_{K'-K}\delta_{j_1'j_1}\delta_{k_1'-k_1}\delta_{m'-m}\delta_{j_2'j_2}\delta_{k_2'-k_2}
$$

the nondiagonal elements

$$
\begin{aligned}
&2(-1)^{J+m+P}\delta_{j_1'j_1}\delta_{m'-m}\delta_{j_2'j_2}\Big[C_{1+}C_{2-}\delta_{K'-K}\delta_{k_1'-k_1-1}\delta_{k_2'-k_2+1}\\
&+C_{1-}C_{2+}\delta_{K'-K}\delta_{k_1'-k_1+1}\delta_{k_2'-k_2-1}-C_{1+}C_{J+}\delta_{K'-K-1}\delta_{k_1'-k_1-1}\delta_{k_2'-k_2}\\
&-C_{1-}C_{J-}\delta_{K'-K+1}\delta_{k_1'-k_1+1}\delta_{k_2'-k_2}-C_{2+}C_{J+}\delta_{K'-K-1}\delta_{k_1'-k_1}\delta_{k_2'-k_2-1}\\
&-C_{2-}C_{J-}\delta_{K'-K+1}\delta_{k_1'-k_1}\delta_{k_2'-k_2+1}\Big]
\end{aligned} \tag{26}
$$

At last, we get the matrix elements of this term:

$$
\begin{aligned}
= \quad & \frac{\delta_{n'n}\delta_{j_1'j_1}\delta_{j_2'j_2}}{2\mu R_n^2\sqrt{(1+\delta_{K'0}\delta_{k_1'0}\delta_{m'0})(1+\delta_{K0}\delta_{k_10}\delta_{m0})}}\times\\
&\Big[E_{Jj_1j_2}[\delta_{m'm}\delta_{K'K}\delta_{k_1'k_1}\delta_{k_2'k_2}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K}\delta_{k_1',-k_1}\delta_{k_2',-k_2}]\\
&+C_{1+}C_{2-}[\delta_{m'm}\delta_{K'K}\delta_{k_1'k_1+1}\delta_{k_2'k_2-1}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K}\delta_{k_1',-k_1-1}\delta_{k_2,-k_2'+1}]\\
&+C_{1-}C_{2+}[\delta_{m'm}\delta_{K'K}\delta_{k_1'k_1-1}\delta_{k_2'k_2+1}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K}\delta_{k_1',-k_1+1}\delta_{k_2,-k_2'-1}]\\
&-C_{1+}C_{J+}[\delta_{m'm}\delta_{K'K+1}\delta_{k_1'k_1+1}\delta_{k_2'k_2}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K-1}\delta_{k_1',-k_1-1}\delta_{k_2',-k_2}]\\
&-C_{1-}C_{J-}[\delta_{m'm}\delta_{K',K-1}\delta_{k_1',k_1-1}\delta_{k_2'k_2}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K+1}\delta_{k_1',k_1+1}\delta_{k_2',-k_2}]\\
&-C_{2+}C_{J+}[\delta_{m'm}\delta_{K'K+1}\delta_{k_1'k_1}\delta_{k_2'k_2+1}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K-1}\delta_{k_1',-k_1}\delta_{k_2',-k_2-1}]\\
&-C_{2-}C_{J-}[\delta_{m'm}\delta_{K'K-1}\delta_{k_1'k_1}\delta_{k_2'k_2-1}+(-1)^{J+m+P}\delta_{m',-m}\delta_{K',-K+1}\delta_{k_1,-k_1}\delta_{k_2',-k_2+1}]\Big]
\end{aligned} \tag{27}
$$

$$
\begin{aligned}
= \quad & \frac{\delta_{n'n}\delta_{j_1'j_1}\delta_{j_2'j_2}\delta_{m'm}}{2\mu R_n^2\sqrt{(1+\delta_{K'0}\delta_{k_1'0}\delta_{m'0})(1+\delta_{K0}\delta_{k_10}\delta_{m0})}} \times \\
& \Big[E_{Jj_1j_2}\delta_{K'K}\delta_{k_1'k_1}\delta_{k_2'k_2}(1+\delta_{K0}\delta_{k_10}\delta_{m0}\delta_{k_20}(-1)^{J+P}) \\
& +C_{1+}C_{2-}\delta_{K'K}\delta_{k_1'k_1+1}\delta_{k_2'k_2-1}(1+\delta_{m0}\delta_{K,0}\delta_{k_1+1,0}\delta_{k_2-1,0}(-1)^{J+P}) \\
& +C_{1-}C_{2+}\delta_{K'K}\delta_{k_1'k_1-1}\delta_{k_2'k_2+1}(1+\delta_{m0}\delta_{K,0}\delta_{k_1-1,0}\delta_{k_2+1,0}(-1)^{J+P}) \\
& -C_{1+}C_{J+}\delta_{K'K+1}\delta_{k_1'k_1+1}\delta_{k_2'k_2}(1+\delta_{m0}\delta_{K+1,0}\delta_{k_1+1,0}\delta_{k_2,0}(-1)^{J+P}) \\
& -C_{1-}C_{J-}\delta_{K'K-1}\delta_{k_1'k_1-1}\delta_{k_2'k_2}(1+\delta_{m0}\delta_{K-1,0}\delta_{k_1-1,0}\delta_{k_2,0}(-1)^{J+P}) \\
& -C_{2+}C_{J+}\delta_{K'K+1}\delta_{k_1'k_1}\delta_{k_2'k_2+1}(1+\delta_{m0}\delta_{K+1,0}\delta_{k_10}\delta_{k_2+1,0}(-1)^{J+P}) \\
& -C_{2-}C_{J-}\delta_{K'K-1}\delta_{k_1'k_1}\delta_{k_2'k_2-1}(1+\delta_{m0}\delta_{K-1,0}\delta_{k_10}\delta_{k_2-1,0}(-1)^{J+P})\Big]
\end{aligned} \tag{28}
$$

where $E_{Jj_1j_2} = \big[J(J+1) + j_1(j_1+1) + j_2(j_2+1) + 2k_1k_2 - 2Kk_1 - 2Kk_2\big]$.

C. The third and forth term of matrix elements

After choosing z'-axis of the molecular(CH_3F or H_2)-fixed frame along the principal rotational axis, the hamiltonian of the molecule is given the following form:

$$\hat{H}_{\mathrm{CH_3F}} = Bj_1^2 + (A-B)j_z^2 - D_Jj_1^4 - D_{JK}j_1^2j_z^2 - D_kj_k^4 \tag{29}$$

$$\hat{H}_{\mathrm{H_2}} = Bj_2^2 \tag{30}$$

and,

$$\hat{H}_{\mathrm{CH_3F}}|j_1k_1m\rangle = Bj_1(j_1+1) + (A-B)k_1^2 - D_Jj_1^2(j_1+1)^2 - D_{jk}j_1(j_1+1)k_1^2 - D_kk_1^4|j_1k_1m\rangle$$

$$\hat{H}_{\mathrm{CH_3F}}|j_1,-k_1,-m\rangle = Bj_1(j_1+1) + (A-B)k_1^2 - D_Jj_1^2(j_1+1)^2 - D_{jk}j_1(j_1+1)k_1^2 - D_kk_1^4|j_1,-k_1,-m\rangle$$

$$\hat{H}_{\mathrm{H_2}}|j_2k_2\rangle = B'j_2(j_2+1)|j_2k_2\rangle$$

$$\hat{H}_{\mathrm{H}_2}|j_2,-k_2\rangle = B'j_2(j_2+1)|j_2,-k_2\rangle$$

We set $E_1 = Bj_1(j_1+1)+(A-B)k_1^2-D_Jj_1^2(j_1+1)^2-D_{jk}j_1(j_1+1)k_1^2-D_kk_1^4$ and $E_2 = B'j_2(j_2+1)$. There are no ladder operators to change the basis in the Hamiltonian. The matrix elements are diagonal. So they are given straightforward.

1. use the first basis

$$\begin{aligned}
&\langle JK'M|\langle j_1'k_1'm|j_2'k_2'|\langle\psi_{n'}|(\hat{H}_{\mathrm{CH_3F}}+\hat{H}_{\mathrm{H}_2})|\psi_n\rangle|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\
=\ &(E_1+E_2)\delta_{n'n}\delta_{K'K}\delta_{j_1'j_1}\delta_{k_1'k_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{k_2'k_2}
\end{aligned} \tag{31}$$

2. use the second basis

$$\begin{aligned}
&\langle\Theta^{JMPK'}_{j_1'k_1'm'j_2'k_2'n'}|(\hat{H}_{\mathrm{CH_3F}}+\hat{H}_{\mathrm{H}_2})|\Theta^{JMPK}_{j_1k_1mj_2k_2n}\rangle \\
=\ &\frac{\delta_{n',n}\delta_{j_1'j_1}\delta_{j_2'j_2}(E_1+E_2)}{\sqrt{(1+\delta_{K'0}\delta_{k_1'0}\delta_{m'0})(1+\delta_{K0}\delta_{k_10}\delta_{m0})}}\left[\delta_{KK'}\delta_{k_1'k_1}\delta_{m'm}\delta_{k_2'k_2}+(-1)^{J+m+P}\delta_{K'-K}\delta_{k_1'-k_1}\delta_{m'-m}\delta_{k_2'-k_2}\right] \\
=\ &\frac{E_1+E_2}{1+\delta_{K0}\delta_{k_10}\delta_{m0}}\left[1+(-1)^{J+P}\delta_{K0}\delta_{k_10}\delta_{k_20}\right]\delta_{n'n}\delta_{K'K}\delta_{j_1'j_1}\delta_{k_1'k_1}\delta_{m'm}\delta_{j_2'j_2}\delta_{k_2'k_2}
\end{aligned} \tag{32}$$

D. The Fifth Term of Matrix Elements

1. use the first basis

The potential energy that invariant under the rotation of the body-fixed frame is independent on the α and β. So the matrix elements about K and K' are diagonal.

$$
\begin{aligned}
&\langle JK'M|\langle j_1'k_1'm'|\langle j_2'k_2'|\langle\psi_{n'}|V(R,\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)|\psi_n\rangle|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\
= \; & \delta_{n'n}\delta_{K'K}\langle j_1'k_1'm'|\langle j_2'k_2'|V_{R_n}(\alpha,\beta,\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)|j_2k_2\rangle|j_1k_1m\rangle \\
= \; & \frac{\sqrt{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}}{32\pi^3}\delta_{n'n}\delta_{K'K}\int_0^{2\pi}d\alpha_1\int_0^{\pi}\sin\beta_1 d\beta_1\int_0^{2\pi}d\gamma\int_0^{2\pi}d\alpha_2\int_0^{\pi}\sin\beta_2 d\beta_2 \\
& e^{-ik_1'\alpha_1}d^{j_1'*}_{k_1'm'}(\beta_1)e^{-im'\gamma}e^{-ik_2'\alpha_2}d^{j_2*}_{k_2'0}(\beta_2)V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)e^{ik_1\alpha_1}d^{j_1}_{k_1m}(\beta_1)e^{im\gamma}e^{ik_2\alpha_2}d^{j_2}_{k_20}(\beta_2) \\
= \; & \frac{\sqrt{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}}{32\pi^3}\delta_{n'n}\delta_{K'K}\int_0^{2\pi}d\alpha_1\int_0^{\pi}\sin\beta_1 d\beta_1\int_0^{2\pi}d\gamma\int_0^{2\pi}d\alpha_2\int_0^{\pi}\sin\beta_2 d\beta_2 \\
& V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)d^{j_1'*}_{k_1'm'}(\beta_1)d^{j_2*}_{k_2'0}(\beta_2)d^{j_1}_{k_1m}(\beta_1)d^{j_2}_{k_20}(\beta_2)\exp i\big[(k_1-k_1')\alpha_1+(m-m')\gamma+(k_2-k_2')\alpha_2\big]
\end{aligned}
$$

We use the Gauss quadrature to gain the result of the integration. The basic idea is that for the integral $\int_0^{2\pi} f(\chi)d\chi$ and $\int_0^{\pi} f(\theta)\sin\theta d\theta$ in which $f(\chi)$ and $f(\theta)$ are general function can be changed to a sum of group of grids. A simple deduce given below.

$$
\begin{aligned}
&\int_0^{\pi} d\theta\sin\theta f(\theta) \\
&= -\int_{\theta=0}^{\pi} d(\cos\theta)f[\cos^{-1}(\cos\theta)] \\
&= \int_{x=-1}^{1} dx f(\cos^{-1}x) \\
&\approx \sum_i w_i^{GL} f(\cos^{-1}x_i^{GL})
\end{aligned}
$$

In which $w_i^{GL}=\triangle x=\frac{2}{N_L},\;\; x_i^{GL}=\frac{2n_L}{N_L}-1(n_L=0.1.2\cdots N_L)$.

$$
\begin{aligned}
\int_{\chi=0}^{2\pi} d\chi f(\chi) &= \int_0^{2\pi} d\chi\sin\chi\frac{f(\chi)}{sin\chi} \\
&= \int_{\chi=0}^{\pi} d\chi sin\chi\frac{f(\chi)}{sin\chi}+\int_{\chi=\pi}^{2\pi} sin\chi\frac{f(\chi)}{sin\chi} \\
&= \int_{\chi=0}^{\pi} d(-\cos\chi)\frac{f(\chi)}{\sqrt{1-\cos^2\chi}}+\int_{\chi=\pi}^{2\pi} d(-\cos\chi)\frac{f(\chi)}{-\sqrt{1-\cos^2\chi}} \\
&= \int_{\cos\chi=-1}^{1} d(\cos\chi)\frac{f(\cos^{-1}(\cos\chi))}{\sqrt{1-\cos^2\chi}}+\int_{\cos\chi=-1}^{1} d(\cos\chi)\frac{2\pi-f(\cos^{-1}(\cos\chi))}{\sqrt{1-\cos^2\chi}} \\
&= \int_{x=-1}^{1} dx\frac{\cos^{-1}x}{\sqrt{1-x^2}}+\int_{x=-1}^{1} dx\frac{2\pi-\cos^{-1}x}{\sqrt{1-x^2}} \\
&= \int_{x=-1}^{1} dx\frac{1}{\sqrt{1-x^2}}[f(\cos^{-1}x)+f(2\pi-cos^{-1}x)] \\
&\approx \sum_i w_i^{GC}[f(\cos^{-1}x_i^{GC})+f(2\pi-\cos^{-1}x_i^{GC})]
\end{aligned}
$$

where w_i^{GC} and x_i^{GC} are the Gauss-Chebyshev weights and grids. Obviously, if we only need n operations to obtain the result with $2n$ grids.

Let's switch to the angular function $A_\Lambda(\alpha_1, \beta_1, \gamma, \alpha_2, \beta_2)$ of the potential energy function which reads:

$$
\begin{aligned}
& A_\Lambda(\alpha_1, \beta_1, \gamma, \alpha_2, \beta_2) \\
= & \sum_{\substack{M_1 \\ M_2=-M_1}} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & 0 \end{pmatrix} D^{L_1 *}_{M_1 K_1}(\alpha_1, \beta_1, \gamma) C^{L_2 *}_{M_2}(\alpha_2, \beta_2) \\
= & \sum_{\substack{M_1 \\ M_2=-M_1}} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & 0 \end{pmatrix} d^{L_1 *}_{M_1 K_1}(\alpha_1, \beta_1, \gamma) P^{M_2}_{L_2 *}(\beta_2) \exp -i(M_1\alpha_1 + K_1\gamma + M_2\alpha) \\
= & \sum_{\substack{M_1 \\ M_2=-M_1}} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & 0 \end{pmatrix} d^{L_1 *}_{M_1 K_1}(\alpha_1, \beta_1, \gamma) P^{M_2}_{L_2 *}(\beta_2) \Big[\cos(M_1\alpha_1 + K_1\gamma + M_2\alpha) \\
& -i \sin(M_1\alpha_1 + K_1\gamma + M_2\alpha) \Big]
\end{aligned} \tag{33}
$$

While, cause the potential energy of the system is real, the complex part is truncated in actual potential energy fitting. Let's see what will happen if the three azimuthal angles changes at the same time under some rule.

$$
\begin{aligned}
& A_\Lambda(2\pi - \alpha_1, \beta_1, 2\pi - \gamma, 2\pi - \alpha_2, \beta_2) \\
= & \sum_{\substack{M_1 \\ M_2=-M_1}} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & 0 \end{pmatrix} d^{L_1 *}_{M_1 K_1}(\alpha_1, \beta_1, \gamma) P^{M_2}_{L_2 *}(\beta_2) \exp -i[M_1(2\pi - \alpha_1) + K_1(2\pi - \gamma) + M_2(2\pi - \alpha)] \\
= & \sum_{\substack{M_1 \\ M_2=-M_1}} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & 0 \end{pmatrix} d^{L_1 *}_{M_1 K_1}(\alpha_1, \beta_1, \gamma) P^{M_2}_{L_2 *}(\beta_2) \Big[\cos[M_1(2\pi - \alpha_1) + K_1(2\pi - \gamma) + M_2(2\pi - \alpha)] \\
& -i \sin[M_1(2\pi - \alpha_1) + K_1(2\pi - \gamma) + M_2(2\pi - \alpha)] \Big] \\
= & \sum_{\substack{M_1 \\ M_2=-M_1}} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & 0 \end{pmatrix} d^{L_1 *}_{M_1 K_1}(\alpha_1, \beta_1, \gamma) P^{M_2}_{L_2 *}(\beta_2) \Big[\cos(M_1\alpha_1 + K_1\gamma + M_2\alpha) \\
& +i \sin(M_1\alpha_1 + K_1\gamma + M_2\alpha) \Big]
\end{aligned} \tag{34}
$$

(33). and (34). are equal, if the complex part are truncated as we did in PES fitting. So we can give these equations:

$$V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) = V_{R_n}[(2\pi-\alpha_1),\beta_1,(2\pi-\gamma),(2\pi-\alpha_2),\beta_2] \tag{35}$$

$$V_{R_n}[\alpha_1,\beta_1,\gamma,(2\pi-\alpha_2),\beta_2] = V_{R_n}[(2\pi-\alpha_1),\beta_1,(2\pi-\gamma),\alpha_2,\beta_2] \tag{36}$$

$$V_{R_n}[\alpha_1,\beta_1,(2\pi-\gamma),\alpha_2,\beta_2] = V_{R_n}[(2\pi-\alpha_1),\beta_1,\gamma,(2\pi-\alpha_2),\beta_2] \tag{37}$$

$$V_{R_n}[\alpha_1,\beta_1,(2\pi-\gamma),(2\pi-\alpha_2),\beta_2] = V_{R_n}[(2\pi-\alpha_1),\beta_1,\gamma,\alpha_2,\beta_2] \tag{38}$$

Let's go back to the integral in (33), and set $f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)$ $= V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) d^{j_1'*}_{k_1'm'}(\beta_1) d^{j_2*}_{k_2'0}(\beta_2) d^{j_1}_{k_1m}(\beta_1) d^{j_2}_{k_20}(\beta_2) \exp i\big[(k_1-k_1')\alpha_1+(m-m')\gamma+(k_2-k_2')\alpha_2\big]$

$$\begin{aligned}
&\int_0^{2\pi} d\alpha_1 \int_0^{2\pi} d\gamma \int_0^{2\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&d^{j_1'*}_{k_1'm'}(\beta_1) d^{j_2*}_{k_2'0}(\beta_2) d^{j_1}_{k_1m}(\beta_1) d^{j_2}_{k_20}(\beta_2) \exp i\big[(k_1-k_1')\alpha_1+(m-m')\gamma+(k_2-k_2')\alpha_2\big] \\
=\;& \int_0^{2\pi} d\alpha_1 \int_0^{2\pi} d\gamma \int_0^{2\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
=\;& \int_0^{\pi} d\alpha_1 \int_0^{\pi} d\gamma \int_0^{\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_0^{\pi} d\alpha_1 \int_0^{\pi} d\gamma \int_{\pi}^{2\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_0^{\pi} d\alpha_1 \int_{\pi}^{2\pi} d\gamma \int_0^{\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_0^{\pi} d\alpha_1 \int_{\pi}^{2\pi} d\gamma \int_{\pi}^{2\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_{\pi}^{2\pi} d\alpha_1 \int_{\pi}^{2\pi} d\gamma \int_{\pi}^{2\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_{\pi}^{2\pi} d\alpha_1 \int_{\pi}^{2\pi} d\gamma \int_0^{\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_{\pi}^{2\pi} d\alpha_1 \int_0^{\pi} d\gamma \int_{\pi}^{2\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
&+\int_{\pi}^{2\pi} d\alpha_1 \int_0^{\pi} d\gamma \int_0^{\pi} d\alpha_2 \int_0^{\pi} \sin\beta_1 d\beta_1 \int_0^{\pi} \sin\beta_2 d\beta_2 \; f(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2) \\
\approx\;& \sum_{i1} w^{GC}_{i1} \sum_{i2} w^{GC}_{i2} \sum_{i3} w^{GC}_{i3} \sum_{l1} w^{GL}_{l1} \sum_{l2} w^{GL}_{l2} \times \\
&\Big[f(\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2}) + f(2\pi-\alpha^{GC}_{i1},\beta^{GL}_{l1},2\pi-\gamma^{GC}_{i3},2\pi-\alpha^{GC}_{i2},\beta^{GL}_{l2}) \\
&+f(\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},2\pi-\alpha^{GC}_{i2},\beta^{GL}_{l2}) + f(2\pi-\alpha^{GC}_{i1},\beta^{GL}_{l1},2\pi-\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2}) \\
&+f(\alpha^{GC}_{i1},\beta^{GL}_{l1},2\pi-\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2}) + f(2\pi-\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},2\pi-\alpha^{GC}_{i2},\beta^{GL}_{l2}) \\
&+f(\alpha^{GC}_{i1},\beta^{GL}_{l1},2\pi-\gamma^{GC}_{i3},2\pi-\alpha^{GC}_{i2},\beta^{GL}_{l2}) + f(2\pi-\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2})\Big]
\end{aligned} \tag{39}$$

in which $\alpha_i^{GC} = \cos^{-1} x_i^{GC}$, $\beta_j^{GL} = \cos^{-1} x_j^{GL}$, $\gamma_i^{GC} = \cos^{-1} x_i^{GC}$. For the first two terms in the bracket,

$$\begin{aligned}
& f(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) + f(2\pi - \alpha_{i1}^{GC}, \beta_{l1}^{GL}, 2\pi - \gamma_{i3}^{GC}, 2\pi - \alpha_{i2}^{GC}, \beta_{l2}^{GL}) \\
= \; & V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL}) \\
& \exp i[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')\alpha_{i2}^{GC}] \\
+ \; & V_{R_n}(2\pi - \alpha_{i1}^{GC}, \beta_{l1}^{GL}, 2\pi - \gamma_{i3}^{GC}, 2\pi - \alpha_{i2}^{GC}, \beta_{l2}^{GL}) d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL}) \\
& \exp i[(k_1 - k_1')(2\pi - \alpha_{i1}^{GC}) + (m - m')(2\pi - \gamma_{i3}^{GC}) + (k_2 - k_2')(2\pi - \alpha_{i2}^{GC})] \\
= \; & V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL}) \\
& \exp i[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')\alpha_{i2}^{GC}] \\
+ \; & V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL}) \\
& \exp -i[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')\alpha_{i2}^{GC}] \\
= \; & 2V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL}) \\
& \cos[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')\alpha_{i2}^{GC}]
\end{aligned} \tag{40}$$

Same theatment can be used to other six terms in the brackets. At last, the matrix elements reads:

$$\begin{aligned}
& \frac{\sqrt{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}}{16\pi^3} \delta_{n'n}\delta_{K'K} \times \\
& \sum_{i1} w_{i1}^{GC} \sum_{i2} w_{i2}^{GC} \sum_{i3} w_{i3}^{GC} \sum_{l1} w_{l1}^{GL} \sum_{l2} w_{l2}^{GL} d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL}) \times \\
& \Big[V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) \cos[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')\alpha_{i2}^{GC}] \\
+ \; & V_{R_n}(2\pi - \alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) \cos[(k_1 - k_1')(2\pi - \alpha_{i1}^{GC}) + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')\alpha_{i2}^{GC}] \\
+ \; & V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, 2\pi - \gamma_{i3}^{GC}, \alpha_{i2}^{GC}, \beta_{l2}^{GL}) \cos[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')(2\pi - \gamma_{i3}^{GC}) + (k_2 - k_2')\alpha_{i2}^{GC}] \\
+ \; & V_{R_n}(\alpha_{i1}^{GC}, \beta_{l1}^{GL}, \gamma_{i3}^{GC}, 2\pi - \alpha_{i2}^{GC}, \beta_{l2}^{GL}) \cos[(k_1 - k_1')\alpha_{i1}^{GC} + (m - m')\gamma_{i3}^{GC} + (k_2 - k_2')(2\pi - \alpha_{i2}^{GC})] \Big]
\end{aligned} \tag{41}$$

2. use the second basis

$$
\begin{aligned}
&\langle\Theta^{JMPK'}_{j_1'k_1'm'j_2'k_2'n'}|V(R,\alpha_1,\beta_1\gamma,\alpha_2,\beta_2)|\Theta^{JMPK}_{j_1k_1mj_2k_2n}\rangle \\
= & \frac{\delta_{n'n}\delta_{K'K}}{2\sqrt{(1+\delta_{k_1',0}\delta_{m',0}\delta_{K',0})(1+\delta_{k_1,0}\delta_{m,0}\delta_{K,0})}}\Big[\langle j_1'k_1'm'|\langle j_2'k_2'|+(-1)^{J+m'+P}\langle j_1'-k_1'-m'|\langle j_2'-k_2'|\Big] \\
& V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)\Big[|j_1k_1m\rangle|j_2k_2\rangle+(-1)^{J+m+P}|j-k_1m\rangle|j_2-k_2\rangle\Big] \\
= & \frac{\delta_{n'n}\delta_{K'K}}{2\sqrt{(1+\delta_{k_1',0}\delta_{m',0}\delta_{K',0})(1+\delta_{k_1,0}\delta_{m,0}\delta_{K,0})}}\Big[\langle j_1'k_1'm'|\langle j_2'k_2'|V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)|j_2k_2\rangle|j_1k_1m\rangle \\
& +(-1)^{J+m+P}\delta_{K,0}\langle j_1'k_1'm'|\langle j_2'k_2'|V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)|j_2-k_2\rangle|j_1-k_1-m\rangle \\
& +(-1)^{J+m'+P}\delta_{K,0}\langle j_1'-k_1'-m'|\langle j_2'-k_2'|V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)|j_2k_2\rangle|j_1k_1m\rangle \\
& +(-1)^{m'+m}\langle j_1'-k_1'-m'|\langle j_2'-k_2'|V_{R_n}(\alpha_1,\beta_1,\gamma,\alpha_2,\beta_2)|j_2-k_2\rangle|j_1-k_1-m\rangle\Big]
\end{aligned} \tag{42}
$$

The appearance of first term in bracket is same as that when we use the first basis. We have get the results before. The way we calculate other three terms is like we the way did in the first basis. So I will't show the detail more and given the results straightforward.

for the second term in bracket,

$$
\begin{aligned}
& \frac{\sqrt{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}}{16\pi^3}(-1)^{J+P+k_1+k_2}\delta_{K,0}\times \\
& \sum_{i1}w^{GC}_{i1}\sum_{i2}w^{GC}_{i2}\sum_{i3}w^{GC}_{i3}\sum_{l1}w^{GL}_{l1}\sum_{l2}w^{GL}_{l2}d^{j_1'*}_{k_1'm'}(\beta^{GL}_{l1})d^{j_2*}_{k_2'0}(\beta^{GL}_{l2})d^{j_1}_{k_1m}(\beta^{GL}_{l1})d^{j_2}_{k_20}(\beta^{GL}_{l2})\times \\
& \Big[V_{R_n}(\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2})\cos\left[(k_1+k_1')\alpha^{GC}_{i1}+(m+m')\gamma^{GC}_{i3}+(k_2+k_2')\alpha^{GC}_{i2}\right] \\
+ & V_{R_n}(2\pi-\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2})\cos\left[(k_1+k_1')(2\pi-\alpha^{GC}_{i1})+(m+m')\gamma^{GC}_{i3}+(k_2+k_2')\alpha^{GC}_{i2}\right] \\
+ & V_{R_n}(\alpha^{GC}_{i1},\beta^{GL}_{l1},2\pi-\gamma^{GC}_{i3},\alpha^{GC}_{i2},\beta^{GL}_{l2})\cos\left[(k_1+k_1')\alpha^{GC}_{i1}+(m+m')(2\pi-\gamma^{GC}_{i3})+(k_2+k_2')\alpha^{GC}_{i2}\right] \\
+ & V_{R_n}(\alpha^{GC}_{i1},\beta^{GL}_{l1},\gamma^{GC}_{i3},2\pi-\alpha^{GC}_{i2},\beta^{GL}_{l2})\cos\left[(k_1+k_1')\alpha^{GC}_{i1}+(m+m')\gamma^{GC}_{i3}+(k_2+k_2')(2\pi-\alpha^{GC}_{i2})\right]\Big]
\end{aligned} \tag{43}
$$

for the third term in bracket,

$$
\frac{\sqrt{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}}{16\pi^3}(-1)^{J+P+k_1'+k_2'}\delta_{K',0}\times
$$

$$
\sum_{i1} w_{i1}^{GC}\sum_{i2} w_{i2}^{GC}\sum_{i3} w_{i3}^{GC}\sum_{l1} w_{l1}^{GL}\sum_{l2} w_{l2}^{GL} d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL})\times
$$

$$
\Big[V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1+k_1')\alpha_{i1}^{GC}+(m+m')\gamma_{i3}^{GC}+(k_2+k_2')\alpha_{i2}^{GC}\right] \tag{44}
$$

$$
+\ V_{R_n}(2\pi-\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1+k_1')(2\pi-\alpha_{i1}^{GC})+(m+m')\gamma_{i3}^{GC}+(k_2+k_2')\alpha_{i2}^{GC}\right]
$$

$$
+\ V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},2\pi-\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1+k_1')\alpha_{i1}^{GC}+(m+m')(2\pi-\gamma_{i3}^{GC})+(k_2+k_2')\alpha_{i2}^{GC}\right]
$$

$$
+\ V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},2\pi-\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1+k_1')\alpha_{i1}^{GC}+(m+m')\gamma_{i3}^{GC}+(k_2+k_2')(2\pi-\alpha_{i2}^{GC})\right]\Big]
$$

for the forth term in bracket

$$
\frac{\sqrt{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}}{16\pi^3}(-1)^{m'+m}\times
$$

$$
\sum_{i1} w_{i1}^{GC}\sum_{i2} w_{i2}^{GC}\sum_{i3} w_{i3}^{GC}\sum_{l1} w_{l1}^{GL}\sum_{l2} w_{l2}^{GL} d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL})\times
$$

$$
\Big[V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1-k_1')\alpha_{i1}^{GC}+(m-m')\gamma_{i3}^{GC}+(k_2-k_2')\alpha_{i2}^{GC}\right] \tag{45}
$$

$$
+\ V_{R_n}(2\pi-\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1-k_1')(2\pi-\alpha_{i1}^{GC})+(m-m')\gamma_{i3}^{GC}+(k_2-k_2')\alpha_{i2}^{GC}\right]
$$

$$
+\ V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},2\pi-\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1-k_1')\alpha_{i1}^{GC}+(m-m')(2\pi-\gamma_{i3}^{GC})+(k_2-k_2')\alpha_{i2}^{GC}\right]
$$

$$
+\ V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},2\pi-\alpha_{i2}^{GC},\beta_{l2}^{GL})\cos\left[(k_1-k_1')\alpha_{i1}^{GC}+(m-m')\gamma_{i3}^{GC}+(k_2-k_2')(2\pi-\alpha_{i2}^{GC})\right]\Big]
$$

Sum the four term and put them back in the equation (43)., we can get

$$
\frac{\delta_{n'n}\delta_{K'K}}{16\pi^3}\sqrt{\frac{(2j_1'+1)(2j_2'+1)(2j_1+1)(2j_2+1)}{(1+\delta_{k_1'0}\delta_{m'0}\delta_{K'0})(1+\delta_{k_10}\delta_{m0}\delta_{K0})}} \tag{46}
$$

$$
\times\ \sum_{i1} w_{i1}^{GC}\sum_{i2} w_{i2}^{GC}\sum_{i3} w_{i3}^{GC}\sum_{l1} w_{l1}^{GL}\sum_{l2} w_{l2}^{GL} d_{k_1'm'}^{j_1'*}(\beta_{l1}^{GL}) d_{k_2'0}^{j_2*}(\beta_{l2}^{GL}) d_{k_1m}^{j_1}(\beta_{l1}^{GL}) d_{k_20}^{j_2}(\beta_{l2}^{GL})
$$

$$
\times\ \Big\{V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\Big[A\cos\left[(k_1-k_1')\alpha_{i1}^{GC}+(m-m')\gamma_{i3}^{GC}+(k_2-k_2')\alpha_{i2}^{GC}\right]
$$

$$
+(-1)^{J+P}\delta_{K0}\cos\left[(k_1+k_1')\alpha_{i1}^{GC}+(m+m')\gamma_{i3}^{GC}+(k_2+k_2')\alpha_{i2}^{GC}\right]\Big]
$$

$$
+\ V_{R_n}(2\pi-\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\Big[A\cos\left[(k_1-k_1')(2\pi-\alpha_{i1}^{GC})+(m-m')\gamma_{i3}^{GC}+(k_2-k_2')\alpha_{i2}^{GC}\right]
$$

$$
+(-1)^{J+P}\delta_{K0}\cos\left[(k_1+k_1')(2\pi-\alpha_{i1}^{GC})+(m+m')\gamma_{i3}^{GC}+(k_2+k_2')\alpha_{i2}^{GC}\right]\Big]
$$

$$
+\ V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},2\pi-\gamma_{i3}^{GC},\alpha_{i2}^{GC},\beta_{l2}^{GL})\Big[A\cos\left[(k_1-k_1')\alpha_{i1}^{GC}+(m-m')(2\pi-\gamma_{i3}^{GC})+(k_2-k_2')\alpha_{i2}^{GC}\right]
$$

$$
+(-1)^{J+P}\delta_{K0}\cos\left[(k_1+k_1')\alpha_{i1}^{GC}+(m+m')(2\pi-\gamma_{i3}^{GC})+(k_2+k_2')\alpha_{i2}^{GC}\right]\Big]
$$

$$
+\ V_{R_n}(\alpha_{i1}^{GC},\beta_{l1}^{GL},\gamma_{i3}^{GC},2\pi-\alpha_{i2}^{GC},\beta_{l2}^{GL})\Big[A\cos\left[(k_1-k_1')\alpha_{i1}^{GC}+(m-m')\gamma_{i3}^{GC}+(k_2-k_2')(2\pi-\alpha_{i2}^{GC})\right]
$$

$$
+(-1)^{J+P}\delta_{K0}\cos\left[(k_1+k_1')\alpha_{i1}^{GC}+(m+m')\gamma_{i3}^{GC}+(k_2+k_2')(2\pi-\alpha_{i2}^{GC})\right]\Big]\Big\}
$$

where $A = 1 + (-1)^{m+m'}$.

E. Dipole matrix elements

The transition line strength is defined as:

$$S_{ii'} = 3 \sum_{M,M'} |\langle \Phi_{i'} | \mu | \Phi_i \rangle|^2 \tag{47}$$

Where Φ_i is the wave function of the lower line, $\Phi_{i'}$ is the wave function of the upper line. Because H_2 molecule has no contribution to the dipole moment. The idea, the derivation and the matrix elements of this system are same as them of CH_3F-He system. I will not repeat them and give the results directly.

1. *use the first basis*

$$\mu_0^{\mathrm{SF}} = \mu_{\mathrm{CH_3F}} \sum_{\sigma=-1}^{-1} D^1_{0,\sigma}(\alpha,\beta,0)^* D^1_{\sigma,0}(\alpha_1,\beta_1,\gamma)^* \tag{48}$$

$$\begin{aligned}
&\langle J'K'M'|\langle j_1'k_1'm|\langle j_2'k_2'|\langle \psi_{n'}|\mu_{\mathrm{CH_3F}} \sum_{\sigma=-1}^{-1} D^1_{0,\sigma}(\alpha,\beta,0)^* D^1_{\sigma,0}(\alpha_1,\beta_1,\gamma)^* |\psi_n\rangle|j_2k_2\rangle|j_1k_1m\rangle|JKM\rangle \\
= \;& \mu_{\mathrm{CH_3F}} \delta_{n',n}\delta_{j_2'j_2}\delta_{k_2'k_2} \sum_{\sigma=-1}^{1} \langle JK'M'|D^1_{0,\sigma}(\alpha,\beta,0)^*|JKM\rangle \langle j_1'k_1'm'|D^1_{\sigma,0}(\alpha_1,\beta_1,\gamma)^*|j_1k_1m\rangle \\
= \;& \mu_{\mathrm{CH_3F}} \delta_{n',n}\delta_{j_2'j_2}\delta_{k_2'k_2} \sum_{\sigma=-1}^{1} (-1)^{k_1'+k_1}\sqrt{2j_1'+1}\sqrt{2j_1+1} \begin{pmatrix} j_1' & 1 & j_1 \\ -m' & 0 & m \end{pmatrix} \begin{pmatrix} j_1' & 1 & j_1 \\ -k_1' & \sigma & k_1 \end{pmatrix} \\
& \times (-1)^{K'+M'}\sqrt{2J+1}\sqrt{2J'+1} \begin{pmatrix} J' & 1 & J \\ -K' & \sigma & K \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ -M' & 0 & M \end{pmatrix} \\
= \;& \mu_{\mathrm{CH_3F}} \delta_{n',n}\delta_{j_2'j_2}\delta_{k_2'k_2} (-1)^{K'+M'+k_1'+k_1}\sqrt{2j_1+1}\sqrt{2j_1'+1}\sqrt{2J+1}\sqrt{2J'+1} \\
& \sum_{\sigma=-1}^{1} \begin{pmatrix} j_1' & 1 & j_1 \\ -m' & 0 & m \end{pmatrix} \begin{pmatrix} j_1' & 1 & j_1 \\ -k_1' & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ -K' & \sigma & K \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ -M' & 0 & M \end{pmatrix} \\
= \;& \mu_{\mathrm{CH_3F}} (-1)^{M'} \begin{pmatrix} J' & 1 & J \\ -M' & 0 & M \end{pmatrix} A_{JPE}
\end{aligned}$$

In which,

$$A_{JPE} = \delta_{n',n}\delta_{j_2'j_2}\delta_{k_2'k_2}(-1)^{K'+M'+k_1'+k_1}[j_1][j_1'][J][J'] \sum_{\sigma=-1}^{1} \begin{pmatrix} j_1' & 1 & j_1 \\ -m & 0 & m \end{pmatrix} \begin{pmatrix} j_1' & 1 & j_1 \\ -k_1' & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ -K' & \sigma & K \end{pmatrix} \tag{49}$$

and $[X] = \sqrt{X+1}$, X for j_1, j_1', J and $J^{'}$.

So Eq. (47) becomes

$$S_{ii'} = 3 \sum_{M=-J_{\min}}^{J_{\min}} |\mu_{\mathrm{CH_3F}}(-1)^{M} \begin{pmatrix} J^{'} & 1 & J \\ -M^{'} & 0 & M \end{pmatrix} \sum_{j_1'k_1'mj_2'k_2'K'} \sum_{j_1k_1mj_2k_2K} C^{J'K'M'P'i^{'}}_{j_1'k_1'm'j_2'k_2'n'} C^{JKMPi}_{j_1k_1mj_2k_2n} A_{JPE}|^2 \tag{50}$$

where J_{min} = min(J$^{'}$, J).

2. use the second basis

$$\langle\Theta^{J'K'M'P'}_{j_1'k_1'm'j_2'k_2'}|\langle R_{n'}|\mu_Z^{SFF}|R_n\rangle|\Theta^{JKMP}_{j_1k_1mj_2k_2}\rangle = \frac{\mu_{CH_3F}\delta_{n',n}\delta_{j_2'j_2}\delta_{k_2'k_2}}{2\sqrt{(1+\delta_{K^{'},0}\delta_{k_1^{'},0}\delta_{m'0})(1+\delta_{K,0})\delta_{k_1,0}\delta_{m0}}}$$
$$(-1)^{K^{'}+M'+k_1'+k_1}[j_1][j_1^{'}][J][J^{'}] \begin{pmatrix} J^{'} & 1 & J \\ -M & 0 & M \end{pmatrix} \sum_{\sigma=-1}^{1} \Bigg\{ \begin{pmatrix} j^{'} & 1 & j \\ -m^{'} & 0 & m \end{pmatrix} \begin{pmatrix} j^{'} & 1 & j \\ -k_1^{'} & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J^{'} & 1 & J \\ -K^{'} & \sigma & K \end{pmatrix}$$
$$+ (-1)^{J+P+k_1} \begin{pmatrix} j_1^{'} & 1 & j_1 \\ -m & 0 & -m \end{pmatrix} \begin{pmatrix} j_1^{'} & 1 & j_1 \\ -k_1^{'} & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J^{'} & 1 & J \\ -K^{'} & \sigma & -K \end{pmatrix}$$
$$+ (-1)^{J^{'}+P^{'}+k_1^{'}} \begin{pmatrix} j_1^{'} & 1 & j_1 \\ m & 0 & m \end{pmatrix} \begin{pmatrix} j_1^{'} & 1 & j_1 \\ k_1 & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J^{'} & 1 & J \\ K^{'} & \sigma & K \end{pmatrix}$$
$$+ (-1)^{J+J^{'}+P+P^{'}+k+k^{'}} \begin{pmatrix} j_1^{'} & 1 & j_1 \\ m' & 0 & -m \end{pmatrix} \begin{pmatrix} j_1^{'} & 1 & j_1 \\ k_1 & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J^{'} & 1 & J \\ K^{'} & \sigma & -K \end{pmatrix} \Bigg\}$$
$$= \mu_{\mathrm{CH_3F}}(-1)^{M'} \begin{pmatrix} J^{'} & 1 & J \\ -M^{'} & 0 & M \end{pmatrix} A_{JPE} \tag{51}$$

the definition of A_{JPA} is

$$A_{JPA} = \frac{\delta_{n',n}\delta_{j_2'j_2}\delta_{k_2'k_2}}{2\sqrt{(1+\delta_{K',0}\delta_{k_1',0}\delta_{m'0})(1+\delta_{K,0})\delta_{k_1,0}\delta_{m0}}}$$
$$(-1)^{K'+k_1'+k_1}[j_1][j_1'][J][J'] \sum_{\sigma=-1}^{1} \Bigg\{ \begin{pmatrix} j' & 1 & j \\ -m' & 0 & m \end{pmatrix} \begin{pmatrix} j' & 1 & j \\ -k_1' & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ -K' & \sigma & K \end{pmatrix}$$
$$+ (-1)^{J+P+k_1} \begin{pmatrix} j_1' & 1 & j_1 \\ -m & 0 & -m \end{pmatrix} \begin{pmatrix} j_1' & 1 & j_1 \\ -k_1' & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ -K' & \sigma & -K \end{pmatrix} \tag{52}$$
$$+ (-1)^{J'+P'+k_1'} \begin{pmatrix} j_1' & 1 & j_1 \\ m & 0 & m \end{pmatrix} \begin{pmatrix} j_1' & 1 & j_1 \\ k_1 & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ K' & \sigma & K \end{pmatrix}$$
$$+ (-1)^{J+J'+P+P'+k+k'} \begin{pmatrix} j_1' & 1 & j_1 \\ m' & 0 & -m \end{pmatrix} \begin{pmatrix} j_1' & 1 & j_1 \\ k_1 & \sigma & k_1 \end{pmatrix} \begin{pmatrix} J' & 1 & J \\ K' & \sigma & -K \end{pmatrix} \Bigg\}$$

So Eq. (47) becomes

$$S_{ii'} = 3 \sum_{M=-J_{\min}}^{J_{\min}} |\mu_{\mathrm{CH_3F}}(-1)^M \begin{pmatrix} J' & 1 & J \\ -M' & 0 & M \end{pmatrix} \sum_{j_1'k_1'mj_2'k_2'K'} \sum_{j_1k_1mj_2k_2K} C^{J'K'M'P'i'}_{j_1'k_1'm'j_2'k_2'n'} C^{JKMPi}_{j_1k_1mj_2k_2n} A_{JPE}|^2 \tag{53}$$

where $J_{min} = \min(\mathrm{J}', \mathrm{J})$.