Simulating reaction of Ne* + OCS collision

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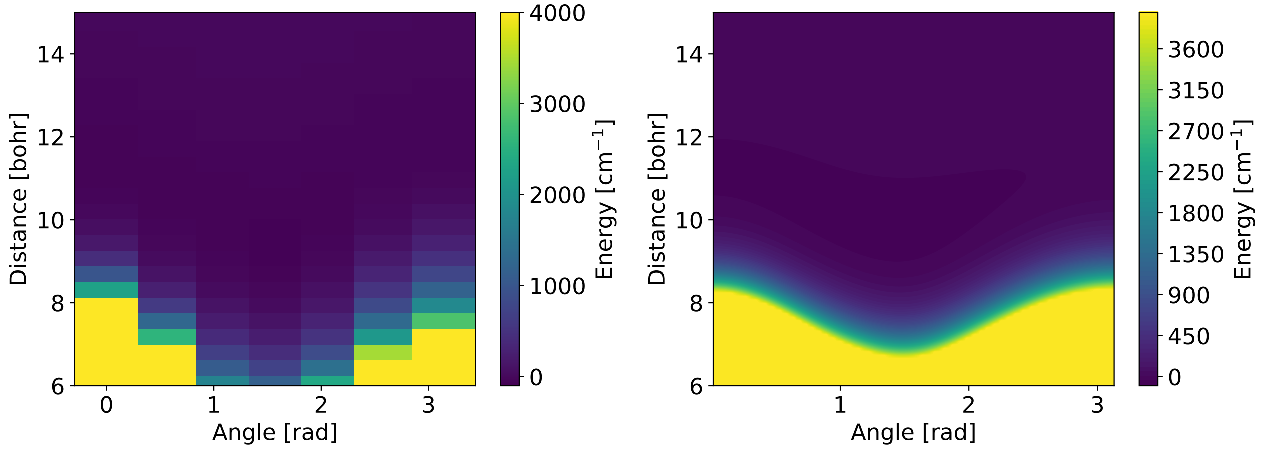
1 Improved potential interpolations

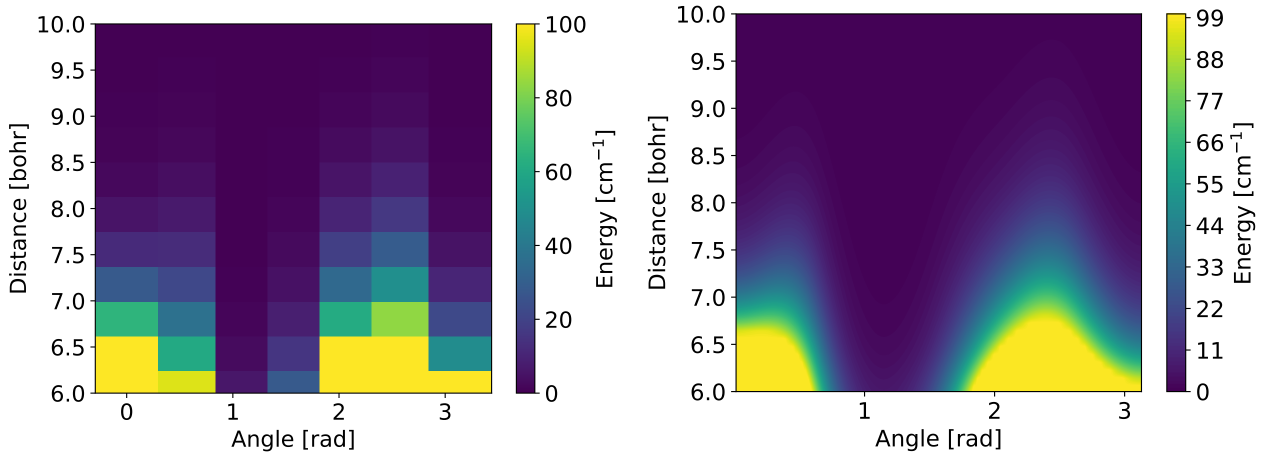
Potential is now fitted to the force field instead of interpolation, because of smoothness issues. Gamma potentials now have correct vanishing derivatives for angles 0 and π.



(a) Data

(b) Fitted

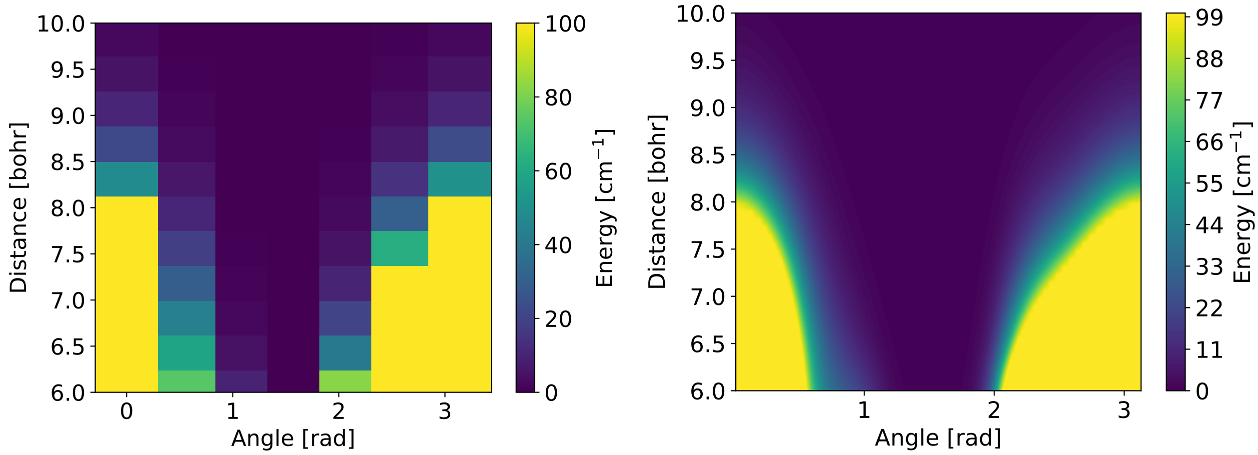
Figure 1: Fitting of intermolecular potential



(a) Data

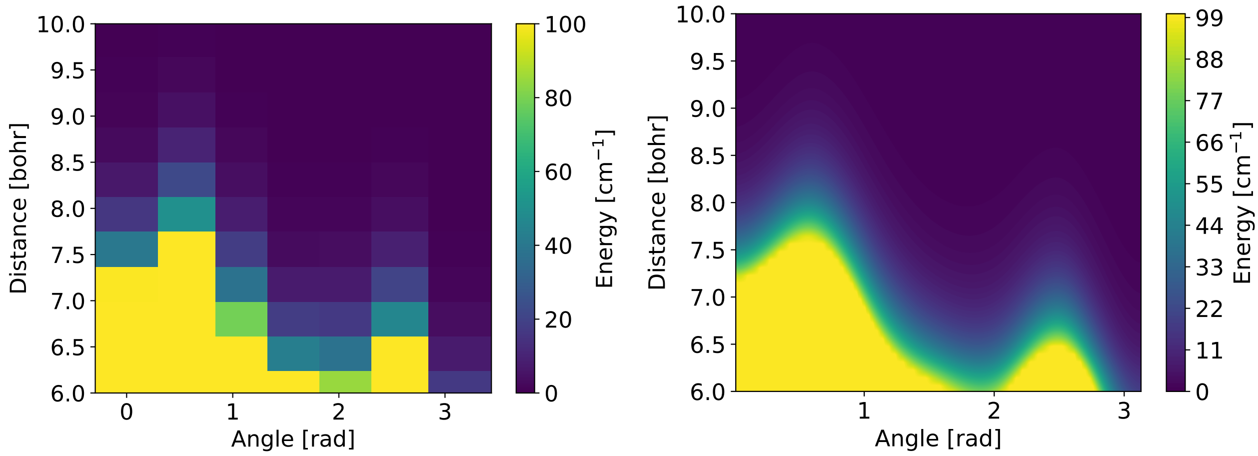
(b) Interpolated

Figure 2: Interpolation of $X\Pi$ gamma potential



(a) Data (b) Interpolated

Figure 3: Interpolation of $B\Sigma$ gamma potential

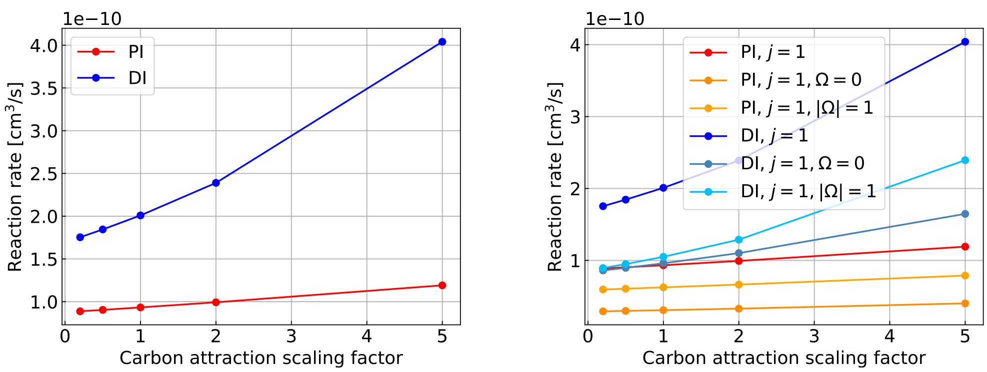


(a) Data (b) Interpolated

Figure 4: Interpolation of $A\Pi$ gamma potential

2 Reaction rate ratio dependence on scaling force field parameters

Scaling the parameters of the force field (fitted as well as with theoretical values) did not change significantly the reaction rate ratio between different initial j from value of 1. As an example we show carbon attraction strength scaling of fitted force field.



(a) Reaction rate for $j = 0$. (b) Reaction rate for $j = 1$.

Figure 5: Calculated reaction rates dependence on carbon attraction strength.

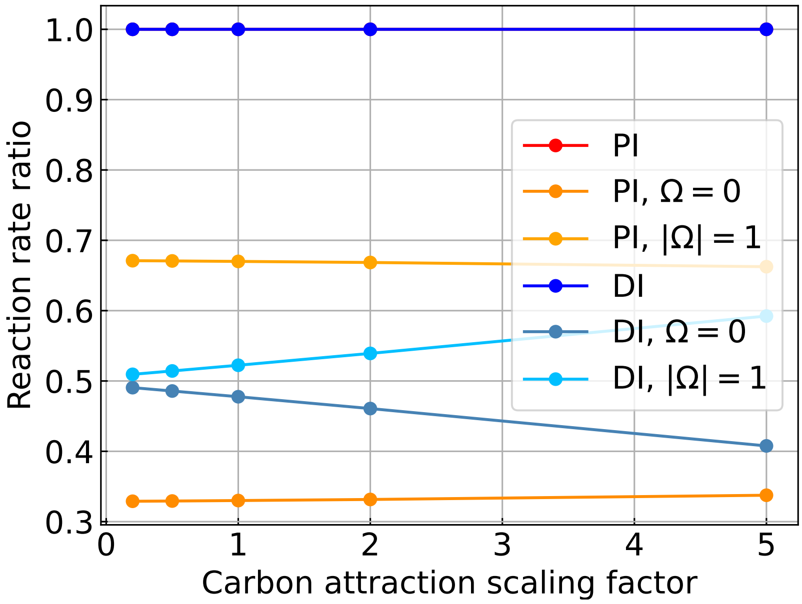


Figure 6: Calculated reaction rate ratio between $j = 1$ and $j = 0$, dependence on carbon attraction strength.

3 Truth tests for coriolis effect inclusion

To test coriolis effect inclusion, we compare the ground state energy of the modified harmonic oscillator using different reference frames.

The system defined by the space-fixed Hamiltonian is

$$\hat{H} = -\frac{1}{2\mu}\frac{\partial^2}{\partial R^2} + \frac{\hat{L}^2}{2\mu R^2} + V(R, \theta), \tag{1}$$

this frame is equivalent to the body-fixed frame that we use for calculations, with rotational constant $B = 0$, the Hamiltonian is

$$\hat{H} = -\frac{1}{2\mu}\frac{\partial^2}{\partial R^2} + \frac{(\hat{J} - \hat{j})^2}{2\mu R^2} + V(R, \theta). \tag{2}$$

The conserved quantities for the first system is the angular momentum projection number m_l, in case of the body-fixed frame the conserved quantity is the total angular momentum J.

We use following harmonic oscillator potential

$$V(r, \theta) = \frac{\mu\omega^2}{2}(r - r_0)^2. \tag{3}$$

The calculated ground state energy of the space-fixed Hamiltonian is equal to ω for projection $m_l = 0$. For the body-fixed Hamiltonian the calculated ground state for $J = 2$ is also ω with coriolis effect and 1.08ω if we neglect the coriolis effect and set $\Omega_{\text{init}} = 0$. Calculated wave-functions of the calculated ground state with and without coriolis effect are shown below.

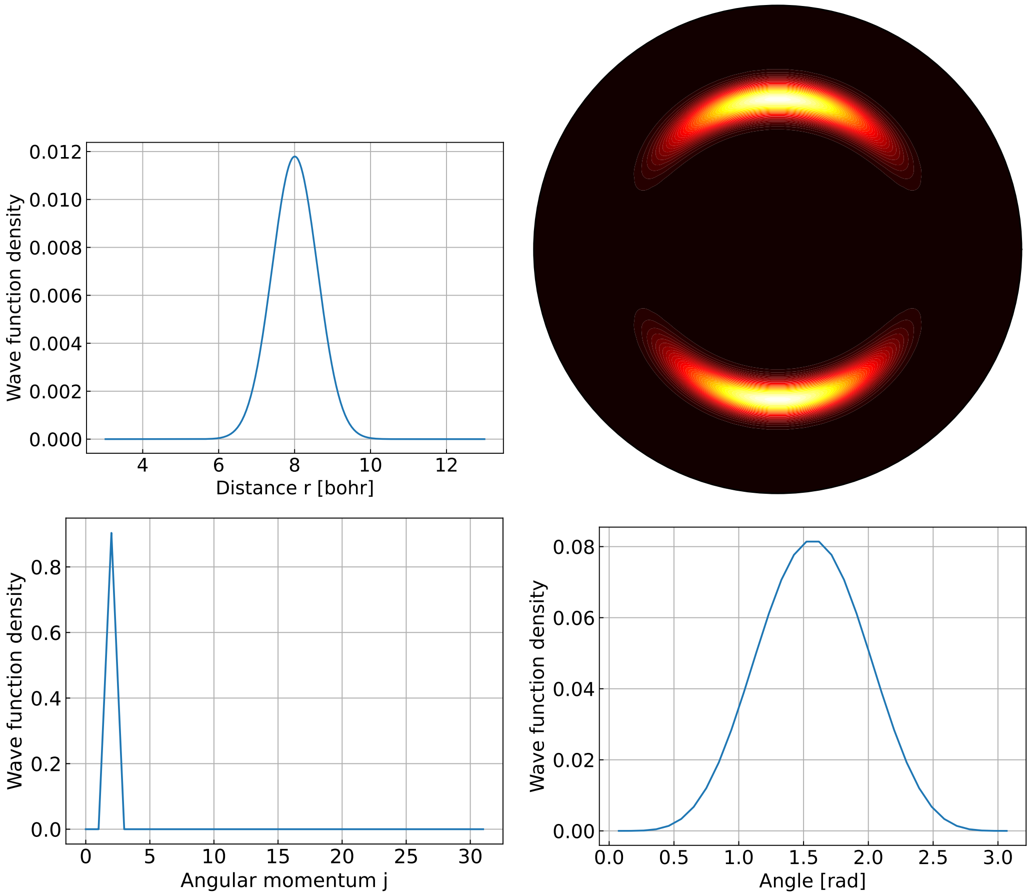


Figure 7: Wave function densities for calculated ground state with $J = 2$ and without coriolis effect.

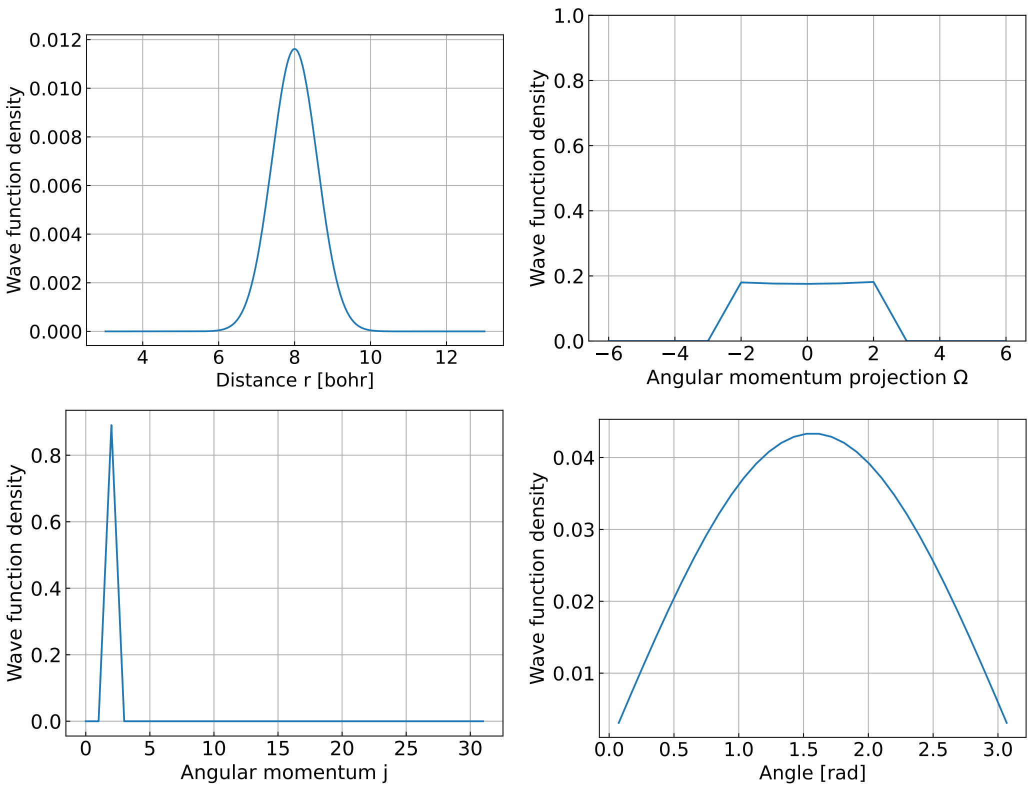


Figure 8: Wave function densities for calculated ground state with $J = 2$ and coriolis effect.

For the potential of the form $V(r, \theta, \phi)$ the space-fixed Hamiltonian doesn't have any conserved angular numbers, however in the body-fixed frame we have still J conserved. It can be deduced that in the case of $B = 0$, J gives additional infinite degeneracy. Because of that, for every conserved J, the ground state is ω, however it is only achievable with the inclusion of the coriolis effect.

Additionally, we test the direct effect of the coriolis effect by multiplying the coriolis term by a factor, results are shown below.

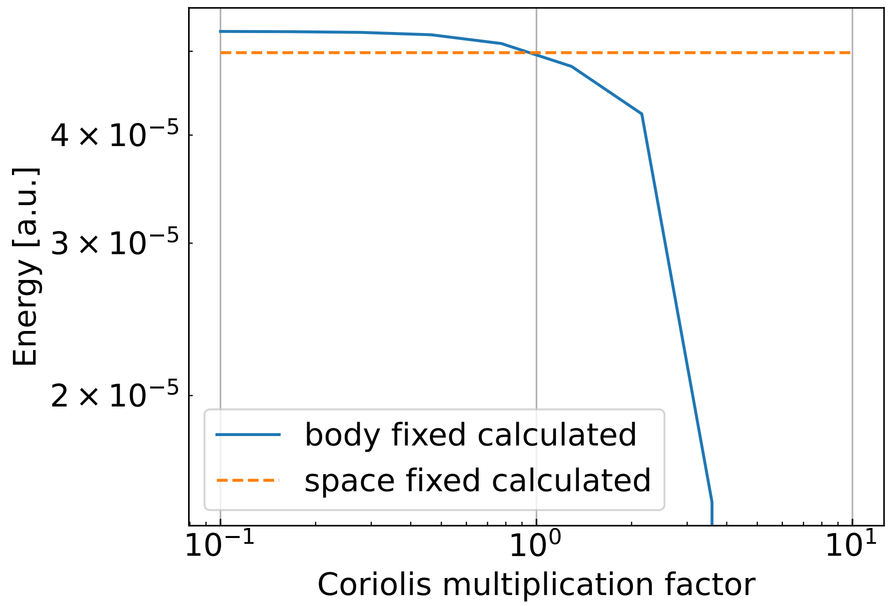
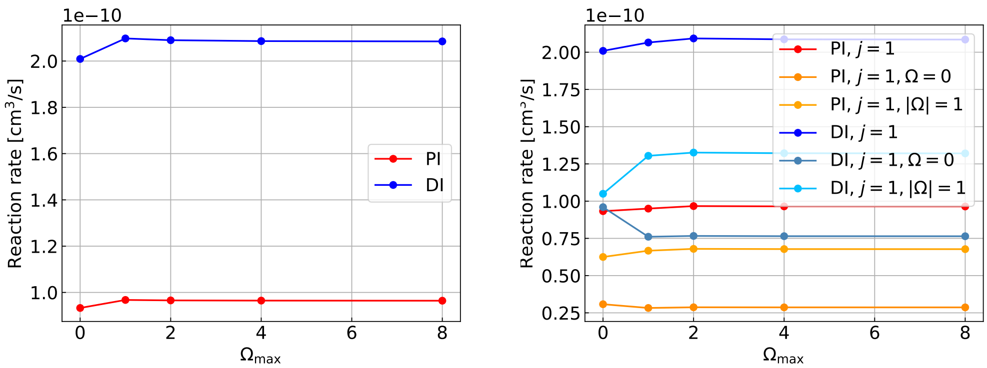


Figure 9: Calculated ground state energy dependence on the coriolis multiplication factor.

4 Reaction rate dependence on the coriolis effect body-fixed projection cutoff

Animations of collision with high $\Omega_{\max}$ showed that the mixing of the total projection is up to $\Omega = 45$. However reaction rate calculations converge for $\Omega_{\max} = 2$ as shown below, which means that mixing of Ω happens mostly after the collision.



(a) Reaction rate for $j = 0$.

(b) Reaction rate for $j = 1$.

Figure 10: Calculated reaction rates dependence on $\Omega_{\max}$.

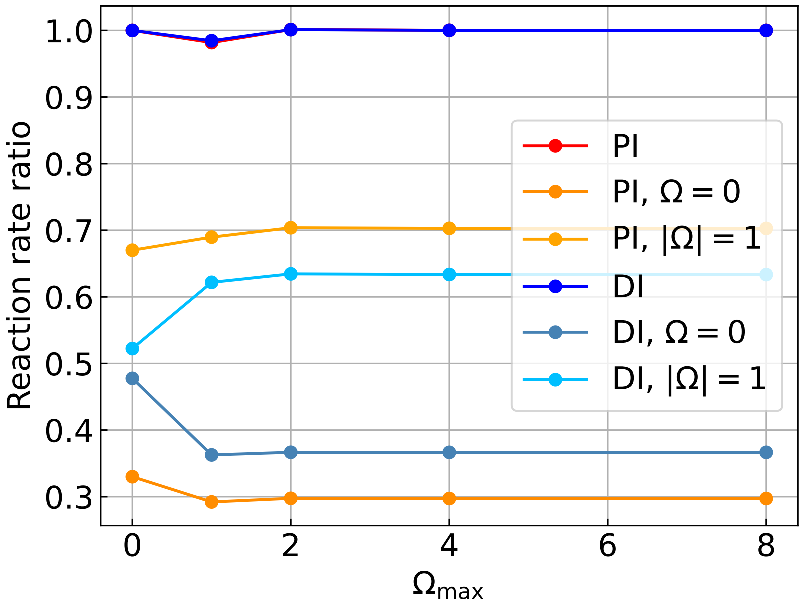


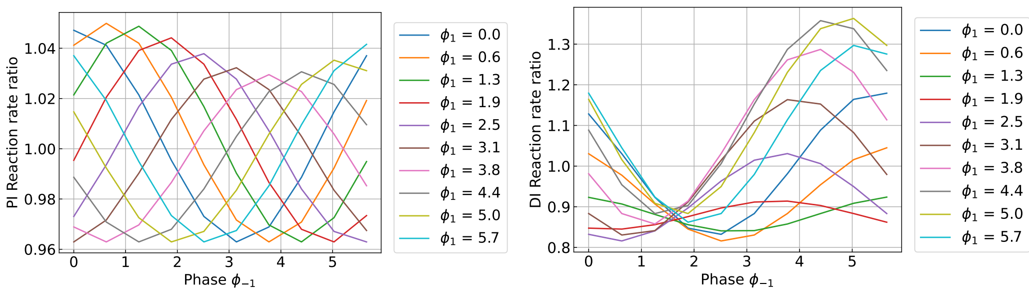
Figure 11: Calculated reaction rate ratio between $j = 1$ and $j = 0$, dependence on $\Omega_{\max}$.

5 Reaction rate for superposition states

For the initial state $j = 1$ being the superposition of initial body-fixed projections

$$|\psi\rangle = \frac{1}{\sqrt{3}} \left(e^{i\phi_{-1}} \left|-1\right\rangle + \left|0\right\rangle + e^{\phi_1} \left|1\right\rangle \right), \tag{4}$$

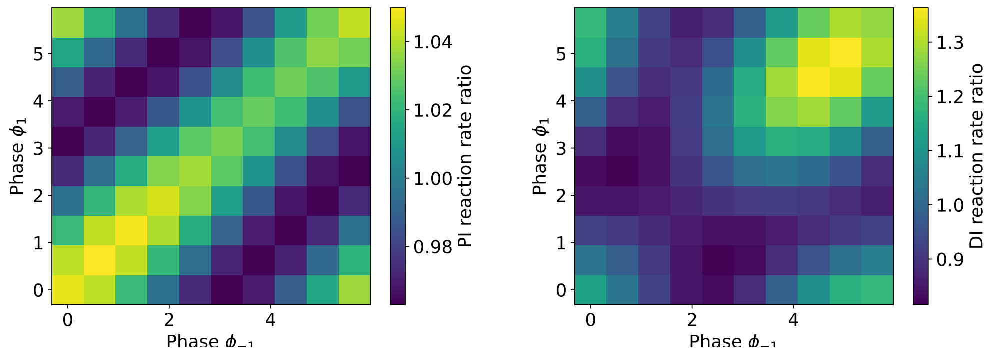
the reaction rate for different initial relative phases is shown below.



(a) Penning ionization reaction rate ratios for different relative phases.

(b) Dissociative ionization reaction rate ratios for different relative phases.

Figure 12: Reaction rate ratios for different relative phases.



(a) Penning ionization reaction rate ratios for different relative phases.

(b) Dissociative ionization reaction rate ratios for different relative phases.

Figure 13: Reaction rate ratios for different relative phases.

Notice that the reaction rate ratio changes from 1 far more for DI channel than the PI channel, which is consistent with the results of the experiment for which ratio for PI is much closer to 1 and DI is not.

We additionally calculated, for every relative phases, the alignment evolution as shown below.

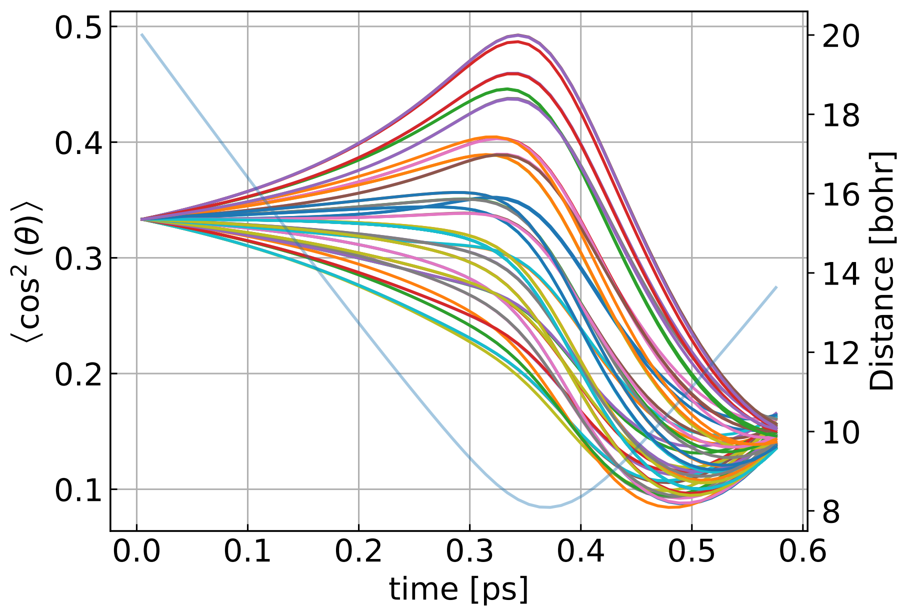


Figure 14: Calculated alignments during the collision for different relative phases.

This brings the idea that after the OCS free flight there is a coherent relative phase in the case of $j = 1$, this relative coherence might be a result of the geometry of the experimental setup or various electric/magnetic fields.