Quantum Interference in Chemical Reactions

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Introduction

- Interferences emerge when multiple pathways coexists together leading towards the same result.
- Experiments[1,2]have shown that if there exists multiple reaction pathways in a chemical reaction then these pathways may interfere with each other producing interference patterns.

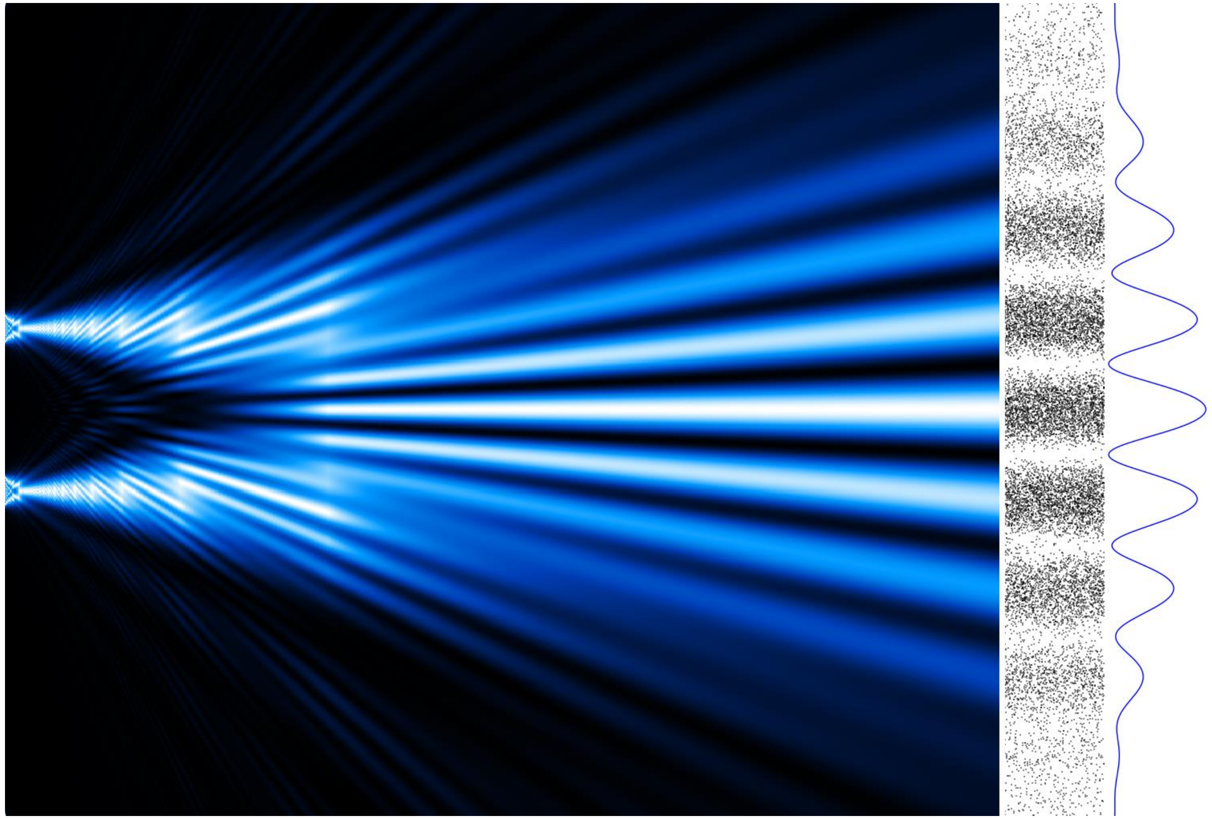
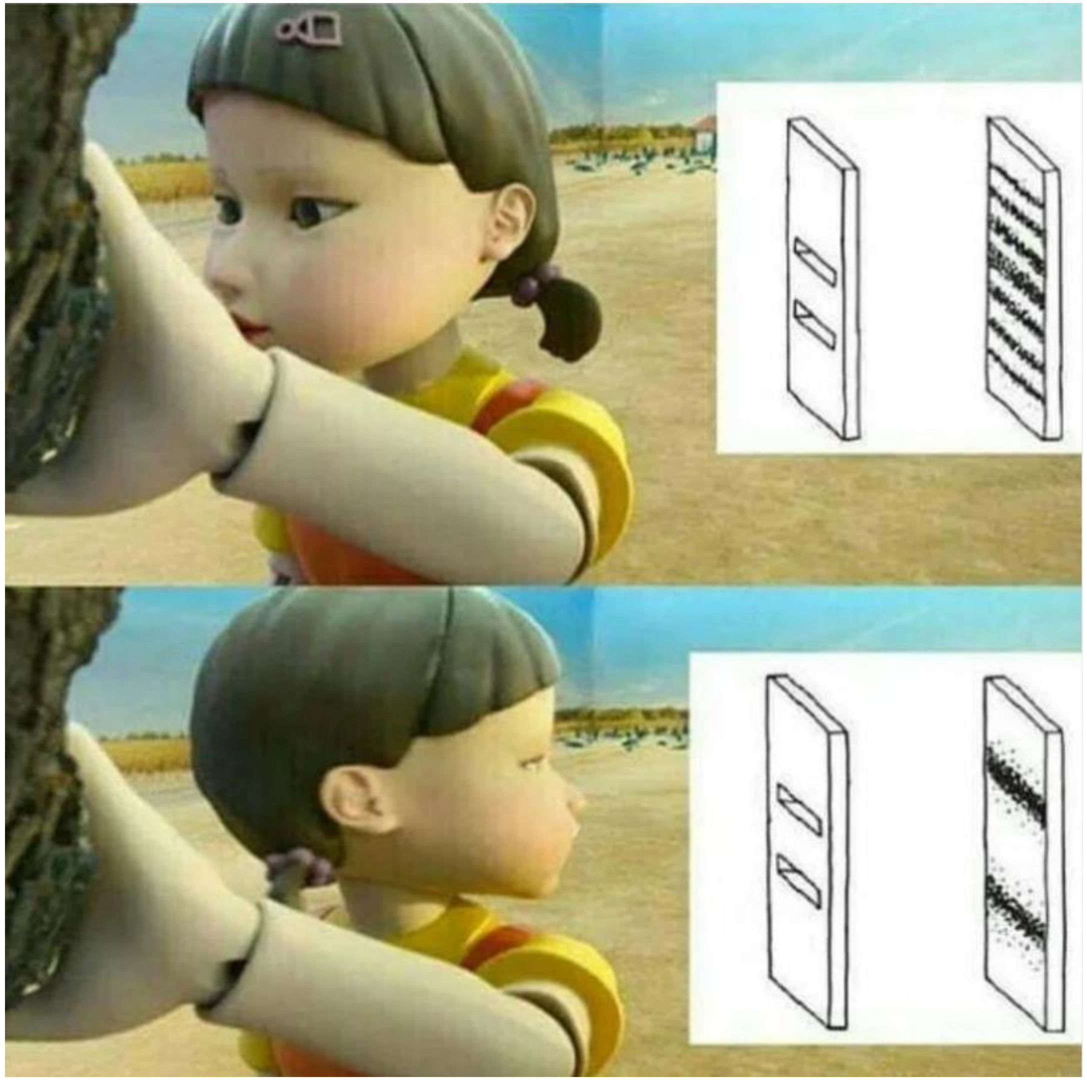


Fig 1: The double-slit experiment with electrons.

1: Nature chemistry, 7(8):661, 2015
2: The Journal of chemical physics, 145(2):024308, 2016

Superposition is Important!!



$$|\psi_{tot}\rangle = C_1|\psi_1\rangle + C_2|\psi_2\rangle$$

$$|\psi_1\rangle \text{ or } |\psi_2\rangle$$

Fig 2: Meme Explaining essentiality of Superposition to observe Quantum interference.

Is the simplest Chemical reaction so simple?

$$H + D_2(v = 0, j = 0) \rightarrow HD\ (v' = 1, j' = 0) + D$$

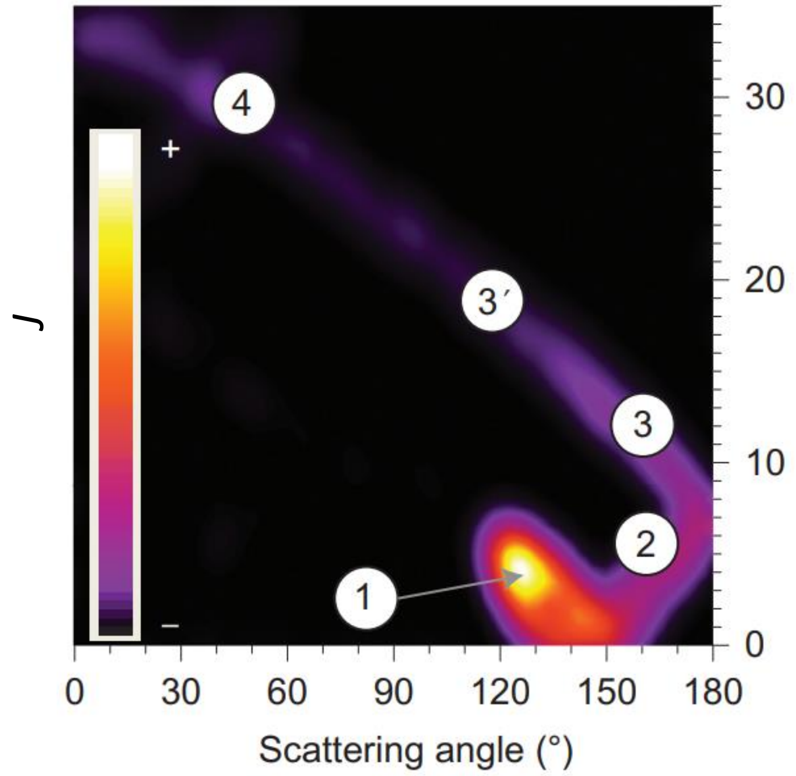


Fig 3a: The classical deflection function (Chemical Reactivity) *for product in* **(v′ = 1, j′ = 0)**

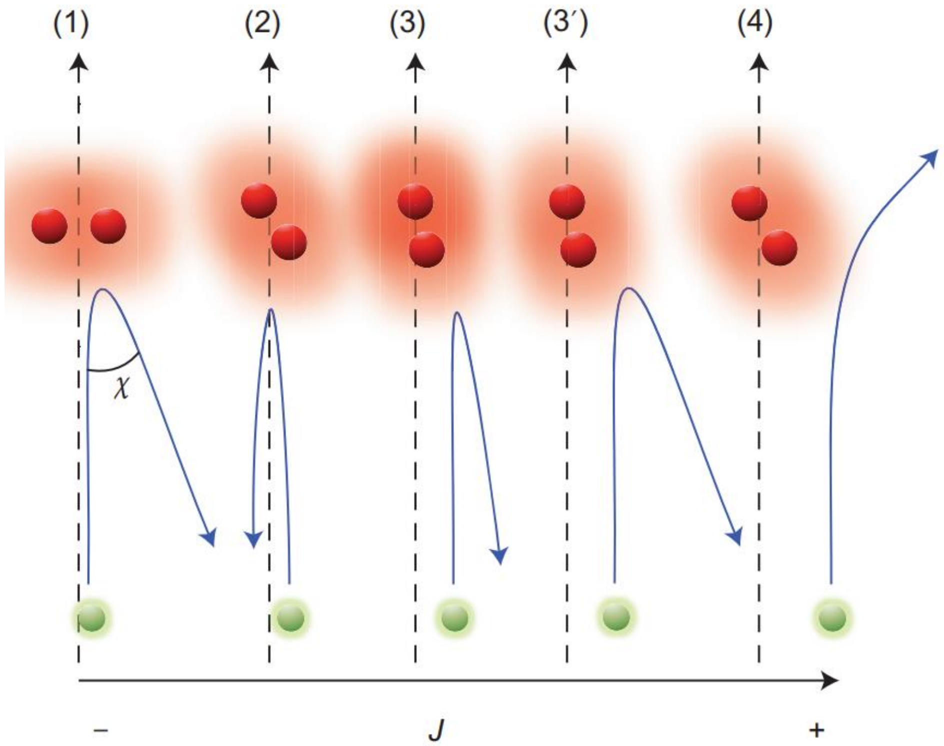


Fig 3b: Sketches of mechanisms that correspond to the various regions

1: Jambrina P. et al, Nature chemistry, 7(8):661, 2015

$$H + D_2(v = 0, j = 0) \rightarrow HD\,(v' = 1, j' = 0) + D$$

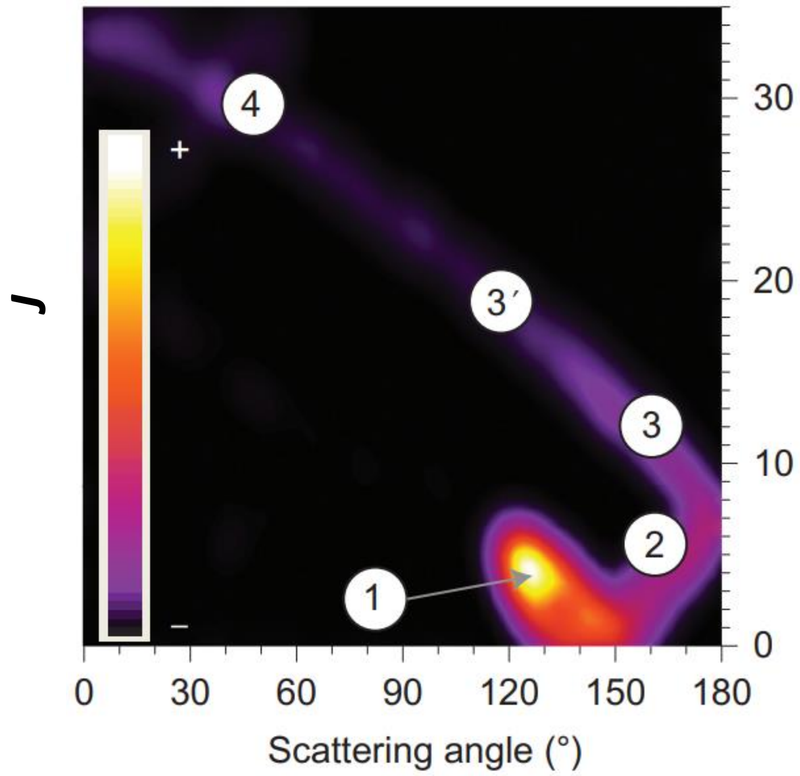


Fig 3a: The classical deflection function (Chemical Reactivity) *for product in* **(*v′* = 1, *j′* = 0)**

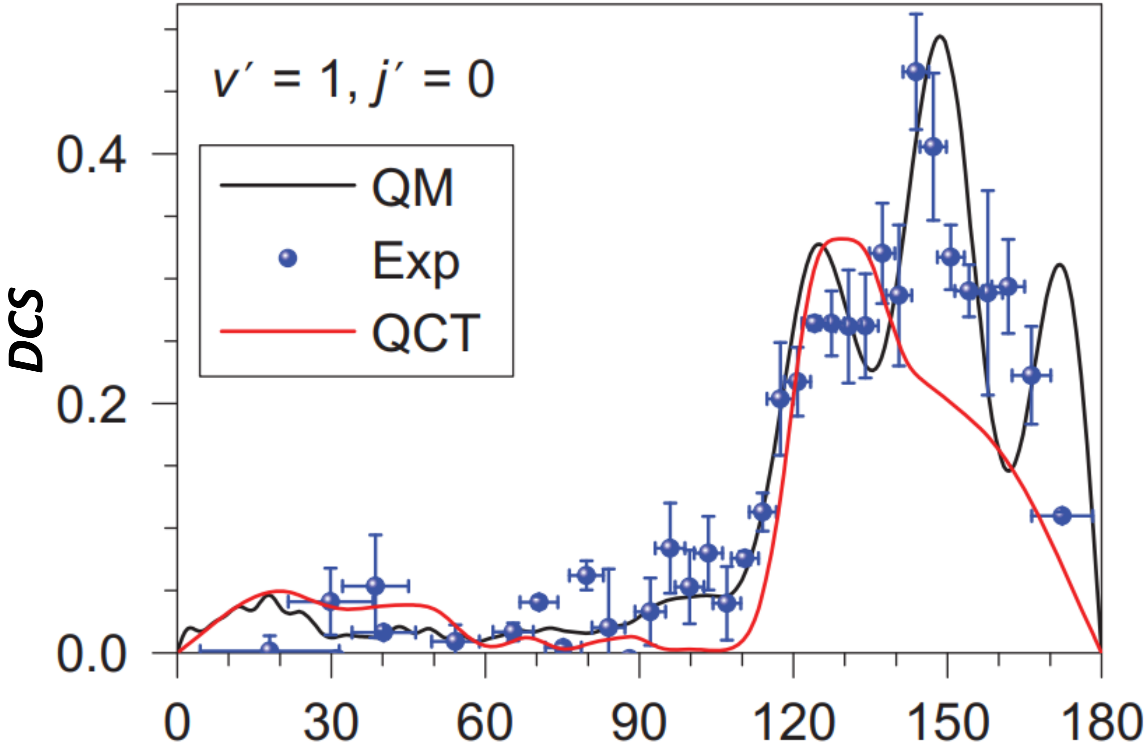
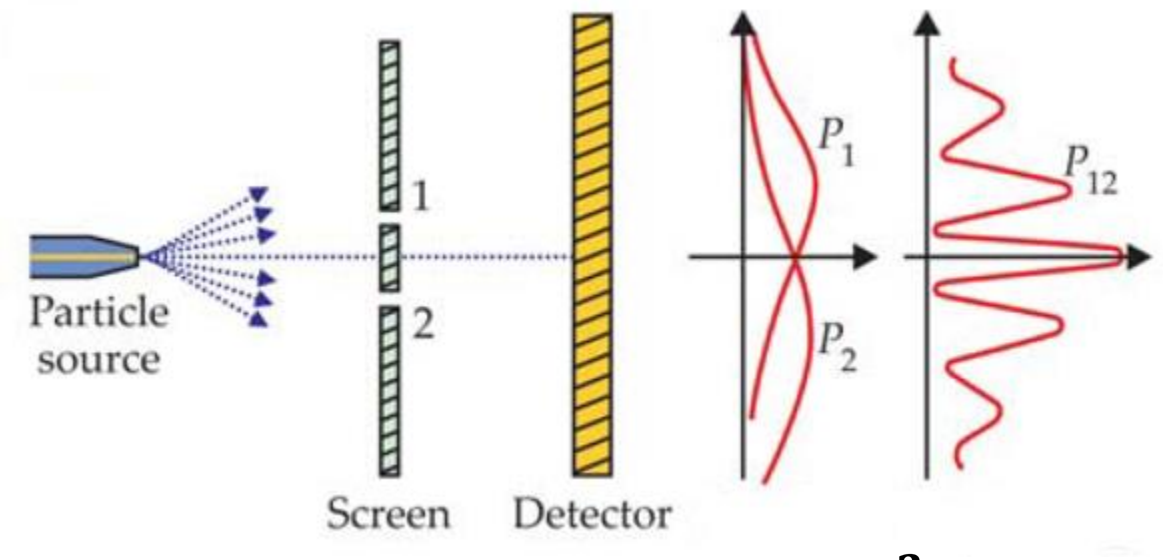


Fig 3c: DCS vs θ for **(*v′* = 1, *j′* = 0)**

1: Jambrina P. et al, Nature chemistry, 7(8):661, 2015

Why is it so important?

Fig 4: Interference (Adapted from Feynman Lectures of Physics)



$$\boldsymbol{P_1 = |A_1\psi_1|^2}$$
$$\boldsymbol{P_2 = |A_2\psi_2|^2}$$

$$\boldsymbol{P_{12} = |A_1\psi_1 + A_2\psi_2|^2}$$

Mechanism 1 (Fig 1b) has 10 times the amplitude of Mechanism 3'

$$(\boldsymbol{A_1 = 10\ A_2})$$

- In-coherent addition of Prob. DCS varies $[10^2 - 1^2, 10^2 + 1^2]$ $(\pm 1\%)$
- Coherent addition it varies between $(10 - 1)^2$ and $(10 + 1)^2$ i.e. [81,121] $(\pm 20\%)$ effect!!!

Small mixing, but big effect

The Photo-association reaction of ^{87}Rb.

- What happens when the reactants are prepared in a quantum superposition! [3]
- Laser Induced molecule formation of ^{87}**Rb (**F = 1**)** BEC.
- Spin Selective Chemical Reaction
- The target in the experiment was to get product molecule with F = 0, m_f = 0.

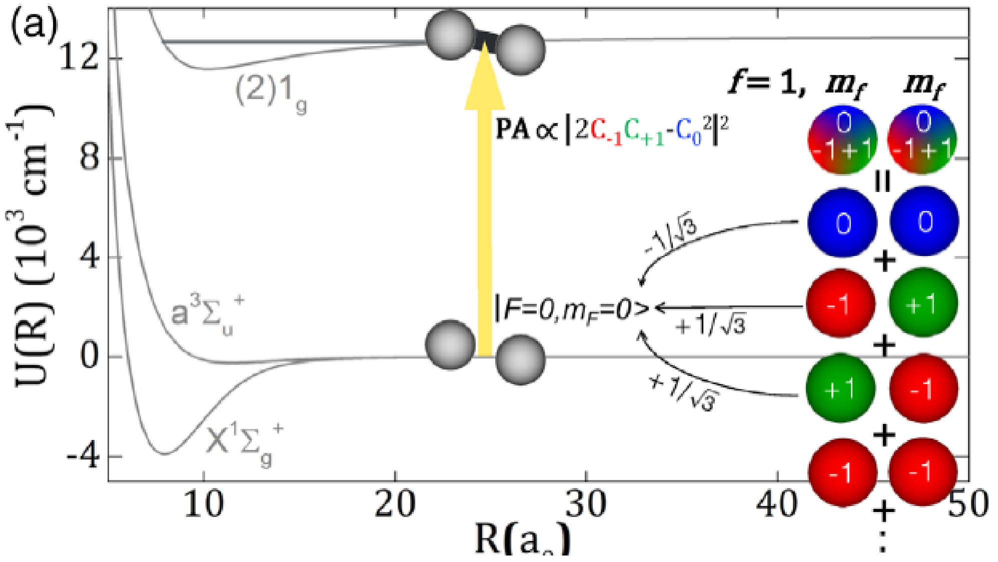


Fig 5: Molecular potential energy curves.

3: Physical review letters, 121(7):073202, 2018.

Spin-momentum Superposition

- ^{87}Rb BEC is prepared in $f = 1$ hyperfine state via optical evaporation.
- The magnetic field tuning during the optical evaporation results in a BEC with bare $m_f = -1,0,+1$ spin states.
- The superposition is achieved by applying two counter-propagating Raman lasers adiabatically which drives transitions between these atomic Zeeman levels.

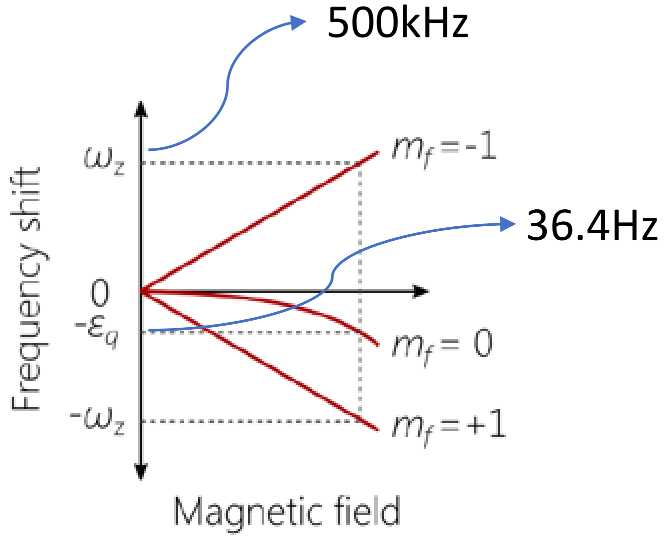


Fig. 6: Schematic representation of Zeeman splitting under bias magnetic field

Hamiltonian and the Chemical Reaction

- As a result, the Rb atom makes a transition from an m_f to an $m_f - 1$ hyperfine Zeeman state by absorbing and emitting a photon.
- This process induces, in addition, a change in the momentum of the atom by $2k_r$.[4]
- The Hamiltonian that describes such a spin (m_f) momentum (K) coupling can be written in the spin-momentum basis:

$$|m_f, K\rangle = \{|-1, q + 2k_r\rangle, |0, q\rangle, |+1, q - 2k_r\rangle\}$$

$$H_0 = \begin{pmatrix} \frac{\hbar^2}{2m}(q+2k_r)^2 - \delta & \frac{\Omega_r}{2} & 0 \\ \frac{\Omega_r}{2} & \frac{\hbar^2}{2m}q^2 - \epsilon(B) & \frac{\Omega_r}{2} \\ 0 & \frac{\Omega_r}{2} & \frac{\hbar^2}{2m}(q-2k_r)^2 + \delta \end{pmatrix}$$

4: Nature, 471(7336):83–86, 2011

Theoretical Investigation of Destructive Interference

- Total scattered wavefunction of one particle is denoted by:

$$\Psi_a = C_0 e^{i\vec{q}\vec{r}_a} |1,0\rangle_a + C_{+1} e^{i(\vec{q}+\vec{k}_r)\vec{r}_a} |1,1\rangle_a + C_{-1} e^{i(\vec{q}-\vec{k}_r)\vec{r}_a} |1,-1\rangle_a$$

$$\Gamma_{sup} \propto \left|\langle \phi_m(\vec{r}_{ab}) \big| \langle F=0, m_f=0 | \Psi_{scat} \rangle \right|^2$$

$$\Psi_{scat} = C_0^2 e^{i\vec{q}(\vec{r}_a+\vec{r}_b)} (|1,0\rangle_a + |1,0\rangle_b) + C_{+1}C_{-1} e^{i\vec{q}\vec{r}_{ab}} (|1,1\rangle_a + |1,-1\rangle_b) + C_{-1}C_{+1} e^{i\vec{q}\vec{r}_{ba}} (|1,-1\rangle_a + |1,+1\rangle_b) + \ldots$$

$$= e^{i\vec{q}.(\vec{r}_a+\vec{r}_b)} [C_0^2 (\sqrt{\frac{2}{3}} |2,0\rangle - \sqrt{\frac{1}{3}} |0,0\rangle)$$

$$+ C_1 C_{-1} e^{i\vec{k}_r.(\vec{r}_a-\vec{r}_b)} (\sqrt{\frac{1}{6}} |2,0\rangle - \sqrt{\frac{1}{2}} |1,0\rangle + \sqrt{\frac{1}{3}} |0,0\rangle)$$

$$+ C_{-1} C_1 e^{i\vec{k}_r.(\vec{r}_b-\vec{r}_a)} (\sqrt{\frac{1}{6}} |2,0\rangle - \sqrt{\frac{1}{2}} |1,0\rangle + \sqrt{\frac{1}{3}} |0,0\rangle) + \ldots]$$

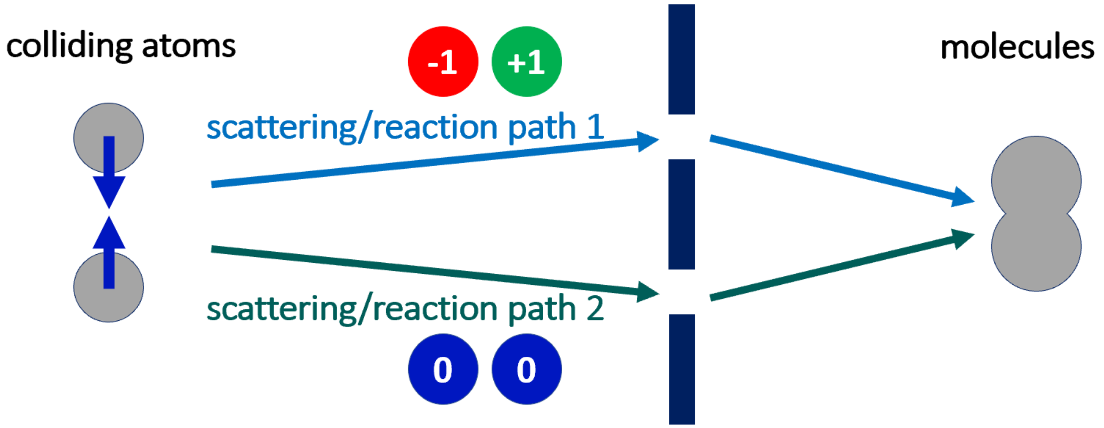
- The spin sensitive PA laser is then applied. Which selectively photo-associates colliding atoms total angular momentum and spin

$$| F = f_a + f_b, m_f = m_{f,a} + m_{f,b} \rangle = |0,0\rangle$$

- Using a single particle basis $|f_a, m_{f,a}\rangle \, |f_b, m_{f,b}\rangle$

$$\langle F = 0, m_f = 0 | \Psi_{scat} \rangle = (C_{+1} C_{-1} |1,-1\rangle |1,+1\rangle - C_0^2 |1,0\rangle |1,0\rangle + C_{-1} C_{+1} |1,+1\rangle |1,-1\rangle)/\sqrt{3}$$

$$= (C_0^2 |1,0\rangle |1,0\rangle - 2C_{-1} C_{+1} |1,+1\rangle |1,-1\rangle)/\sqrt{3}$$



$$\Gamma_{sup} \propto \left|\langle\phi_m(\vec{r}_{ab})\middle|\langle F=0, m_f=0|\Psi_{scat}\rangle\right|^2$$
$$\propto \left|\int \phi_m^*(\vec{r}_{ab})\phi_{F=0}(\vec{r}_{ab})\, d\vec{r}_{ab}\right|^2 \left|C_0^2 - 2C_{+1}C_{-1}\right|^2$$

$C_{+1} = C_{-1} = 0$ and $C_0 = 1$

$$\Gamma_{0,0} \propto \left|\int \phi_m^*(\vec{r}_{ab})\phi_{F=0}(\vec{r}_{ab})\, d\vec{r}_{ab}\right|^2$$

$$\frac{k_{sup}}{k_{0,0}} = \frac{\Gamma_{sup}}{\Gamma_{0,0}} = \left|C_0^2 - 2C_{+1}C_{-1}\right|^2$$

- For the reaction with Channel $|F=0, m_f=0\rangle$ the ratio of reaction rates:

$$\frac{k_{sup}}{k_{0,0}} = |C_0^2|^2 + 4|C_{-1}C_{+1}|^2 - 4Re[C_0^2 C_{-1} C_{+1}]$$

- The experiment $|\, F = 0, m_f = 0\rangle$ employs θ/ϵ PA lines of the spectrum. These are spin sensitive wrt m_f = 0.

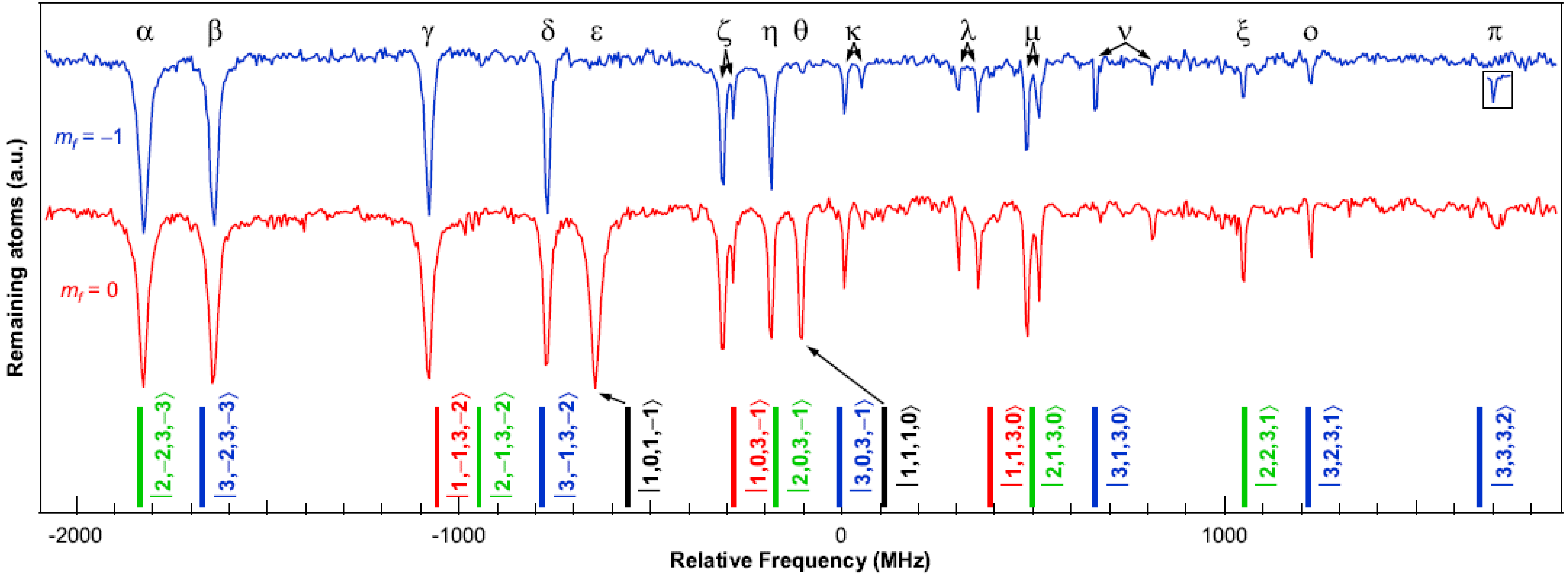


Fig. 7: Hamley et. al. Phys. Rev. A 79, 23401 (2009)

Reaction Rate vs Ω_r
Calculations for $|F = 0, m_f = 0\rangle$

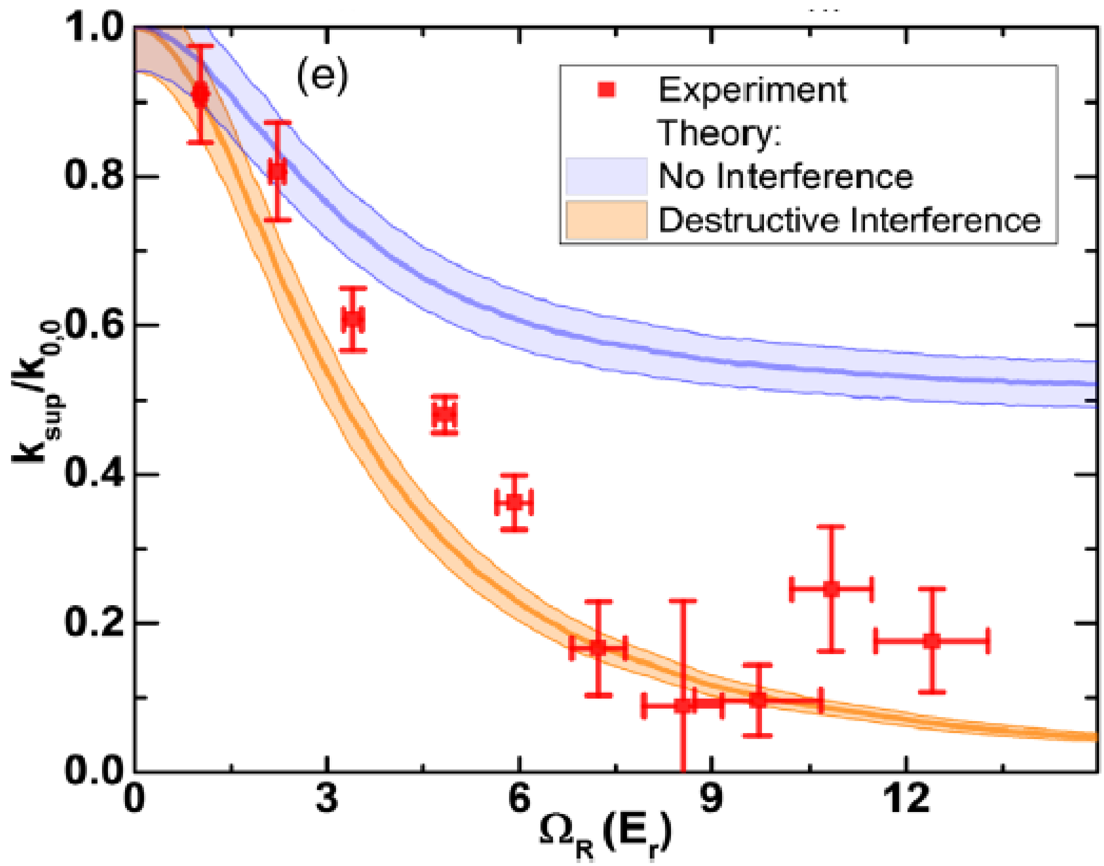


Fig. 8a: Normalized photo-association rate $k_{sup}/k_{0,0}$ as a function of Ω_r/E_r (Blasing et. al. , PRL 2018)

$$\frac{k_{sup}}{k_{0,0}} = \left|C_0^2 - 2C_{+1}C_{-1}\right|^2$$

$$|C_{-1}| = |C_1| = 1/\sqrt{2}|C_0|$$

Reaction Rate vs $\boldsymbol{\delta}_r$
Calculations for $|F = 0, m_f = 0\rangle$

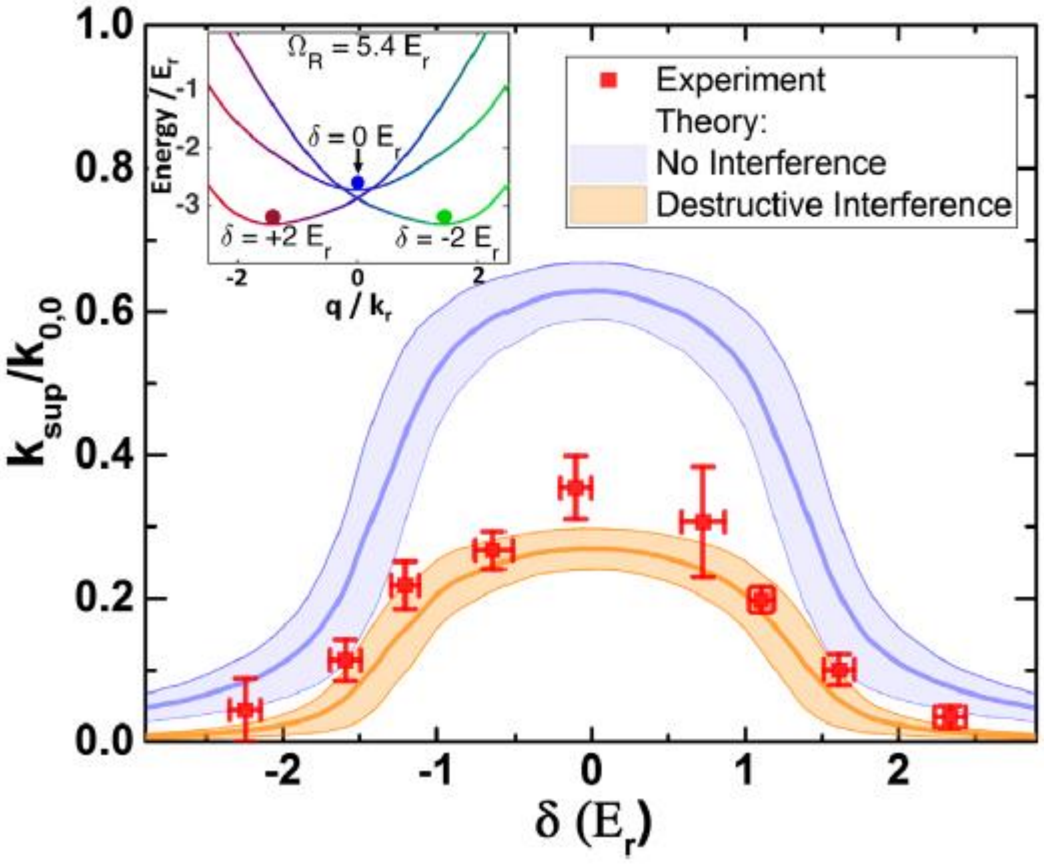


Fig. 9a: Normalized photo-association rate $k_{sup}/k_{0,0}$ as a function of $\boldsymbol{\delta}_r/E_r at\ \boldsymbol{\Omega}_r = 5.4\ E_r$ (Blasing et. al. , PRL 2018)

Achieving Constructive Interference

- Changing the reaction scheme where the molecular hyperfine state F = 1 couples to a pair of colliding atoms whose total angular momentum is $|\, F = f_a + f_b, m_f = m_{f,a} + m_{f,b}\rangle$ = |2,0⟩.
- Using a single particle basis $|f_a, m_{f,a}\rangle\, |f_b, m_{f,b}\rangle$

$$\langle \boldsymbol{F = 2, m_f = 0 | \Psi_{scat}} \rangle \ = (\boldsymbol{C_{+1} C_{-1} |1, -1\rangle |1, +1\rangle + 2C_0^2 |1, 0\rangle |1, 0\rangle + C_{-1} C_{+1} |1, +1\rangle |1, -1\rangle})/\sqrt{\boldsymbol{6}}$$

- Following the same procedure. We get the ratio of reaction rates of superposition to bare stat. mixture:

$$\frac{k_{sup}}{k_{0,0}} = |C_0^2|^2 + |C_{-1} C_{+1}|^2 + 2Re[C_0^2 C_{-1} C_{+1}] \qquad (4)$$

Reaction Rate Calculations for $|F = 2, m_f = 0\rangle$

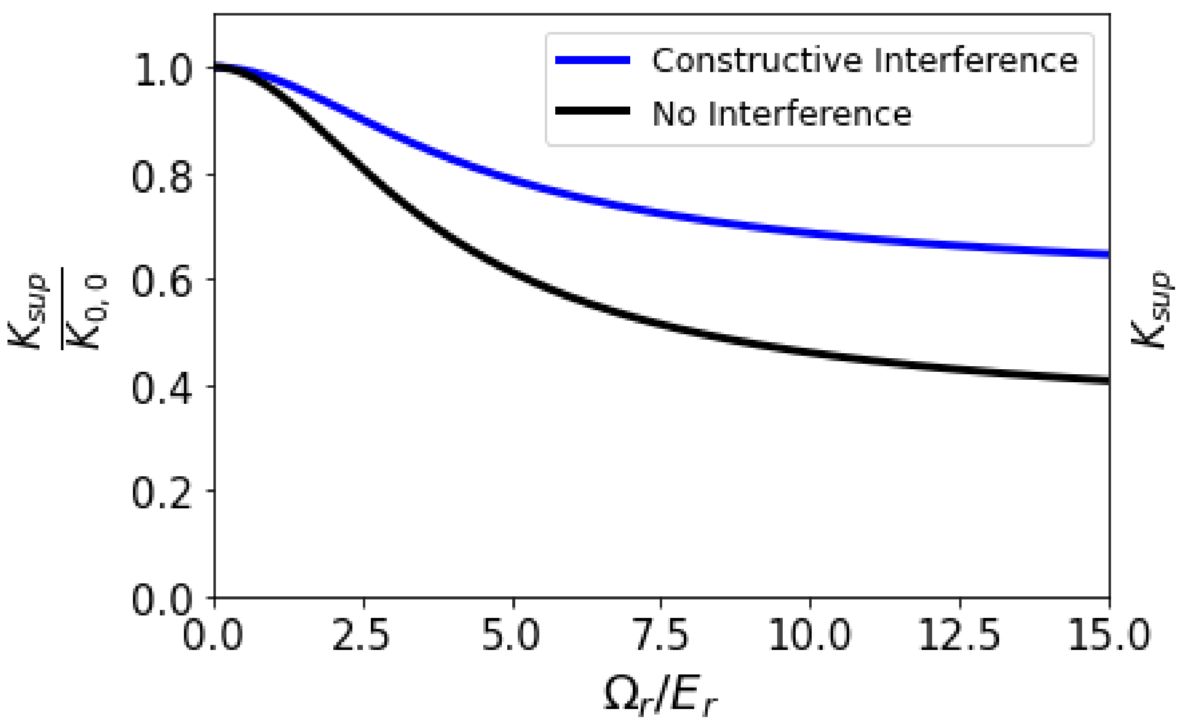


Fig. 10a: Normalized photo-association rate $k_{sup}/k_{0,0}$ as a function of $\boldsymbol{\Omega}_r/E_r$

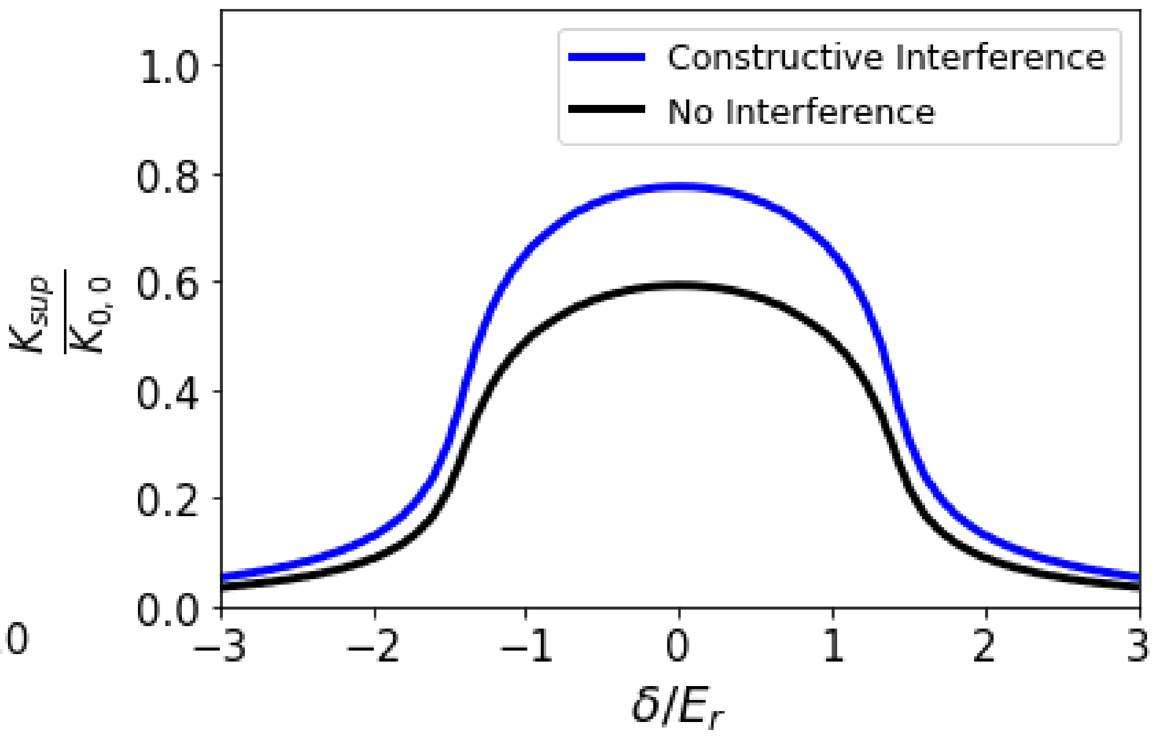


Fig. 10b: Normalized photo-association rate $k_{sup}/k_{0,0}$ as a function of $\boldsymbol{\delta}_r/E_r$ at $\boldsymbol{\Omega}_r = 5.4E_r$

Using RF beam to create superposition

- By changing the rotation angle θ_y around y axis we can change the population in $m_f = 0$ and $m_f = \pm 1$
- Recent study by Esat[4] et. al. realized these rotations in the system (Specifically single $\frac{\pi}{2}$ rotation around y axis)

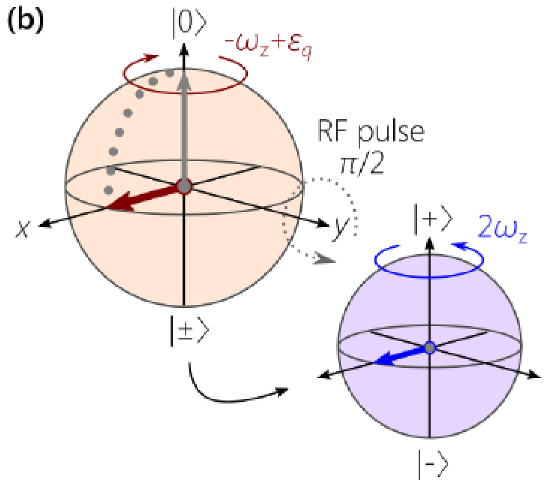


Fig. 11a: A Bloch-sphere representation of three level quantum state

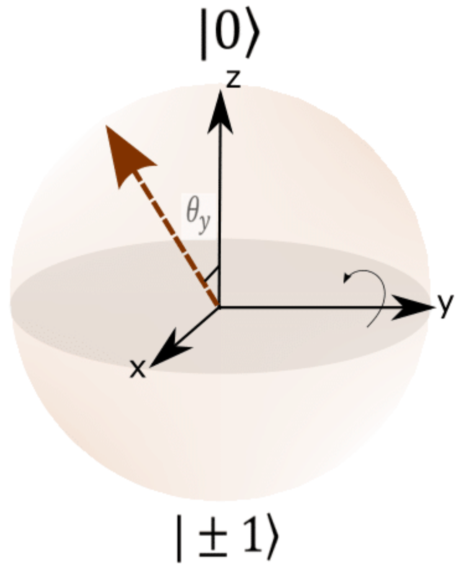
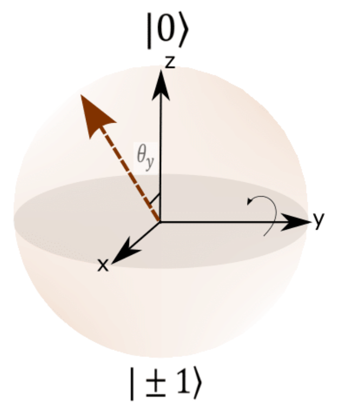


Fig. 11b: Bloch sphere illustration showing the rotation θ around y

4: 2020 Conference on Lasers and Electro-Optics (CLEO), pages 1–2. IEEE, 2020

- Total scattered wavefunction of one particle is denoted by:

$$\Psi_a = C_0|1,0\rangle_a + C_{+1}\ |1,1\rangle_a + C_{-1}\ |1,-1\rangle_a$$



- Simulation was done on Qiskit state vector simulator and the real quantum device.

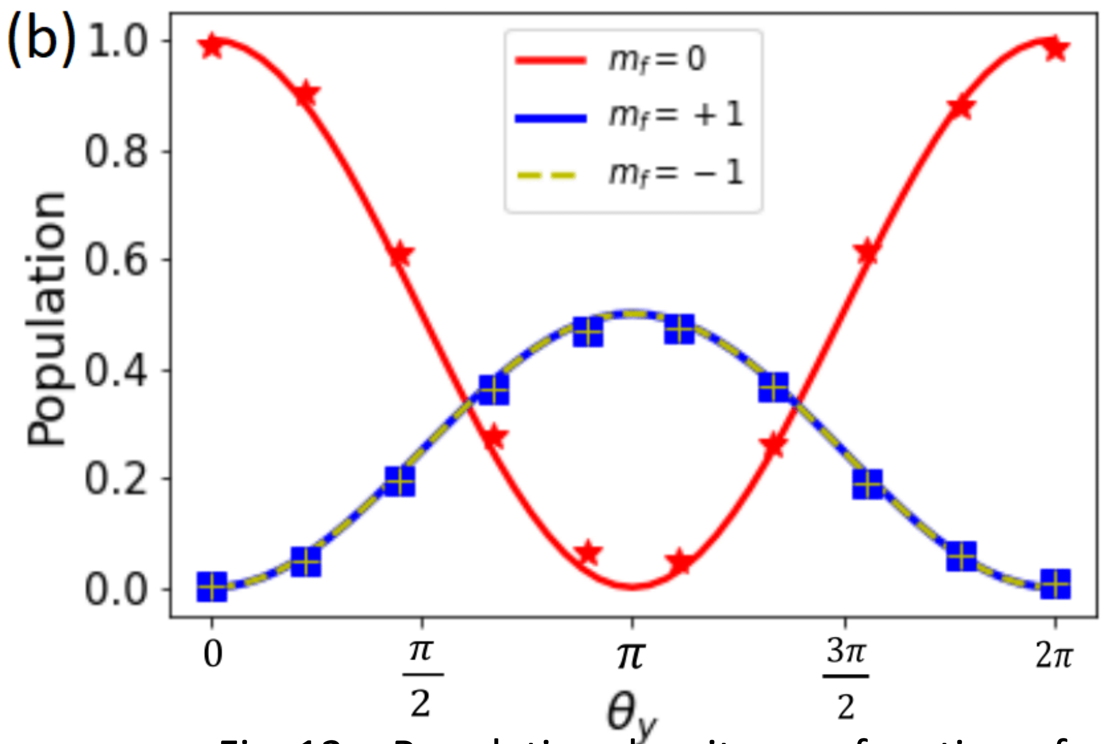


Fig. 12a: Population density as a function of rotation θ around y axis

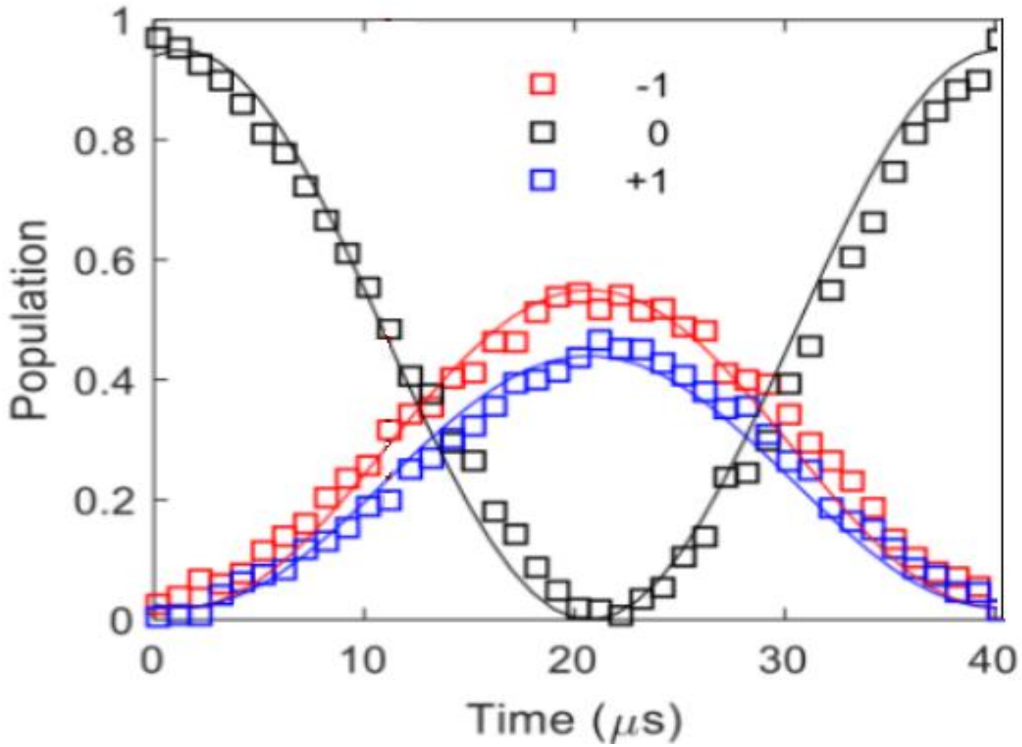


Fig. 12b: Experimental results by Esat et. al.

Reaction Rate Calculations

$$|\,F = 0, m_f = 0\rangle$$

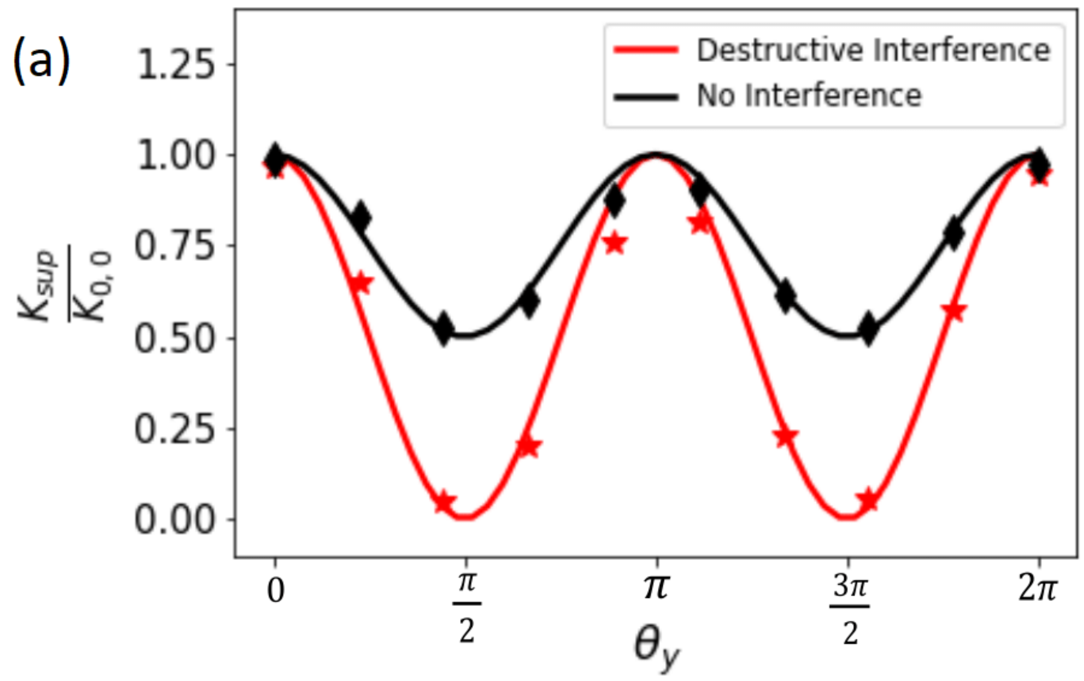


Fig. 13a: Normalized photo-association rate $k_{sup}/k_{0,0}$ as a function of θ_y

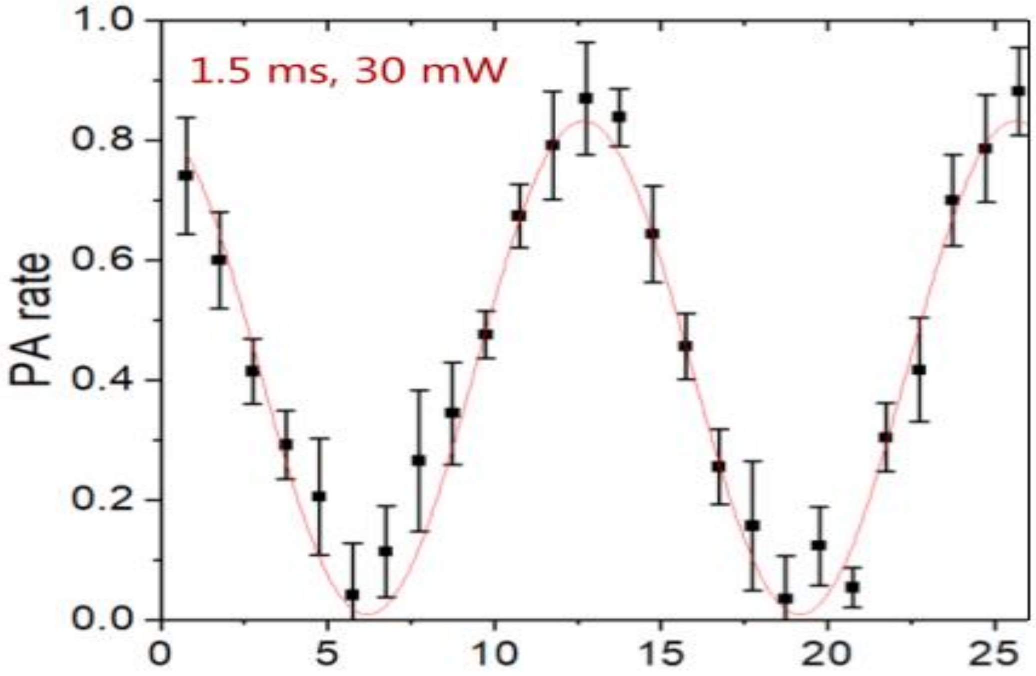


Fig. 13c: Experimental Results

$$\frac{k_{sup}}{k_{0,0}} = |C_0^2|^2 + 4|C_{-1}C_{+1}|^2 - 4Re[C_0^2 C_{-1} C_{+1}]$$

Reaction Rate Calculations

$$|\, F = 2, m_f = 0\rangle$$

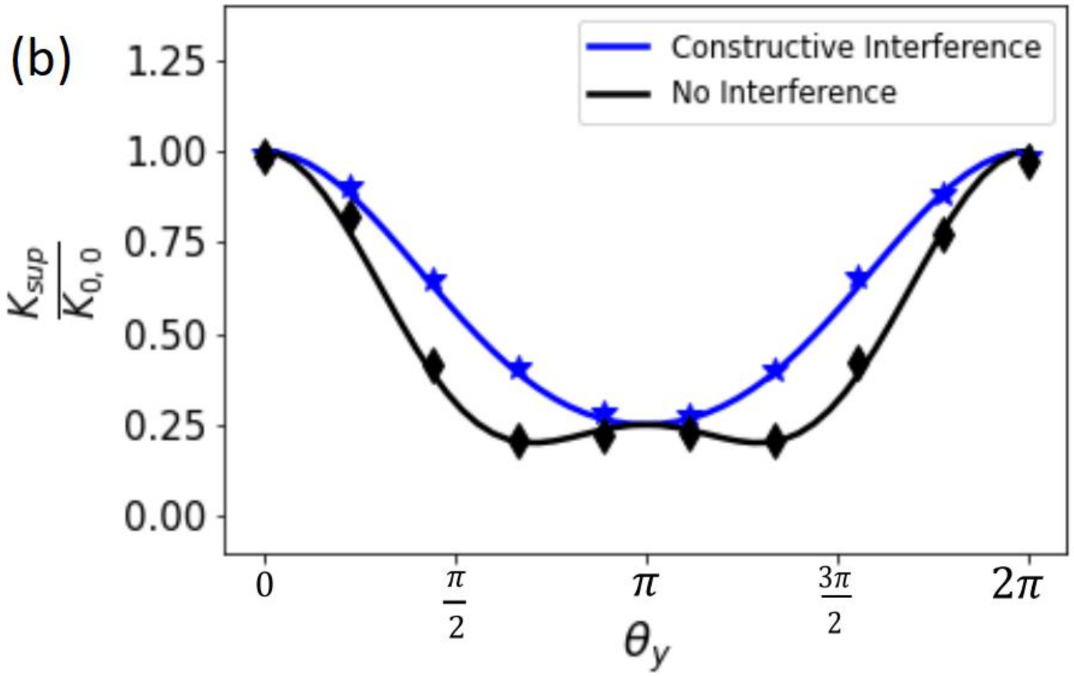


Fig. 13b: Normalized photo-association rate $k_{sup}/k_{0,0}$ as a function of θ_y

$$\frac{k_{sup}}{k_{0,0}} = |C_0^2|^2 + |C_{-1}C_{+1}|^2 + 2Re[C_0^2 C_{-1} C_{+1}]$$

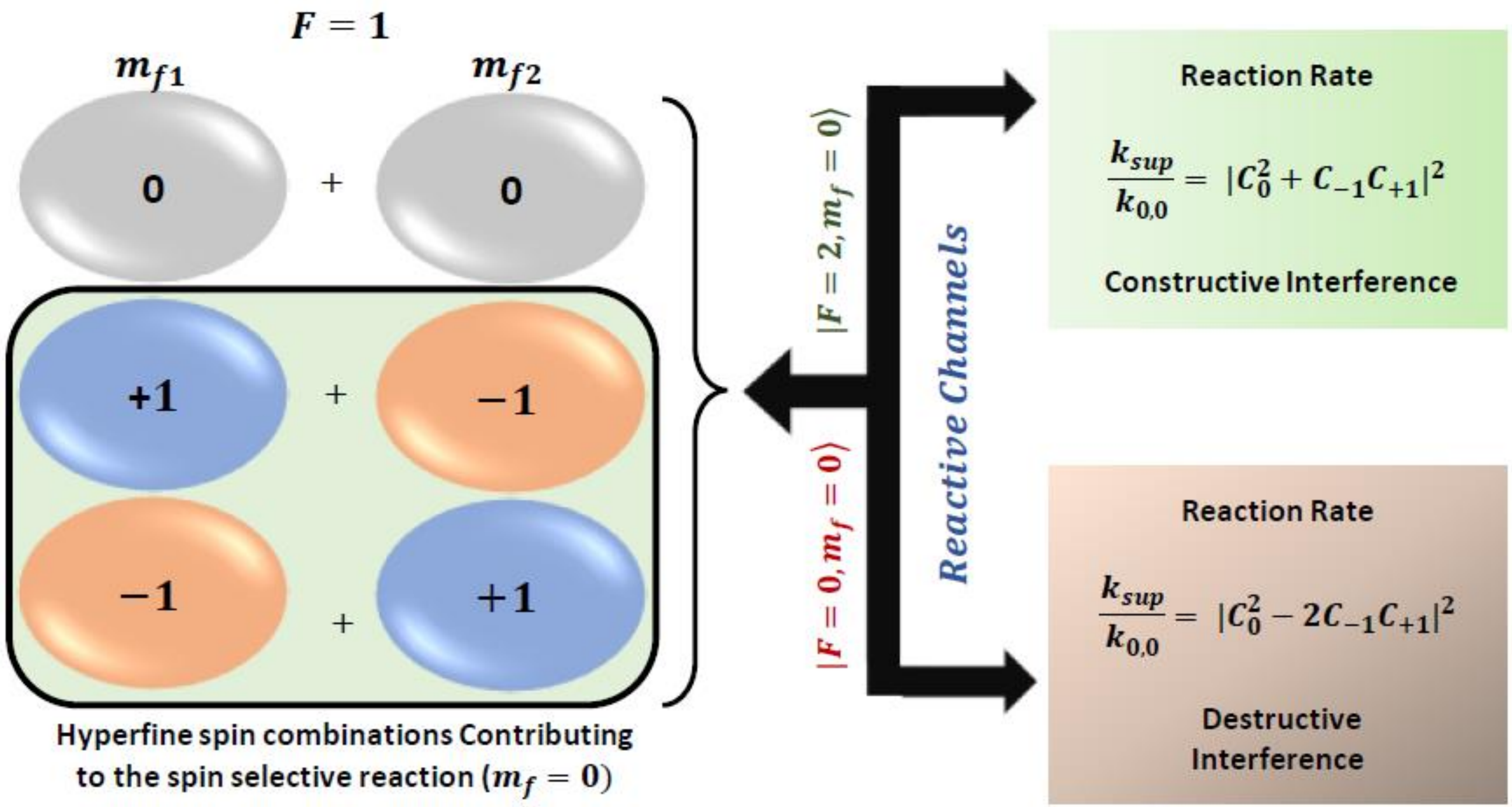


Fig. 14: Overview of the Reaction Schemes

Interferometric Control Over the reaction rate

- After keeping the rotation angle $\theta_y = \frac{\pi}{2}$
  We will introduce θ_z (Rotation around Z)
- Additional phase between $m_f = 0$ and $m_f = \pm 1$

$$\Psi_a = C_0|1,0\rangle_a + e^{i\theta_z}\left[C_{+1}|1,1\rangle_a + C_{-1}|1,-1\rangle_a\right]$$

- How would that affect the reaction rate ratio?

$$|F = 0, m_f = 0\rangle$$

$$\frac{k_{sup}}{k_{0,0}} = |C_0^2|^2 + 4|C_{-1}C_{+1}|^2 - 4Re[C_0^2 C_{-1} C_{+1}]$$

$\cos(\theta_z)$

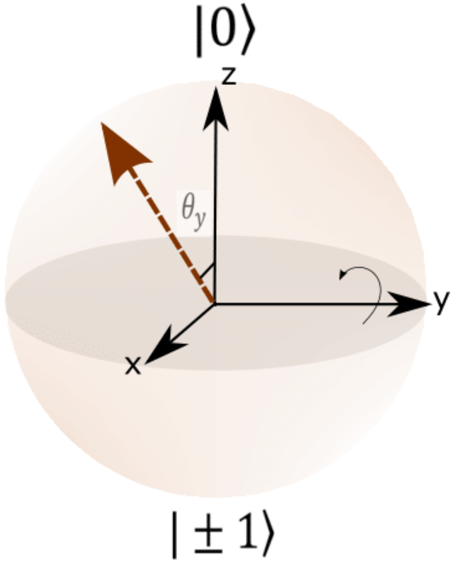


Fig. 15: Bloch sphere illustration showing the rotation θ around y

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Constructive Quantum Interference in Photochemical Reactions

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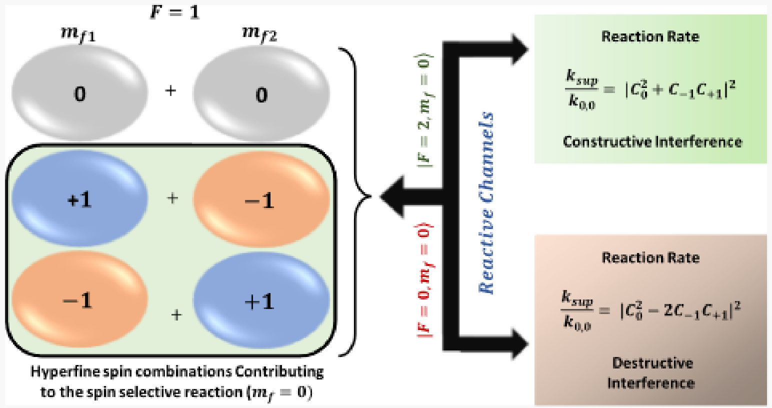
ABSTRACT: Interferences emerge when multiple pathways coexist together, leading toward the same result. Here, we report a theoretical study for a reaction scheme that leads to constructive quantum interference in a photoassociation (PA) reaction of a ^{87}Rb Bose–Einstein condensate where the reactant spin state is prepared in a coherent superposition of multiple bare spin states. This is achieved by changing the reactive scattering channel in the PA reaction. As the origin of coherent control comes from the spin part of the wavefunction, we show that it is sufficient to use radio frequency (RF) coupling to achieve the superposition state. We simulate the RF coupling on a quantum processor (IBMQ Lima), and our results show that interferences can be used as a resource for the coherent control of photochemical reactions. The approach is general and can be employed to study a wide spectrum of chemical reactions in the ultracold regime.



What is Entanglement?

- Entanglement is a characteristic feature of Quantum Mechanics.
- Schrödinger termed it as "Verschränkung".

"The best possible knowledge of whole does not
include the best possible knowledge of its parts"

- Used as a resource in Quantum Computing & essential for Quantum Speedup.
- Example: Bell's state

$|0\rangle$ = 

$|1\rangle$ = 

$|\psi_{bell}\rangle = \frac{1}{\sqrt{2}}$ (|   $\rangle$ + |   $\rangle$)

Spin-momentum entanglement in BECs

- The Hamiltonian that describes spin (m_f) momentum (K) coupling can be written in the spin-momentum basis:

$$|m_f, K\rangle = \{|-1, q+2k_r\rangle, |0, q\rangle, |1, q-2k_r\rangle\}$$

$$H_0 = \begin{pmatrix} \frac{\hbar^2}{2m}(q+2k_r)^2 - \delta & \frac{\Omega_r}{2} & 0 \\ \frac{\Omega_r}{2} & \frac{\hbar^2}{2m}q^2 - \epsilon(B) & \frac{\Omega_r}{2} \\ 0 & \frac{\Omega_r}{2} & \frac{\hbar^2}{2m}(q-2k_r)^2 + \delta \end{pmatrix}$$

$$\Psi_{scat,g} = C_0\,|q,0\rangle_a + C_{+1}\,|q-2\vec{k}_r, 1\rangle_a + C_{-1}\,|q+2\vec{k}_r, -1\rangle_a$$

- Use Spin & momentum degrees of freedom as two independent computing units
- One qutrit corresponds to $\{|q+2k_r\rangle, |q\rangle, |q-2k_r\rangle\}$ and the second qutrit corresponds to $\{|-1\rangle, |0\rangle, |+1\rangle\}$

Entanglement Measure (Von Neumann Entropy)

- Using Schmidt decomposition, the ground state wavefunction can be recast into

$$|\psi\rangle = \sum_{i=1}^{d} \alpha_i |u_i\rangle_{\mathrm{A}} |v_i\rangle_{\mathrm{B}}$$

- For a bipartite pure system, A and B the Von Neumann entropy is

$$S = -\sum_{i=1}^{d} |\alpha_i|^2 \log_3 |\alpha_i|^2$$

- Measures how random the Subsystem A is when ignoring all information from subsystem B.

Entanglement Measure (Concurrence or EOF)

- Based on the concept of spin flipped states. For a bipartite system of two qutrits. The steps involve
- Calculating the density matrix $\rho = |\psi_{scat,g}\rangle \langle\psi_{scat,g}|$
- Constructing spin flipped density matrix $\widetilde{\rho} = (\sigma_y \bigotimes \sigma_y)\rho^*(\sigma_y \bigotimes \sigma_y)$
- Calculating eigenvalues $\lambda_1, \lambda_2, \lambda_3 \ldots$ of $\rho\widetilde{\rho}$
- Concurrence $= \max[0, \sqrt{\lambda_1} - \sqrt{\lambda_2} - \sqrt{\lambda_3} - \ldots]$

$$\begin{pmatrix} 0 & -i & i \\ i & 0 & -i \\ -i & i & 0 \end{pmatrix}$$

Results

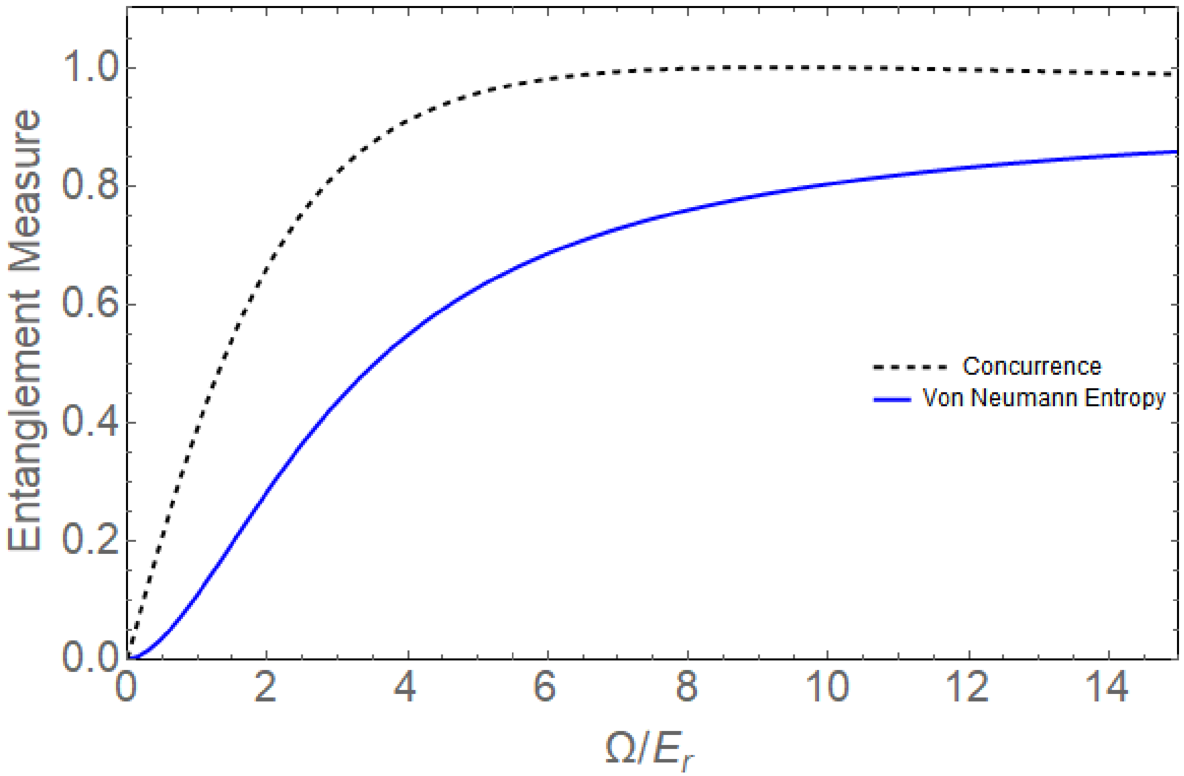


Fig. 16a: Entanglement Measures vs $\boldsymbol{\Omega}_r/E_r$ at $\boldsymbol{\delta}_r$ =0

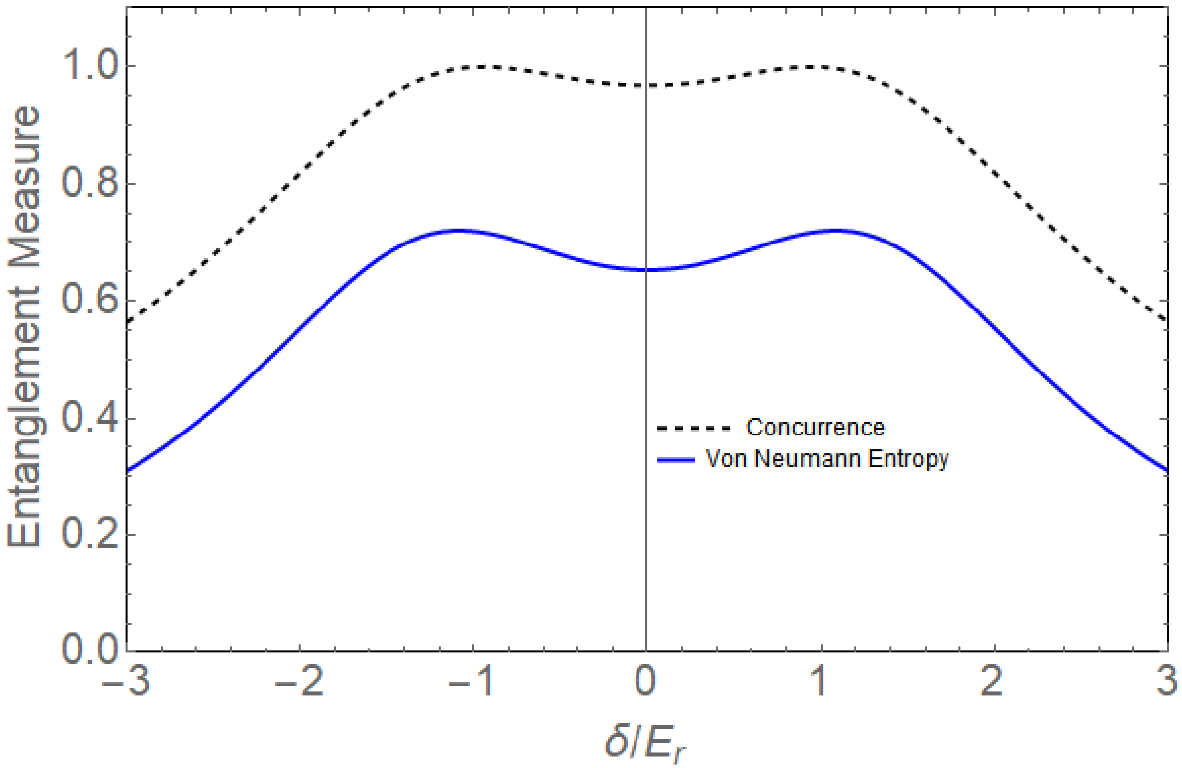


Fig. 16b: Entanglement measures vs $\boldsymbol{\delta}_r/E_r$ at $\boldsymbol{\Omega}_r = 5.4E_r$

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Spin-momentum entanglement in a Bose–Einstein condensate

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Entanglement is at the core of quantum information processing and may prove essential for quantum speed-up. Inspired by both theoretical and experimental studies of spin-momentum coupling in systems of ultra-cold atoms, we investigate the entanglement between the spin and momentum degrees of freedom of an optically trapped BEC of ^{87}Rb atoms. We consider entanglement that arises due to the coupling of these degrees of freedom induced by Raman and radio-frequency fields and examine its dependence on the coupling parameters by evaluating von Neumann entropy as well as concurrence as measures of the entanglement attained. Our calculations reveal that under proper experimental conditions significant spin-momentum entanglement can be obtained, with von Neumann entropy of 80% of the maximum attainable value. Our analysis sheds some light on the prospects of using BECs for quantum information applications.

Summary and Outlook

- Can Control chemical reaction using quantum mechanical properties.
- Realizing constructive interference in PA reaction with two different ways:
  - Using Raman beam to superimpose different hyperfine spins and then letting the reaction happen through $|\mathrm{F}=2, m_f=0\rangle$.
  - Changing the population in in different hyperfine spin levels by introducing a y-rotation using rf.
- Possible to achieve interferometric control over the reaction.
- A single boson can be used as a pair of quitrit.
- Extension to a triatomic system and it's simulation on Quantum Computers.

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The Kais research group and Collaborators

Thank You All!

- The analysis shows that the total nuclear spin of the molecular state must be 1 for F = 0 scattering channel and predominantly I = 3 for the F = 2 scattering channel.

- To conduct an exp. For F = 2 there should be a line which is spin selective and which corresponds to molecular states $|\, F = 1/2/3, I \ = 3\rangle$

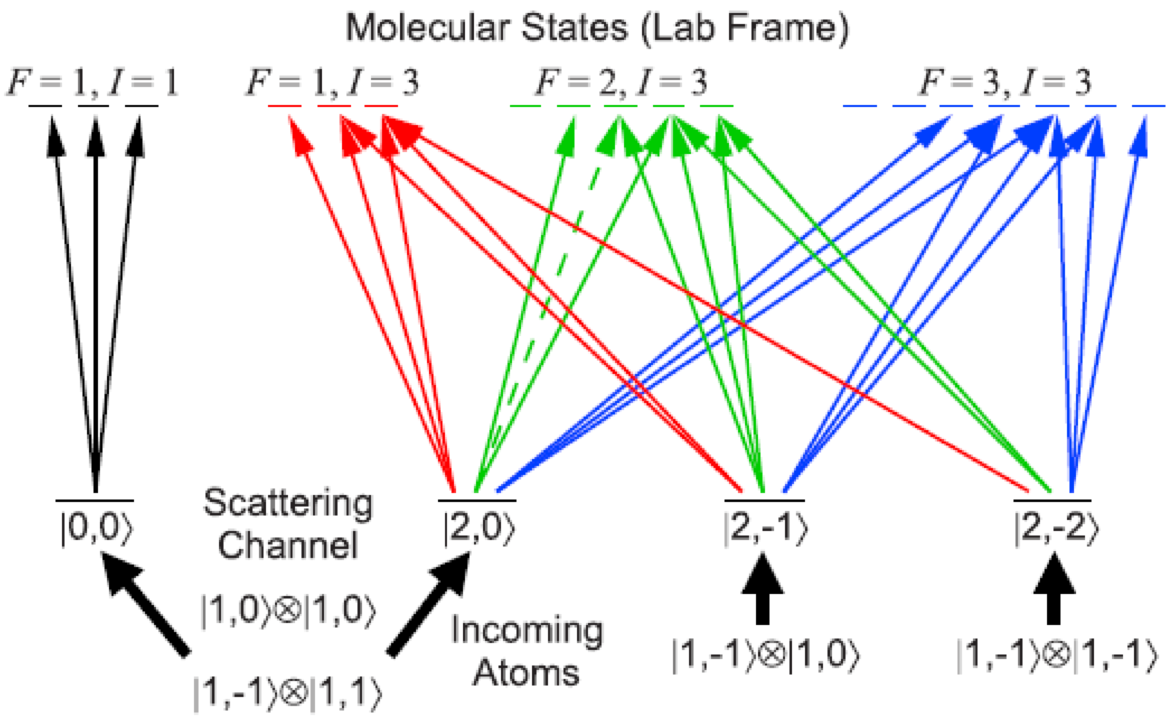

Fig. 8: Coupling Schema Hamley et. al. Phys. Rev. A 79, 23401 (2009)

Experimental Overview of Superposition

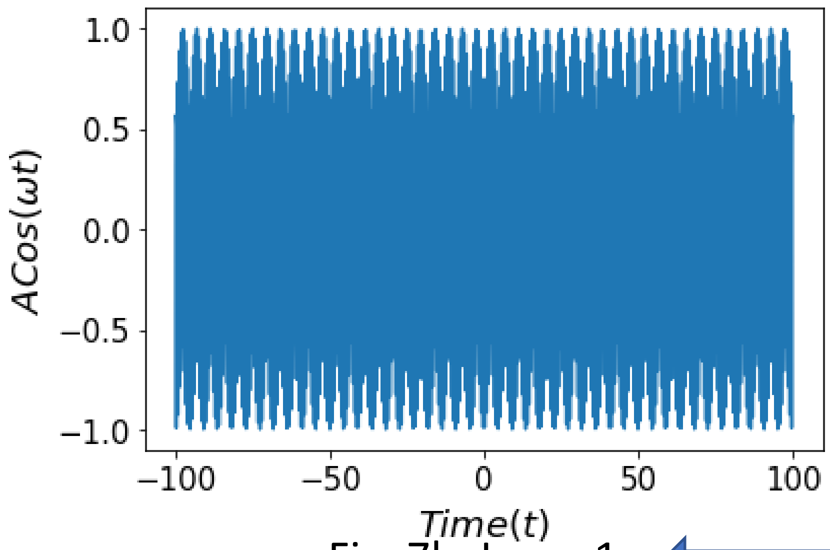

Fig. 7b: Laser 1

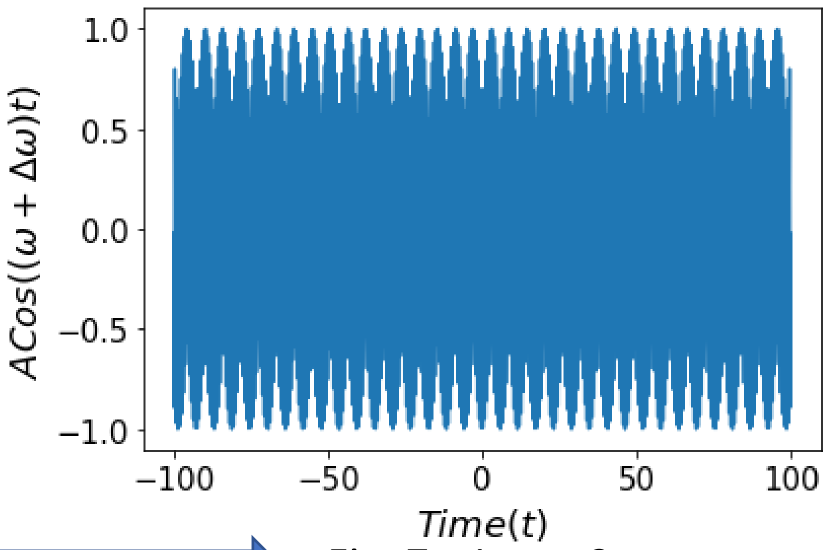

Fig. 7c: Laser 2

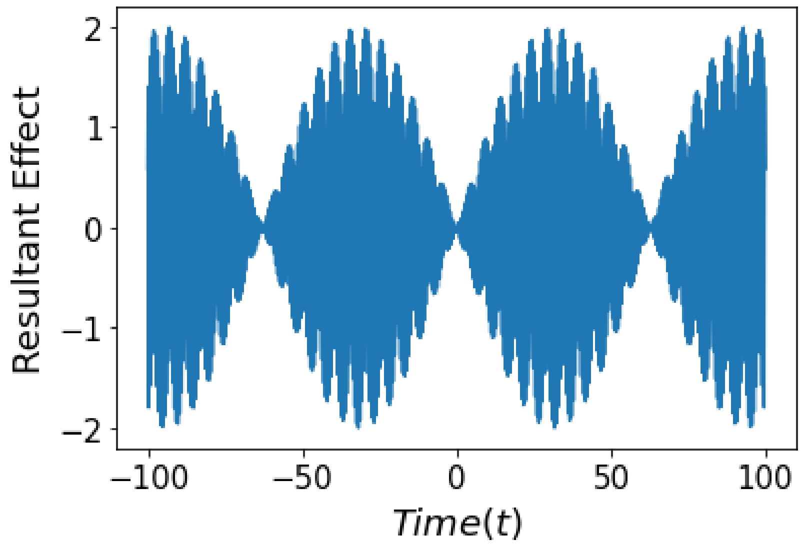

Fig. 7d: Resultant Effect

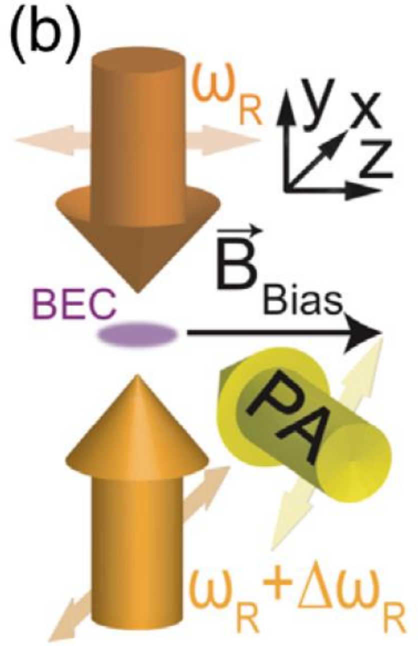

Fig. 7a: Experimental Setup