Supplemental Materials:
Observation of Quantum Interference and Coherent Control in a Photo-Chemical Reaction

David B. Blasing,[1] Jesús Pérez-Ríos,[2] Yangqian Yan,[1] Sourav Dutta,[1,3], Chuan-Hsun Li,[4] Qi Zhou,[1] and Yong P. Chen[1,4,5]
[1]*Department of Physics and Astronomy, Purdue University, West Lafayette, IN 47907*
[2]*School of Materials Sciences and Technology, Universidad del Turabo, Gurabo, Puerto Rico 00778*
[3]*Department of Physics, Indian Institute of Science Education and Research, Bhopal 462066, India*
[4]*School of Electrical and Computer Engineering, Purdue University, West Lafayette, IN 47907*
[5]*Purdue Quantum Center, Purdue University, West Lafayette, IN 47907*
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For the dressed atoms scattering in the $|F=0, m_F=0>$ channel, the stimulated transition rate to the excited molecular state is

$$\Gamma_{sup} \propto |\langle\phi_m(\vec{r}_{ab})|\langle F=0, m_F=0|\psi_{\text{scat}}\rangle|^2 \tag{1}$$

where $|\psi_{\text{scat}}\rangle$ denotes two body scattering wavefunction of the colliding atoms, including both the spin and relative spatial portions. The operator $\langle F=0, m_F=0|$ selects the spatial portion of the scattering wavefunction with total spin $|F=0, m_F=0\rangle$ (the only total spin channel contributing to the PA transition we chose). The spatial wavefunctions for molecule and the bare scattering state in the $|F=0, m_F=0\rangle$ channel are denoted as $\varphi_m(\vec{r}_{ab})$ and $\varphi_{F=0}(\vec{r}_{ab})$ respectively, both with relative coordinate

$$\vec{r}_{ab} = \vec{r}_a - \vec{r}_b, \tag{2}$$

where $\vec{r}_a$ and $\vec{r}_b$ denote the spatial coordinates of the two atoms. Since the BECs in the experiment are always loaded to the single particle ground state with quasimomentum $\vec{q}=(0, q_{min}, 0)$, the total wavefunction of one particle (denoted with a subscript) is

$$C_0 e^{i\vec{q}\cdot\vec{r}_a}|1,0\rangle_a + C_1 e^{i(\vec{q}+\vec{k}_r)\cdot\vec{r}_a}|1,1\rangle_a + C_{-1}e^{i(\vec{q}-\vec{k}_r)\cdot\vec{r}_a}|1,-1\rangle_a, \tag{3}$$

where $\vec{k}_r = (0, -2k_r, 0)$. The product state of two such particles, denoted a and b respectively, is written as

$$\begin{aligned}
&e^{i\vec{q}\cdot(\vec{r}_a+\vec{r}_b)}\big(C_0^2|1,0\rangle_a|1,0\rangle_b + C_1C_{-1}e^{i\vec{k}_r\cdot(\vec{r}_a-\vec{r}_b)}|1,1\rangle_a|1,-1\rangle_b + C_1C_{-1}e^{i\vec{k}_r\cdot(\vec{r}_b-\vec{r}_a)}|1,-1\rangle_a|1,1\rangle_b + ...\big)\\
=&e^{i\vec{q}\cdot(\vec{r}_a+\vec{r}_b)}\big[C_0^2(\sqrt{\tfrac{2}{3}}|2,0\rangle - \sqrt{\tfrac{1}{3}}|0,0\rangle)\\
&+ C_1C_{-1}e^{i\vec{k}_r\cdot(\vec{r}_a-\vec{r}_b)}(\sqrt{\tfrac{1}{6}}|2,0\rangle - \sqrt{\tfrac{1}{2}}|1,0\rangle + \sqrt{\tfrac{1}{3}}|0,0\rangle)\\
&+ C_1C_{-1}e^{i\vec{k}_r\cdot(\vec{r}_b-\vec{r}_a)}(\sqrt{\tfrac{1}{6}}|2,0\rangle + \sqrt{\tfrac{1}{2}}|1,0\rangle + \sqrt{\tfrac{1}{3}}|0,0\rangle) + ...\big]\\
=&e^{i\vec{q}\cdot(\vec{r}_a+\vec{r}_b)}[-\sqrt{\tfrac{1}{3}}C_0^2 + \sqrt{\tfrac{1}{3}}C_1C_{-1}e^{i\vec{k}_r\cdot(\vec{r}_b-\vec{r}_a)} + \sqrt{\tfrac{1}{3}}C_1C_{-1}e^{i\vec{k}_r\cdot(\vec{r}_a-\vec{r}_b)}\big]|0,0\rangle + \dots\\
=&e^{i\vec{q}\cdot(\vec{r}_a+\vec{r}_b)}[-\sqrt{\tfrac{1}{3}}C_0^2 + \sqrt{\tfrac{1}{3}}C_1C_{-1}e^{-i\vec{k}_r\cdot\vec{r}_{ab}} + \sqrt{\tfrac{1}{3}}C_1C_{-1}e^{i\vec{k}_r\cdot\vec{r}_{ab}}\big]|0,0\rangle + \dots.
\end{aligned} \tag{4}$$

where kets with subscripts, a or b, denote the spin states of the two single atoms respectively, and the ones without subscripts correspond to the total spins of two particles. In our model, $|\psi_{\text{scat}}\rangle$ then is

$$[-\sqrt{\tfrac{1}{3}}C_0^2 + \sqrt{\tfrac{1}{3}}C_1C_{-1}e^{-i\vec{k}_r\cdot\vec{r}_{ab}} + \sqrt{\tfrac{1}{3}}C_1C_{-1}e^{i\vec{k}_r\cdot\vec{r}_{ab}}\big]\varphi_{F=0}(\vec{r}_{ab})|0,0\rangle + \dots. \tag{5}$$

where we have multiplied the corresponding terms in the product state by $\varphi_{F=0}(\vec{r}_{ab})$, the relevant spatial wavefunction for the $F=0$ channel, and, we have also suppressed the center of mass term $e^{i\vec{q}\cdot(\vec{r}_a+\vec{r}_b)}$ as it is an overall phase. The

... denotes projections of the scattering state that does not contribute to the PA transition we chose. To describe our experiment, we project $|\psi_{\rm scat}\rangle$ to include only the portion with $|F=0, m_F=0\rangle$:

$$\langle F=0, m_F=0|\psi_{\rm scat}\rangle = [-\sqrt{\frac{1}{3}}C_0^2 + \sqrt{\frac{1}{3}}C_1C_{-1}e^{-i\vec{k}_r\cdot\vec{r}_{ab}} + \sqrt{\frac{1}{3}}C_1C_{-1}e^{i\vec{k}_r\cdot\vec{r}_{ab}}]\varphi_{F=0}(\vec{r}_{ab}) \tag{6}$$

Therefore according to Eq. (1) we have

$$\Gamma_{sup}\propto\Big|-\frac{C_0^2}{\sqrt{3}}\left(\int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})\right)+\frac{C_1C_{-1}}{\sqrt{3}}\left(\int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})e^{i\vec{k}_r\cdot\vec{r}_{ab}}+\int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})e^{-i\vec{k}_r\cdot\vec{r}_{ab}}\right)\Big|^2, \tag{7}$$

where inside the integrals representing the Franck-Condon overlap, we have used $\varphi_{F=0}(\vec{r}_{ab})$, the bare spatial wavefunction for scattering along $F=0$, and the additional phases associated with the Raman beams weighted by the appropriate superposition coefficients. This is justifiable since the size of our molecule is $\sim 10^{-3}\lambda_R$ (recall that $\lambda_R = 2\pi/k_r \approx 15000\,a_0$), so $\vec{k}_r\cdot\vec{r}_{ab}$ is negligibly small. Since the Franck-Condon overlap integrals are determined only by the short-range behavior (relative to the λ_R scale), then $\int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})e^{i\vec{k}_r\cdot\vec{r}_{ab}} \approx \int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})e^{-i\vec{k}_r\cdot\vec{r}_{ab}} \approx \int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})$, therefore,

$$\Gamma_{sup}\propto\frac{1}{3}\Big|\int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})\Big|^2\Big|-C_0^2+2C_1C_{-1}\Big|^2. \tag{8}$$

If we let the stimulated rate for two particles with atomic spin state $|f=0, m_f=0\rangle$ (like when $C_0=1$, and $C_{\pm1}=0$) be denoted by $\Gamma_{0,0}$ and note that they have a projection along $|F=0, m_F=0\rangle$ with CG coefficient $1/\sqrt{3}$, then:

$$\Gamma_{0,0}\propto\frac{1}{3}\Big|\int d\vec{r}_{ab}\varphi_m^*(\vec{r}_{ab})\varphi_{F=0}(\vec{r}_{ab})\Big|^2. \tag{9}$$

Therefore, $\Gamma_{sup}/\Gamma_{0,0} = |-C_0^2+2C_1C_{-1}|^2$. Recalling that $k_{sup}\propto\Gamma_{sup}$ (with a proportional factor here, as well as that in Eq. (1), that do not depend on the spin states of the colliding atoms [1]), we may then obtain Eq. 3 in the main text

$$k_{sup}/k_{0,0} = |C_0^2|^2 + 4|C_{-1}C_{+1}|^2 - 4{\rm Re}[C_0^2C_{-1}^*C_{+1}^*]. \tag{10}$$

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[1] M. Theis, G. Thalhammer, K. Winkler, M. Hellwig, G. Ruff, R. Grimm, and J. Hecker Denschlag, *Phys. Rev. Lett.*, 93, 12300 (2004).