Double-Slit Interference of Two-Electron Wave with Light Phase

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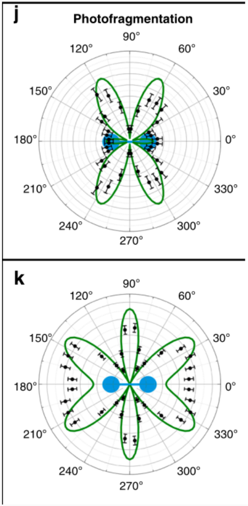
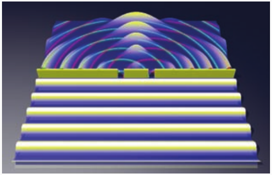
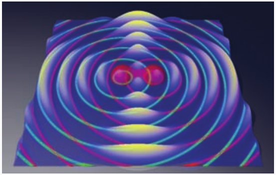
The simplest natural double-slit: H_2







Single ionization to $2s\sigma_g/2p\sigma_u$ state

$$H_2 : |L\rangle\,|R\rangle + |R\rangle\,|L\rangle\,,$$

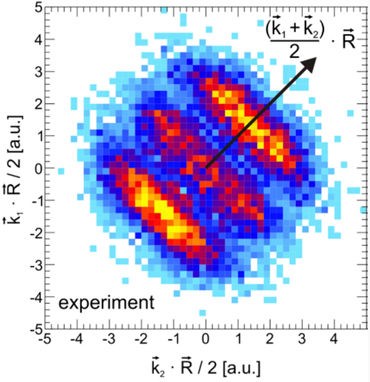
$$H_2^{\;+} : \sigma_g = |L\rangle + |R\rangle \qquad \rightarrow e^{-ip\cdot r_R} + e^{-ip\cdot r_L},$$

$$\sigma_u = |L\rangle - |R\rangle \qquad \rightarrow e^{-ip\cdot r_R} - e^{-ip\cdot r_L}.$$

The phase information of slits are encoded into the interference pattern.

Akoury, D. *et al.* *Science* **318**, 949 (2007)
Waitz, M. *et al.* *Nat. Comm.* **8**, 2266 (2017)

Double-slit interference of entangle electron pair



For one-photon double ionization process, two electrons emit from the same nuclear as an entangled pair.

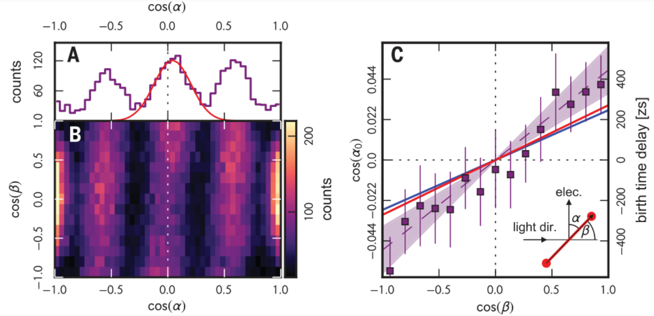
$$|L, p_{e1}\rangle\,|L, p_{e2}\rangle + |R, p_{e1}\rangle\,|R, p_{e2}\rangle$$

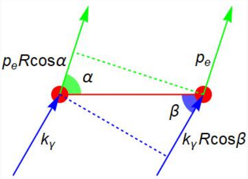
$$\rightarrow e^{i(p_{e1}+p_{e2})\cdot r_L} + e^{i(p_{e1}+p_{e2})\cdot r_R}$$

$$\rightarrow 1 + \cos[(p_{e1} + p_{e2}) \cdot R]$$

Kreidi, K. *et al.* *Phys. Rev. Lett.* **100**, 133005 (2008)
Horner, D. A. *et al.* *Phys. Rev. Lett.* **101**, 183002 (2008)
Waitz, M. *et al.* *Phys. Rev. Lett.* **117**, 083002 (2016)

Travel time that light passing by the molecule





Light reaches two nuclear with a time difference $\Delta t = \hat{k}_\gamma \cdot R/c$, resulting an additional phase difference $\omega\Delta t = k_\gamma \cdot R$.

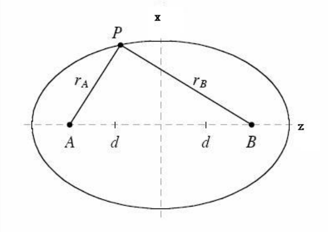
Grundmann, S. *et al. Science*, **370**, 339 (2020)

Towards our work

Question to be answered

- Significant deviation between experiment results and model prediction is found, what is the source?
- Experiment use the double ionization events in which the fast electron takes most of kinetic energy ($E_1/(E_1 + E_2) > 0.96$), and then integrate over the slow electron. So what it the electron correlation effect?

Numerical method: coordinate system



$$\xi = (r_A + r_B)/R, \quad \eta = (r_A - r_B)/R,$$

Expand two electron wavefunction into basis:

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2) = \sum_{\alpha l_1 m_1 \beta l_2 m_2} c_{\alpha l_1 m_1 \beta l_2 m_2} f_\alpha(\xi_1) Y_{l_1}^{m_1}(\eta_1, \phi_1) f_\beta(\xi_2) Y_{l_2}^{m_2}(\eta_2, \phi_2).$$

Re-collect the angular part into different subspaces:

$$\text{Parity} = (l_1 + l_2) \mod 2, \quad \text{total magnetic number} = m_1 + m_2,$$

which corresponding to field-free symmetry of H_2.

H. Liang, *et al. J. Phys. B: At. Mol. Opt. Phys.* **50**, 174002 (2017)

Numerical method: Hamiltonian

The field-free Hamiltonian include kinetic energy, electron-ion interaction and electron-electron interaction

$$T_{lm} = \frac{R}{4}\left[-\partial_\xi(\xi^2-1)\partial_\xi + \frac{m^2}{\xi^2-1} + l(l+1)\right],$$

$$V_{\mathrm{ei}} = -\frac{R^2}{2}\xi,$$

$$V^{\alpha l_1 m_1 \beta l_2 m_2}_{\alpha' l_1' m_1' \beta' l_2' m_2'} = -\delta_{\alpha'\alpha}\delta_{\beta'\beta}\sum_{LM}(-)^{M_{\mathrm{tot}}+M}\rho^{\alpha L,M}_{l_1 m_1 l_1' m_1'}\rho^{\beta L,-M}_{l_2 m_2 l_2' m_2'}[T^{-1}_{LM}]_{\alpha\beta}.$$

The laser-electron interaction is expanded into electric dipole, electric quadrapole and magnetic dipole term

$$H_{\mathrm{int}} = r\cdot E_0(t) - \frac{\hat{k}\cdot r}{2c}[r\cdot\partial_t E_0(t)] + \frac{1}{2c}L\cdot[\hat{k}\times E_0(t)] + O\left(\frac{1}{c^2}\right),$$

L. Tao, C. W. McCurdy, and T. N. Rescigno, *Phys. Rev. A* **82**, 023423 (2010).
H. Liang, *et al.* *Phys. Rev. A* **98**, 063413 (2018).

Numerical method: propagator

Shrödinger's equation for one-photon channel

$$(\mathrm{i}\partial_t - H_0)\,|\psi\rangle = H_{\text{int}}(t)\,|i\rangle\,,$$

can be departed into short time propagators

$$\begin{aligned}|\psi(t+\Delta t)\rangle &= \mathrm{e}^{-\mathrm{i}H_0\Delta t}\left(|\psi(t)\rangle - \mathrm{i}\int_0^{\Delta t}\mathrm{e}^{\mathrm{i}(H_0-E_i)\tau}H_{\text{int}}(t+\tau)\,|i\rangle\,\mathrm{d}\tau\right)\\ &\approx \mathrm{e}^{-\mathrm{i}H_0\Delta t}\,|\psi(t)\rangle + \frac{\mathrm{e}^{-\mathrm{i}(H_0-E_i)\Delta t}-1}{H_0-E_i}\overline{H_{\text{int}}(t+\tau)}\,|i\rangle\,.\end{aligned}$$

The exponential and inversion of H_0 are solved with Krylov subspace technique.

Numerical method: observable

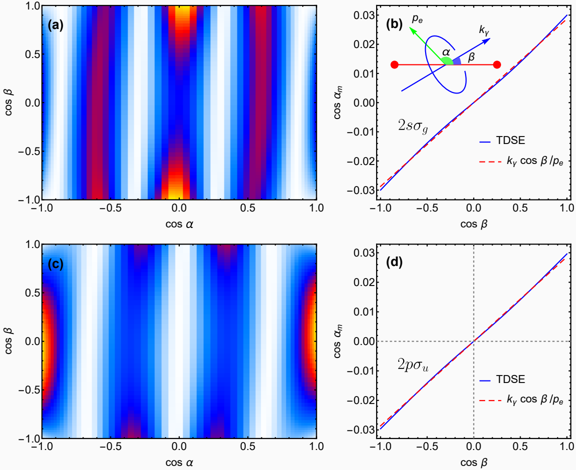
We use a 10-cycle pulse to ionize H_2 and wait for additional 2 cycles in case that e-e interaction could be ignored.

The final state is projected into the symmetric production of one-electron bound/continue states

$$A_{i\to p_1 p_2} = \frac{1}{\sqrt{2}}(\langle p_1| \langle p_2| + \langle p_2| \langle p_1|) |\psi_f\rangle ,$$

$$A_{i\to pe} = \frac{1}{\sqrt{2}}(\langle p| \langle e| + \langle e| \langle p|) |\psi_f\rangle ,$$

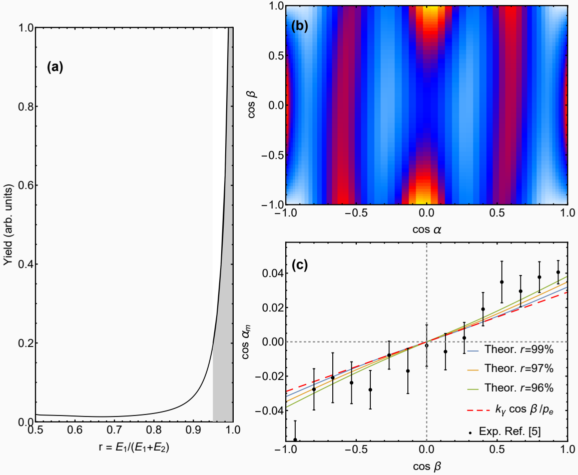
Numerical Results: Singly Ionization



Double-slit with phase difference

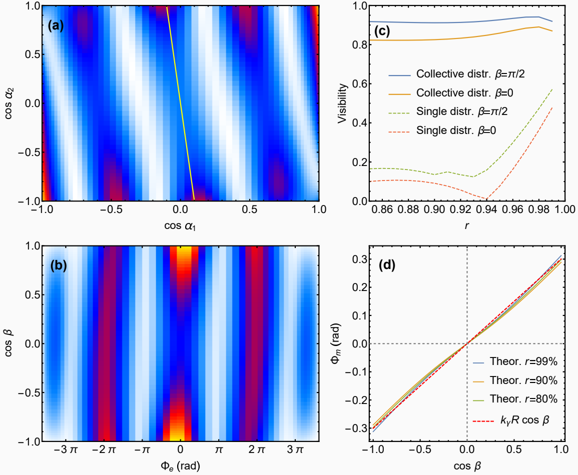
$$\Delta\phi = p_e R \cos\alpha - k_\gamma R \cos\beta$$

Numerical Result: Double Ionization Signal



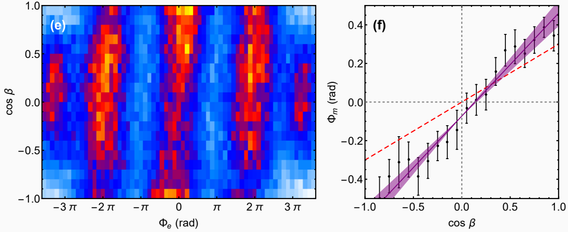
$$P_{\text{single}}(\cos\alpha_1) \equiv \int P_{\text{joint}}(\cos\alpha_1, \cos\alpha_2)\, \mathrm{d}\cos\alpha_2.$$

Numerical Results: Distribution on Collective Phase



$$P_{\text{collect}}(\Phi_e) \equiv \int P_{\text{joint}}(\cos\alpha_1, \cos\alpha_2) \times$$
$$\delta(\Phi_e - p_{e1} R \cos\alpha_1 - p_{e2} R \cos\alpha_2)\, \mathrm{d}\cos\alpha_1 \mathrm{d}\cos\alpha_2.$$

Re-Statistic of Experiment Data



- Visibility enhanced as expected.
- However, experiment data still deviates from the numerical results and model prediction, which calls for further investigations.

Recoil momentum to nuclear

One uses the Coulomb explosion fragments to determine the direction of molecular axis, the corresponding momentum

$$p_n \approx \sqrt{\frac{m_p}{R_e}} \approx 36\,\text{a.u.},$$

while the electron recoil momentum is

$$p_e \approx \sqrt{2\omega} \approx 8\,\text{a.u.},$$

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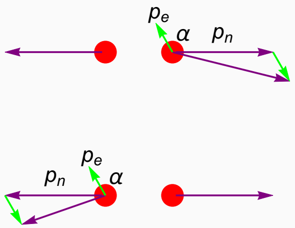

It gives a factor of

$$\sqrt{1 - \left(\frac{p_e}{2p_n}\right)^2} \approx 0.994,$$

thus cannot explain the existing difference.

What if electrons emit from different nuclei?

For two electron emitting from the same side, one can easily write down the interference

$$\langle L, k_\gamma|\,|L, p_{e1}\rangle\,|L, p_{e2}\rangle + \langle R, k_\gamma|\,|R, p_{e1}\rangle\,|R, p_{e2}\rangle$$
$$\to \mathrm{e}^{\mathrm{i}(p_{e1}+p_{e2}-k_\gamma)\cdot r_L} + \mathrm{e}^{\mathrm{i}(p_{e1}+p_{e2}-k_\gamma)\cdot r_R}$$
$$\to 1 + \cos[(p_{e1} + p_{e2} - k_\gamma)\cdot R],$$

but if them emit from different sides, things get confused

$$\langle ?, k_\gamma|\,|L, p_{e1}\rangle\,|R, p_{e2}\rangle + \langle ?, k_\gamma|\,|R, p_{e1}\rangle\,|L, p_{e2}\rangle$$
$$\to \mathrm{e}^{\mathrm{i}(p_{e1}\cdot r_L+p_{e2}\cdot r_R+?)} + \mathrm{e}^{\mathrm{i}(p_{e1}\cdot r_R+p_{e2}\cdot r_L+?)}$$
$$\to 1 + \cos[(p_{e1} - p_{e2} + ?)\cdot R].$$

Shake-off picture may gives an answer, but when energy partition rate continually varies from 0 to 1, problem still exists.

Summary

- We developed a program for two-electron time-dependent Schrödinger equation.
- Double-slit interference in one-photon ionization, including the effect of light propagation and electron correlation, can be reproduced.
- The remaining deviation between experiment data and theoretical prediction will trigger further investigations on this problem.

Liang, H. *et al. Phys. Rev. A*, **98**, 063413 (2018).
Liang, H. *et al. Phys. Rev. Lett.* under review.

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Thanks for listening!

Quantum model for nuclear motion

Consider the one-photon ionization of H_2^+. From the initial state

$$|i\rangle = \Psi(R_1 - R_2)[\psi(r - R_1) + \psi(r - R_2)],$$

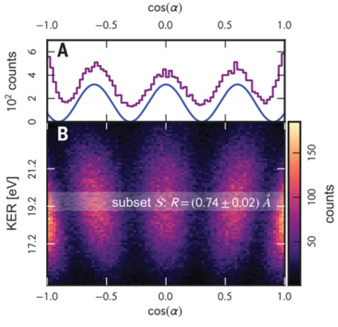
to a final state $|p\rangle = \mathrm{e}^{\mathrm{i}p\cdot r}$ with interaction term $\varepsilon \cdot p\mathrm{e}^{-\mathrm{i}k\cdot r}$.

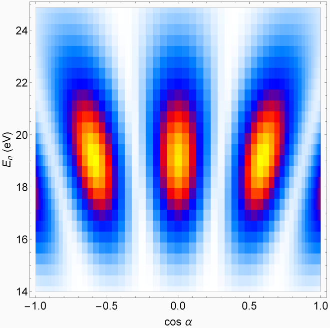
Thus, the final nuclear state is

$$\begin{aligned}\langle p|\varepsilon \cdot p\mathrm{e}^{-\mathrm{i}k\cdot r}|i\rangle =&(\varepsilon \cdot p)\Psi(R_1 - R_2)\phi(p - k)[\mathrm{e}^{-\mathrm{i}(p-k)\cdot R_1} + \mathrm{e}^{-\mathrm{i}(p-k)\cdot R_2}] \\ \propto&(\varepsilon \cdot p)\Psi(R_1 - R_2)\phi(p - k)\times \\ &\sum_l (2l+1) j_l(\frac{pR}{2}) \cos(\frac{k \cdot R}{2} - \frac{l\pi}{2}) P_l(\hat{p} \cdot \hat{k})\end{aligned}$$

By projecting it onto Coulomb continue of nuclear, one can reach the final joint distribution.

Quantum model for nuclear motion - Cont.





The KER dependence is nearly the same from the reflection approximation

$$\text{KER} = \frac{1}{R}.$$

And the $\cos\beta$ dependence is also unaffected by the nuclear motion.