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Classical trajectory perspective on double ionization dynamics of atoms and molecules irradiated by ultrashort intense laser pulses

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Thanks

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Outline of the talk

☆History review

☆Newest experimental developments

☆Difficulty of the existing theories

☆Our semi-classical model **New!**

☆Explanation of the experiments

☆Future perspective

☆Conclusion

1. J. Liu, D. F. Ye, J. Chen, X. Liu, Phys. Rev. Lett. 99, 013003 (2007)
2. D. F. Ye, J. Chen, J. Liu, Phys. Rev. A 77, 013403 (2008)
3. D. F. Ye, X. Liu, J. Liu, arXiv:0802.0041 (2008)

History review 1994

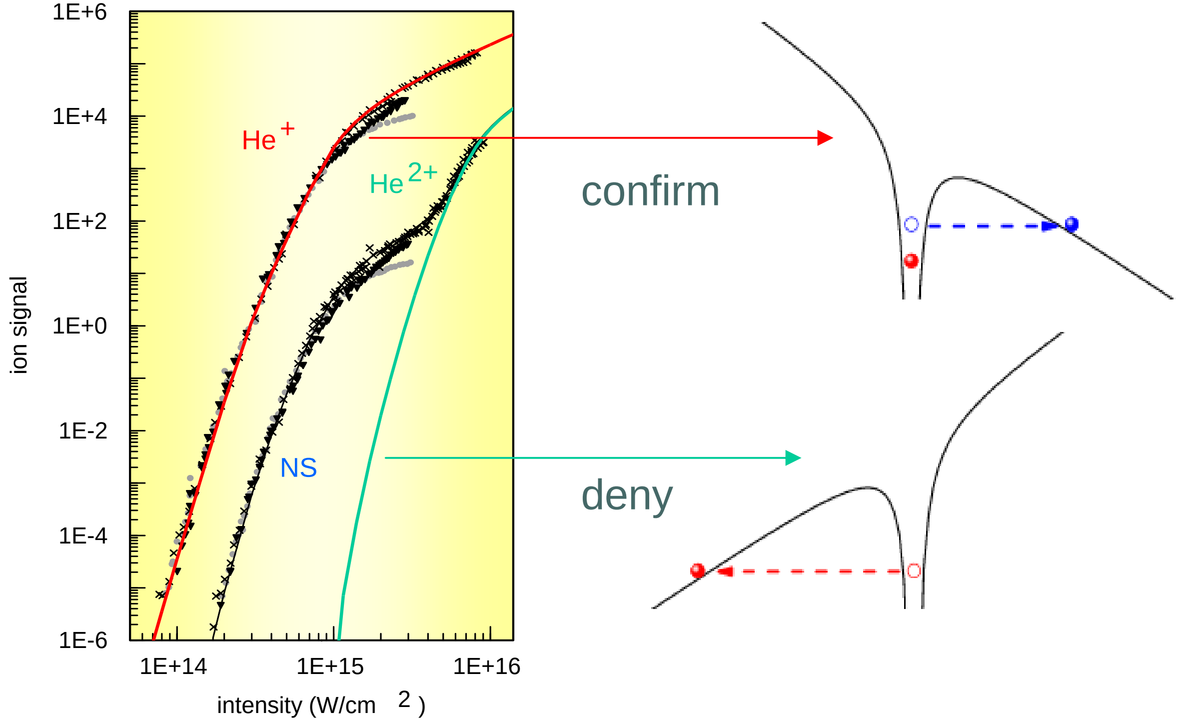
Single ionization

ADK theory √

Double ionization

No!

Experiments in the early 1990s showed considerably more double ionization than predicted by independent-electron model.



Electron correlation should be take into account !

B. Walker et al, Phys. Rev. Lett. 73, 1227 (1994)

History review ……2000

Direct evidence of electron correlation

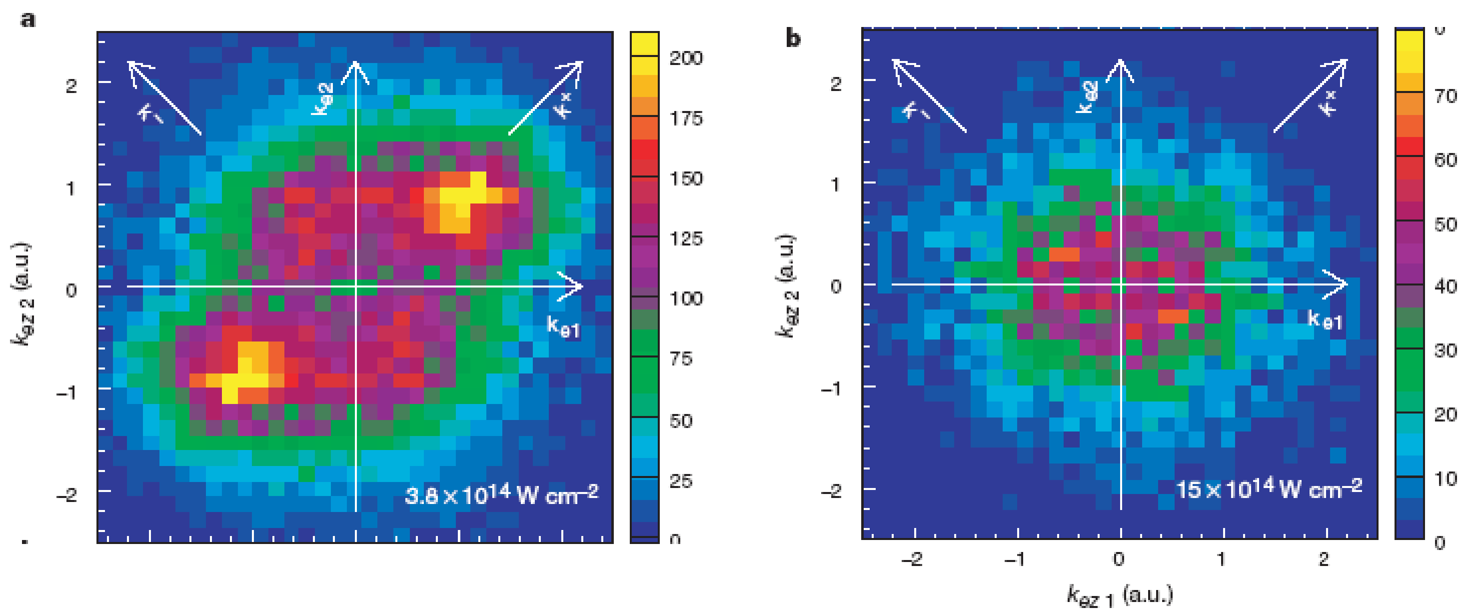


Figure 2 Momentum correlation between the two emitted electrons when an Ar^{2+} ion is produced in the focus of a 220-fs, 800-nm laser pulse at peak intensities of 3.8×10^{14} W cm^{-2} (**a**) and 15×10^{14} W cm^{-2} (**b**). The horizontal axis shows the momentum component of one electron along the polarization of the laser field; the vertical axis shows the same momentum component of the corresponding second electron. The same sign of the momenta for both electrons means emission to the same half sphere. The data are integrated over the momentum components in the direction perpendicular to the polarization. The colour coding shows the differential rate in arbitrary units.

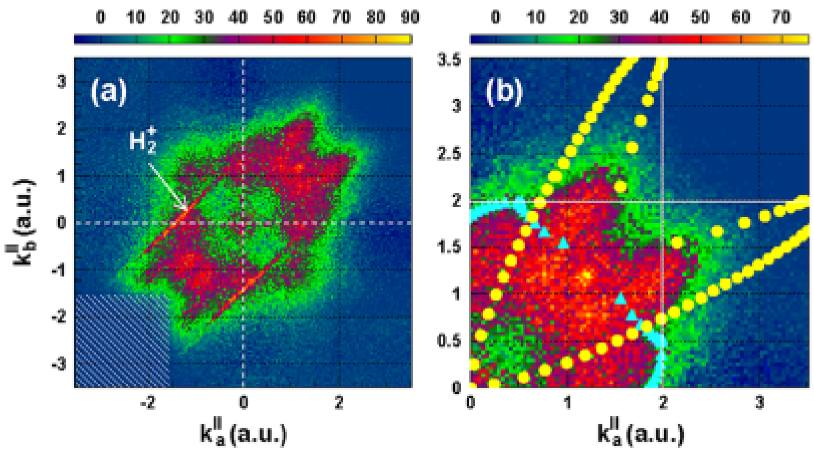
Th. Weber et al, Nature 405, 658 (2000)

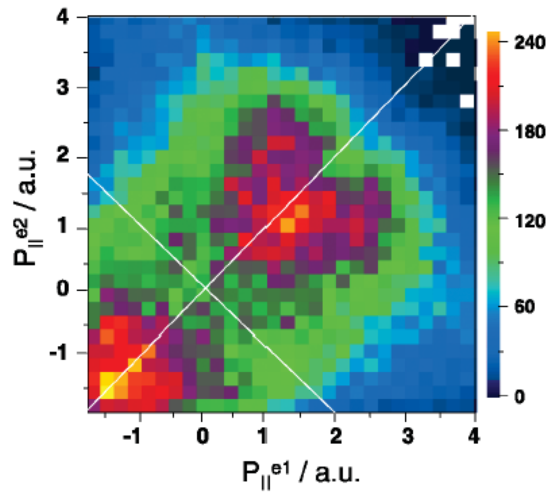
History review ……last day in 2007

PRL **99,** 263002 (2007) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2007

Binary and Recoil Collisions in Strong Field Double Ionization of Helium

A. Staudte,[1,*] C. Ruiz,[2] M. Schöffler,[3] S. Schössler,[3] D. Zeidler,[4] Th. Weber,[5] M. Meckel,[3] D. M. Villeneuve,[1] P. B. Corkum,[1] A. Becker,[2] and R. Dörner[3]





PRL **99,** 263003 (2007) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2007

Correlated Two-Electron Momentum Spectra for Strong-Field Nonsequential Double Ionization of He at 800 nm

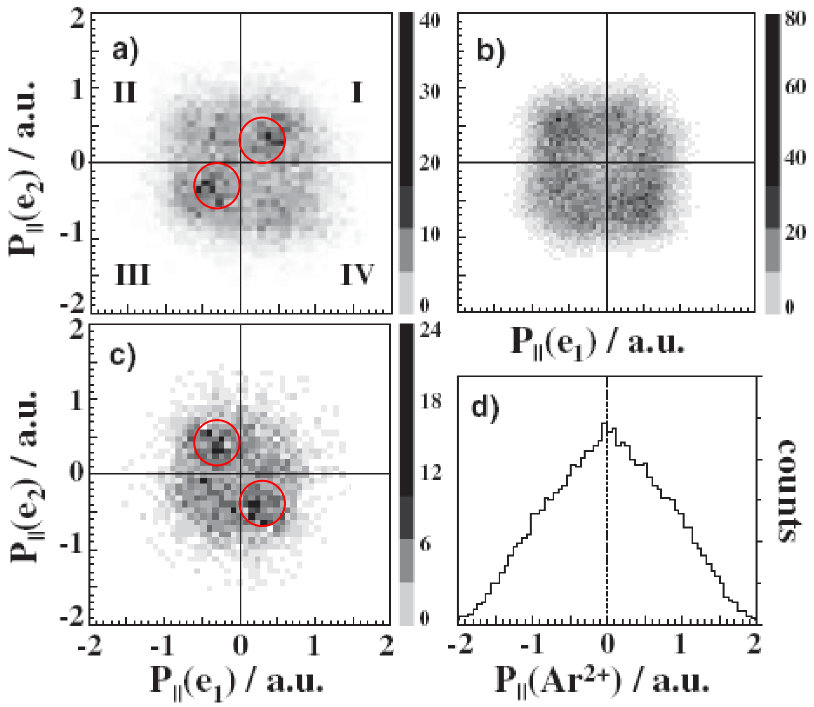
A. Rudenko,[1] V. L. B. de Jesus,[2] Th. Ergler,[1] K. Zrost,[1] B. Feuerstein,[1] C. D. Schröter,[1] R. Moshammer,[1] and J. Ullrich[1]

History review ……just 2 months ago

PRL **101**, 053001 (2008) PHYSICAL REVIEW LETTERS week ending 1 AUGUST 2008

Strong-Field Double Ionization of Ar below the Recollision Threshold

Yunquan Liu,[1] S. Tschuch,[1] A. Rudenko,[1] M. Dürr,[1] M. Siegel,[2] U. Morgner,[2] R. Moshammer,[1] and J. Ullrich[1]



Back-to-Back emission dominates

when the laser field is so weak that even the most energetic electron with maximum returned energy 3.17Up can not directly free the inner electron.

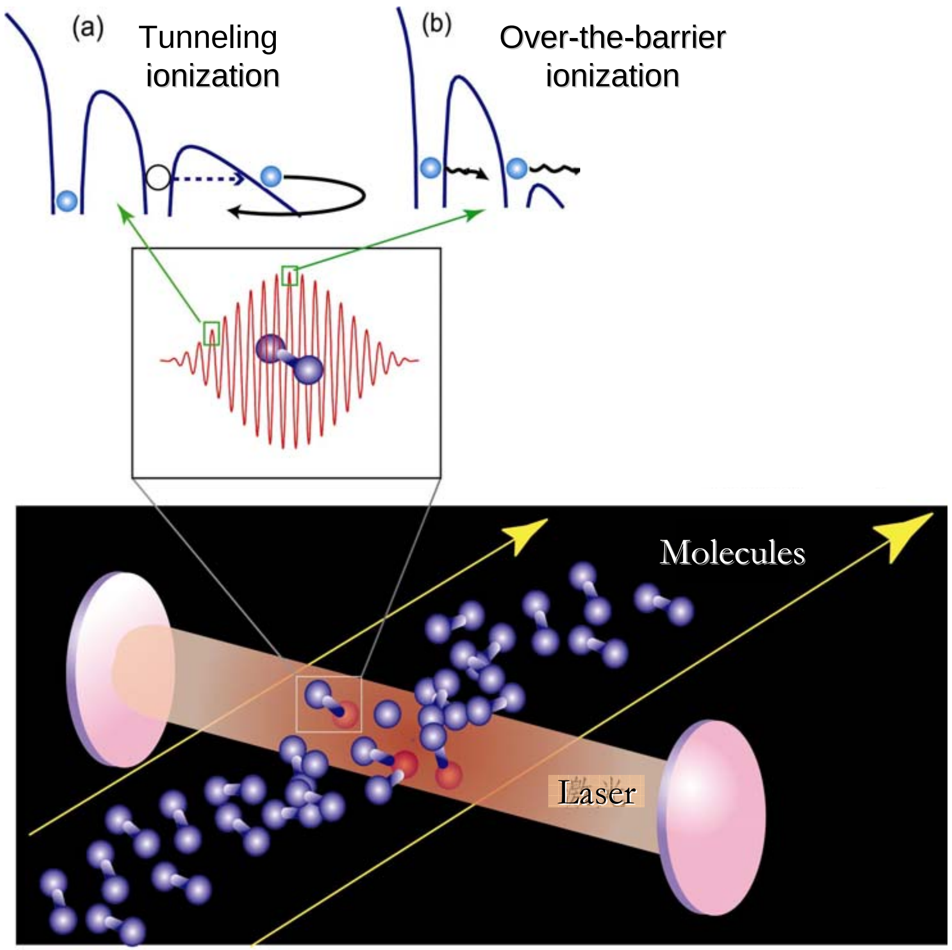
FIG. 2. Correlated longitudinal momentum spectra $P_{\|}(e_1)$ vs $P_{\|}(e_2)$ for Ar double ionization. (a) 9×10^{13} W/cm^2 [11]. (b) 7×10^{13} W/cm^2. (c) 4×10^{13} W/cm^2. (d) Longitudinal momentum distribution of Ar^{2+} ions at 7×10^{13} W/cm^2.

Theoretical Dificulties

• 1. Perturbation theory	The laser field has reached or even exceeded the Coulomb attraction	✗
• 2. 3D quantum calculation	Far exceed the capability even of the best computer	✗
• 3. 1D quantum calculation	Do not include effects such as Coulomb focusing	✗
• 4. Classical MC calculation	Do not include effects such as quantum tunneling	✗

- S-Matrix, Floque theory ……

Semiclassical model (Phys.Rev.Lett. 99, 013003, 2007)



Overview

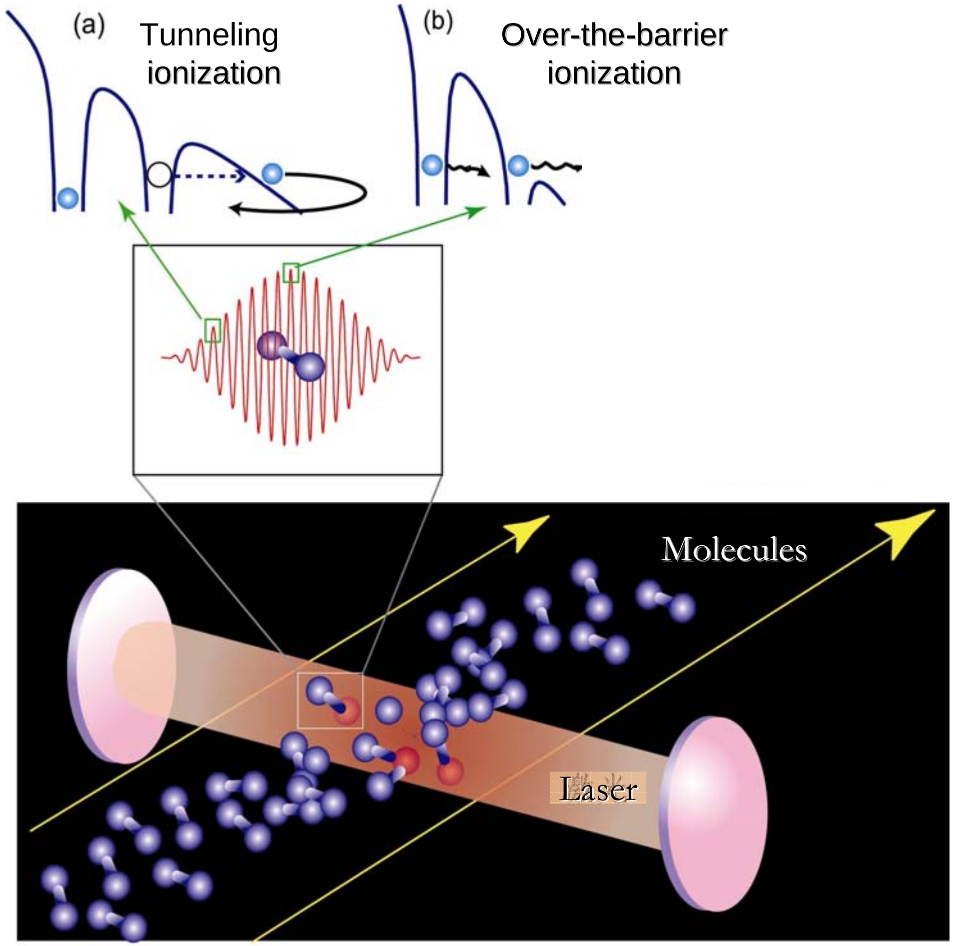
The model is based upon the rescattering picture

It has included all the effects that determine the DI process
- quantum tunneling
- Coulomb focusing
- ……

while keeps the computational capacity still accessible.

The dynamics of two electrons are governed by Newton's equations.

Semiclassical model … continued



Newton's equation

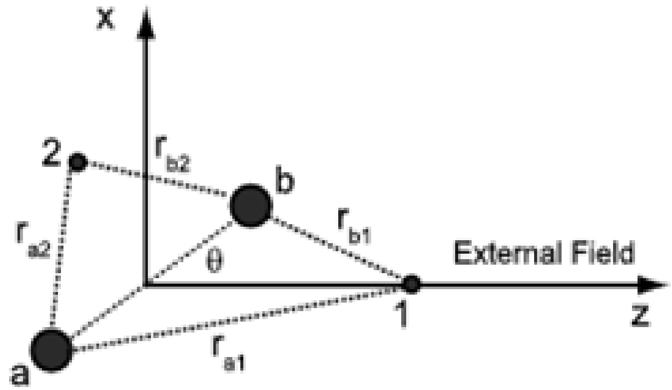
$$\frac{d^2\mathbf{r}_i}{dt^2} = \mathbf{E}(t) - \nabla(V_{ne} + V_{ee})$$

$$\mathbf{E}(t) = (\varepsilon(t)sin\theta, 0, \varepsilon(t)cos\theta)$$

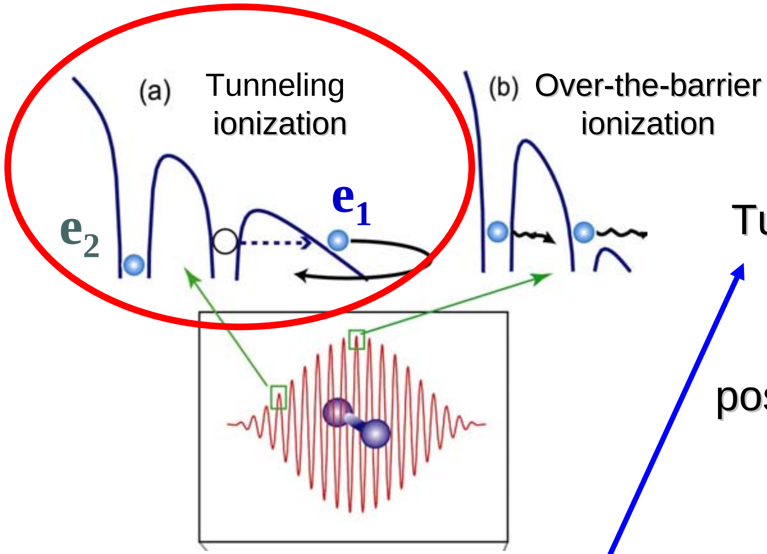
$$\varepsilon(t) = \varepsilon cos(\omega t).$$

$$V_{ne}^{i} = -\frac{Z_{eff}}{r_{ai}} - \frac{Z_{eff}}{r_{bi}};$$

$$V_{ee} = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}.$$



Semiclassical model … continued



J. Chen, J. Liu, L. B. Fu, and W. M. Zheng Phys. Rev. A **63**, 011404(R) (2000); Li-Bin Fu, Jie Liu, Jing Chen, and Shi-Gang Chen Phys. Rev. A **63**, 043416 (2001); Li-Bin Fu, Jie Liu, and Shi-Gang Chen Phys. Rev. A **65**, 021406(R) (2002); J. Chen, J. Liu, and W. M. Zheng Phys. Rev. A **66**, 043410 (2002).

R. Abrines and I.C. Percival, Proc. Phys. Soc. London **88**, 861 (1966); J. G. Leopold and I. C. Percival, J. Phys. B **12**, 709 (1979).

Initial distribution

(a) Tunneling regime

Tunneled electron e_1

position
$$\begin{cases} -\frac{1}{r_{a1}} - \frac{1}{r_{b1}} + \int \frac{\left|\Psi(\mathbf{r}')\right|^2}{|\mathbf{r}_1 - \mathbf{r}'|} d\mathbf{r}' + I_{p1} - z_1 \varepsilon(t_0) = 0, \\ x_1 = y_1 = 0 \end{cases}$$

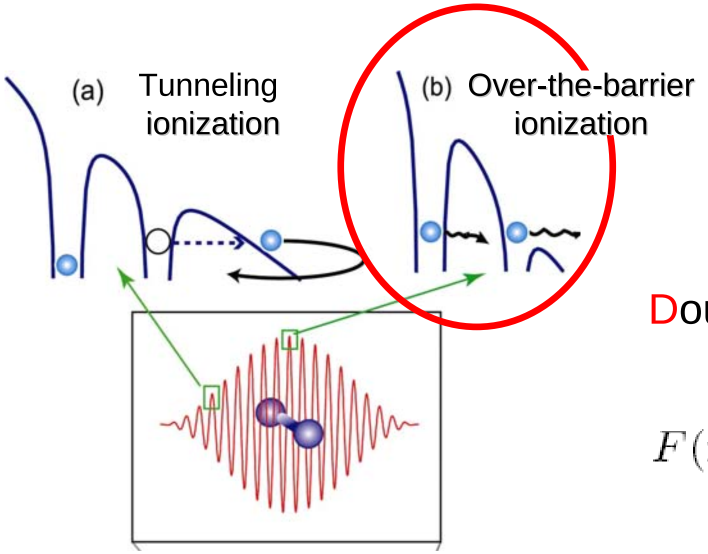
momentum
$$\begin{cases} (v_\perp \cos\varphi, v_\perp \sin\varphi, 0) \\ w(v_\perp) dv_\perp = \frac{2(2I_{p1})^{1/2} v_\perp}{\varepsilon(t_0)} \exp\left(-\frac{v_\perp^2 (2I_{p1})^{1/2}}{\varepsilon(t_0)}\right) dv_\perp \end{cases}$$

Bound electron e_2

Single electron Microcanonical Distribution（SMD）

position + momentum
$$\begin{cases} F(\mathbf{r}_2, \mathbf{p}_2) = k\delta[I_{p2} - \mathbf{p}_2^2/2 - W(r_{a2}, r_{b2})] \\ W(r_{a2}, r_{b2}) = -1/r_{a2} - 1/r_{b2} \end{cases}$$

Semiclassical model … continued



Initial distribution

(b) Over-the-barrier regime

Double electron Microcanonical Distribution（DMD）

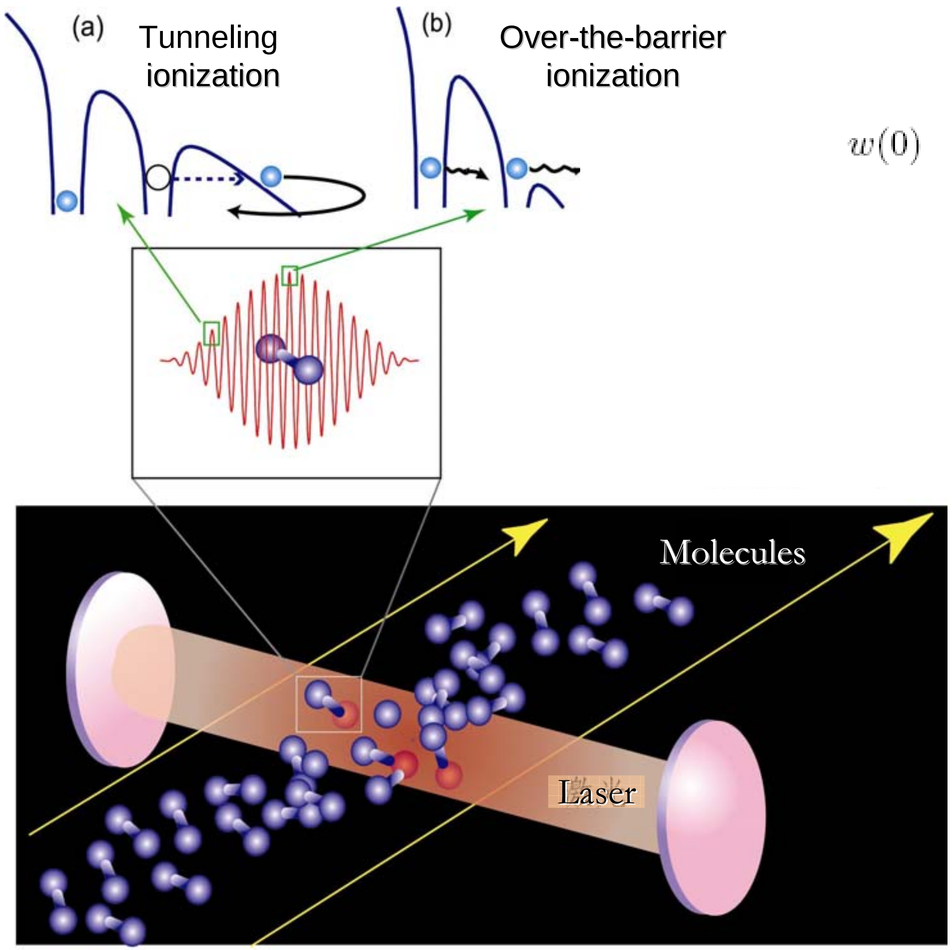
$$F(\mathbf{r}_1, , \mathbf{r}_2, \mathbf{p}_1, \mathbf{p}_2) = \frac{1}{2}[f_\alpha(\mathbf{r}_1, \mathbf{p}_1) f_\beta(\mathbf{r}_2, \mathbf{p}_2) + f_\beta(\mathbf{r}_1, \mathbf{p}_1) f_\alpha(\mathbf{r}_2, \mathbf{p}_2)],$$

$$f_{\alpha,\beta}(\mathbf{r}, \mathbf{p}) = k\delta[I_{p1} - \frac{\mathbf{p}^2}{2} - W(r_a, r_b) - V_{\alpha,\beta}(\mathbf{r})],$$

$$V_{\alpha,\beta}(\mathbf{r}) = \frac{1}{r_{b,a}}[1-(1+\kappa r_{b,a})e^{-2\kappa r_{b,a}}].$$

L. Meng, C. O. Reinhold and R. E. Olson, Phys. Rev. A **40** , 3637 (1989).

Semiclassical model … continued



Each event has different weight → molecular ADK formula:

$$w(0) = \frac{16 \cdot c_1^2 (2I_{p1})^{5/2}}{\varepsilon^2} e^{-\kappa R} \int_0^{2\pi} d\phi \int_0^1 \cosh^2\left(\frac{\kappa R \mu}{2}\right) \frac{\mu d\mu}{[\sin\theta\cos\phi(1-\mu^2)^{1/2} + \mu\cos\theta]^2}$$

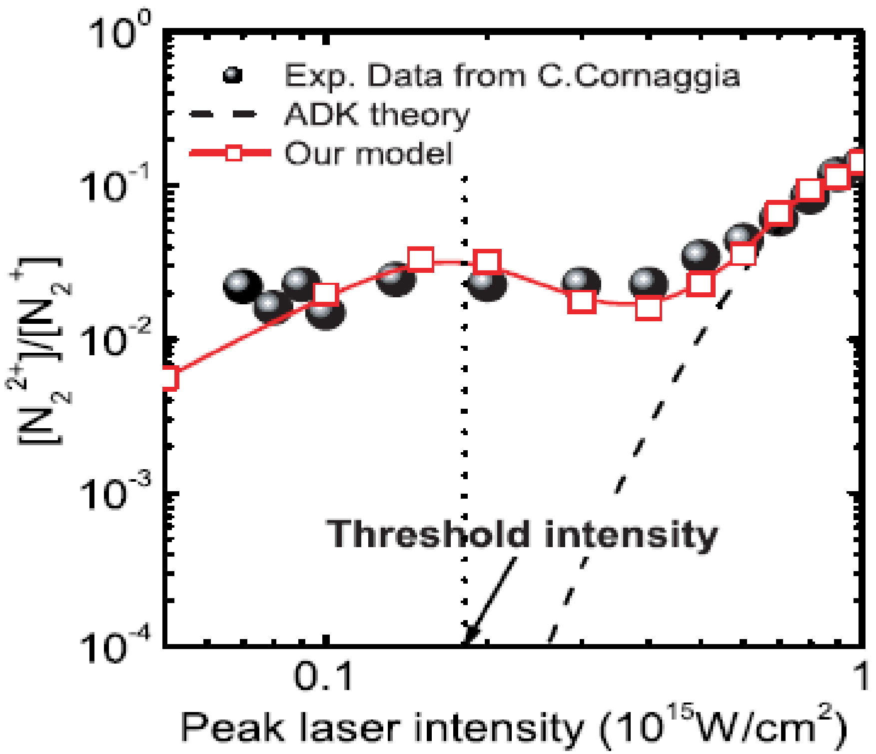
$$\times \quad \exp\left[-\frac{2}{3\varepsilon}\frac{(I_{p1})^{3/2}}{\sin\theta\cos\phi(1-\mu^2)^{1/2} + \mu\cos\theta}\right]$$

NOTE: The total ionization rate should be summed over the

- pulse duration
- interaction volume
- alignment angle

in order to compare with the experimental data.

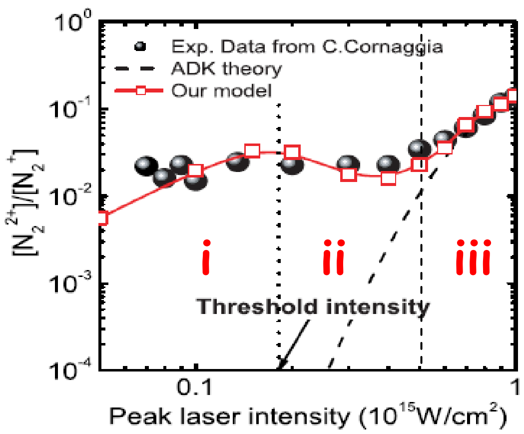
First sight of the model calculation

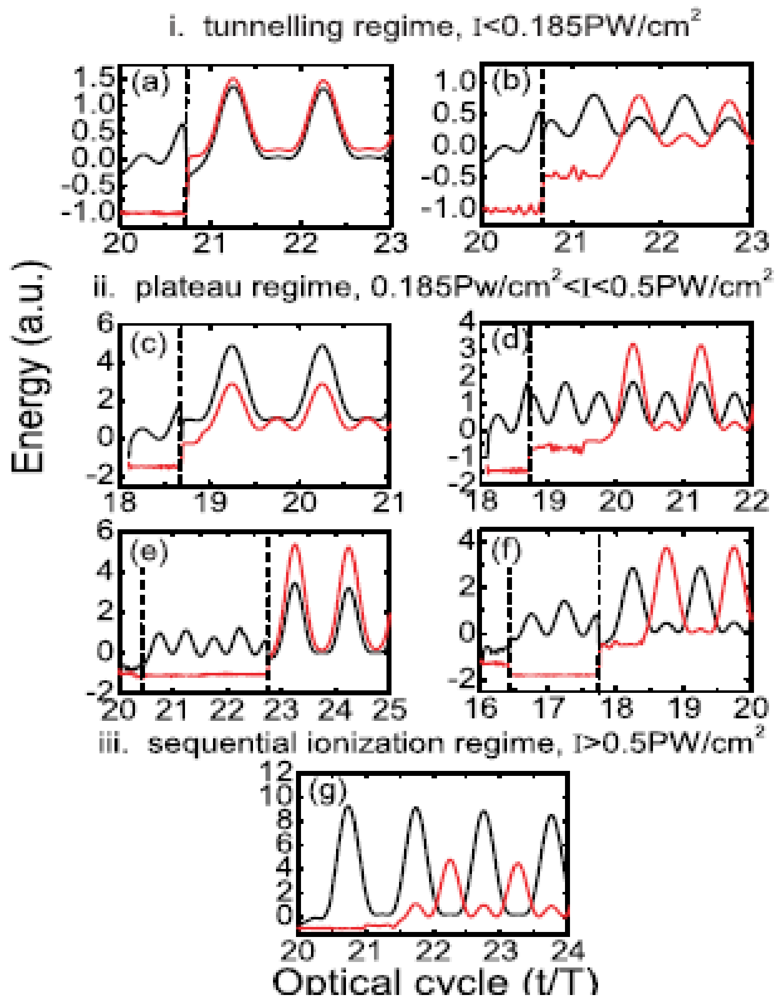


Good Agreement!

C. Cornaggia and Ph.Hering
Phys. Rev. A 62, 023403 (2000)

Subcycle dynamics





(i) Tunneling regime:

collision-ionization (CI)

collision-excitation-ionization (CEI)

(ii) Plateau regime:

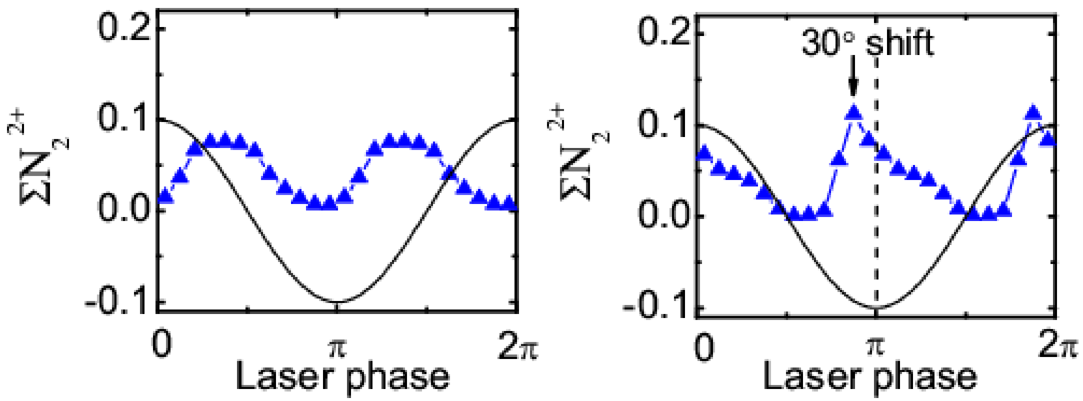
trajectories are much more complicated.

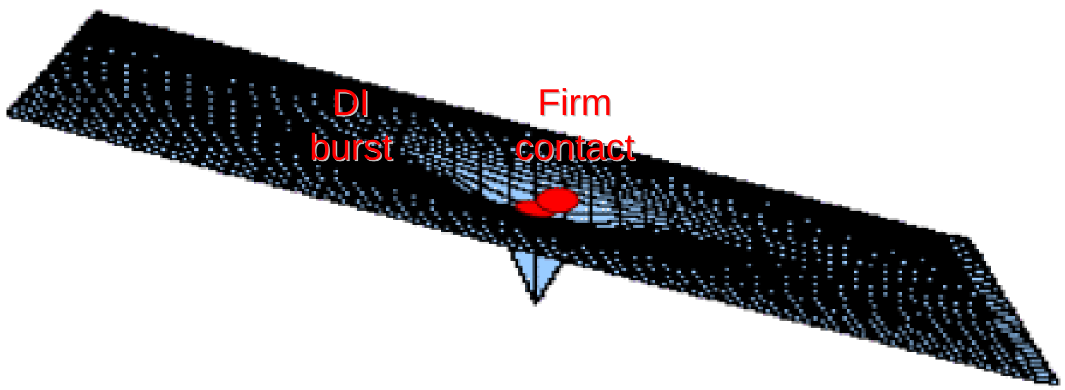
multiple-collision trajectories

(iii) Sequential ionization regime:

ionize independently

Laser phase: at firm contact & DI burst





Two most important instant

Most electron pairs recollide at the laser zero, that is because when the returned electons come back at this time, it obtain the largest kinetic energy, thus would cause double ionization more easily.

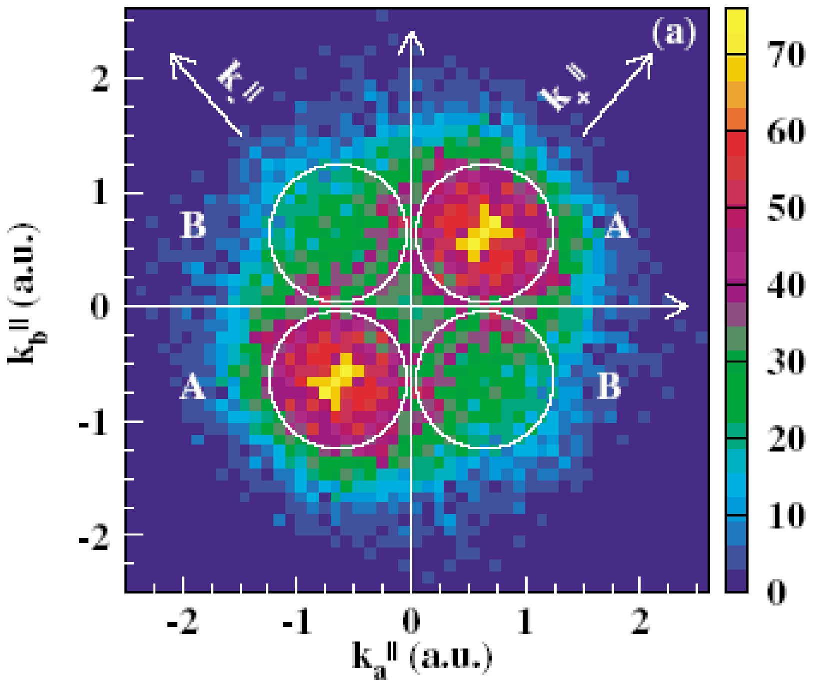
DI occurs a little later, about 30º before the laser peak, leading to the emission of electron pairs with nonzero momentum and in the same direction (e-e correlation), see next slide.

30º shift

VOLUME 92, NUMBER 21 PHYSICAL REVIEW LETTERS week ending 28 MAY 2004

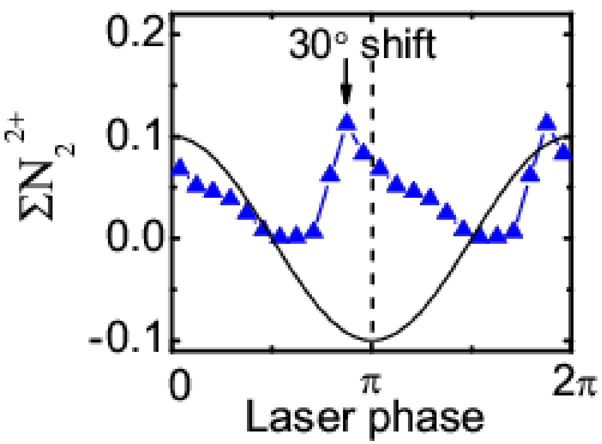
Fully Differential Rates for Femtosecond Multiphoton Double Ionization of Neon

M. Weckenbrock,[1] D. Zeidler,[2] A. Staudte,[1] Th. Weber,[1] M. Schöffler,[1] M. Meckel,[1] S. Kammer,[1] M. Smolarski,[1] O. Jagutzki,[1] V. R. Bhardwaj,[2] D. M. Rayner,[2] D. M. Villeneuve,[2] P. B. Corkum,[2] and R. Dörner[1,*]

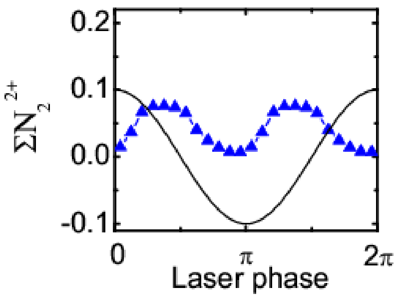


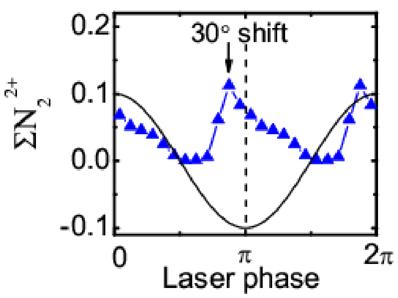
argon [23]). We also find that the two electrons often have similar momentum in the direction parallel to the laser field. Assuming that this momentum is primarily obtained from the laser field, we deduce that both electrons are reemitted at about 30° before or after the maximum of the laser field. Electrons that have similar

$$k^{\|}_{a,b} = 2\sqrt{U_p}\,\sin\omega t_{\rm ion}.$$

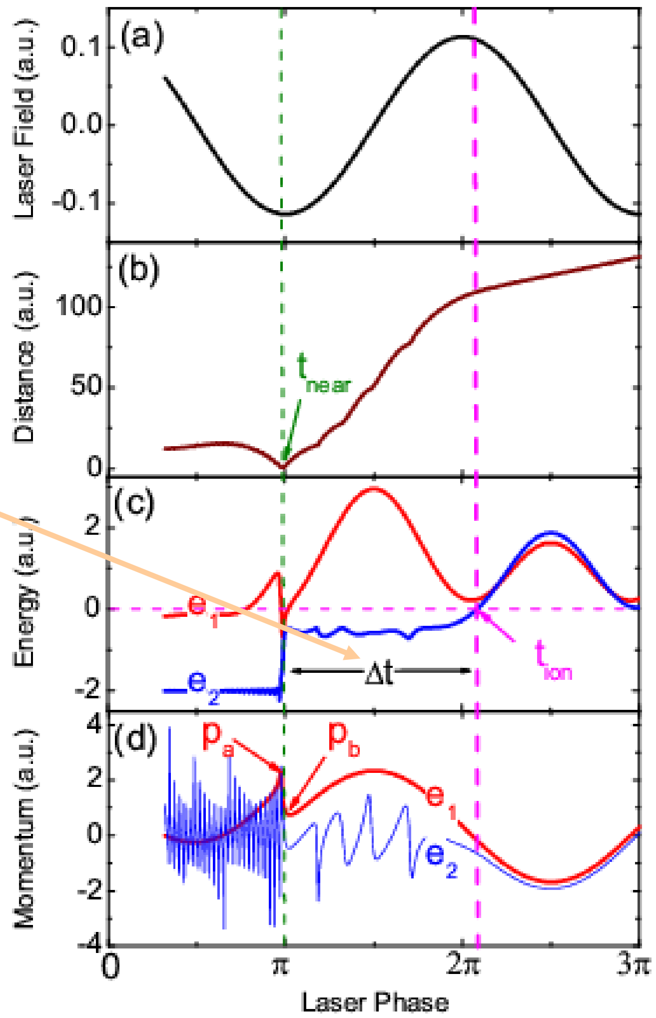


Time delay





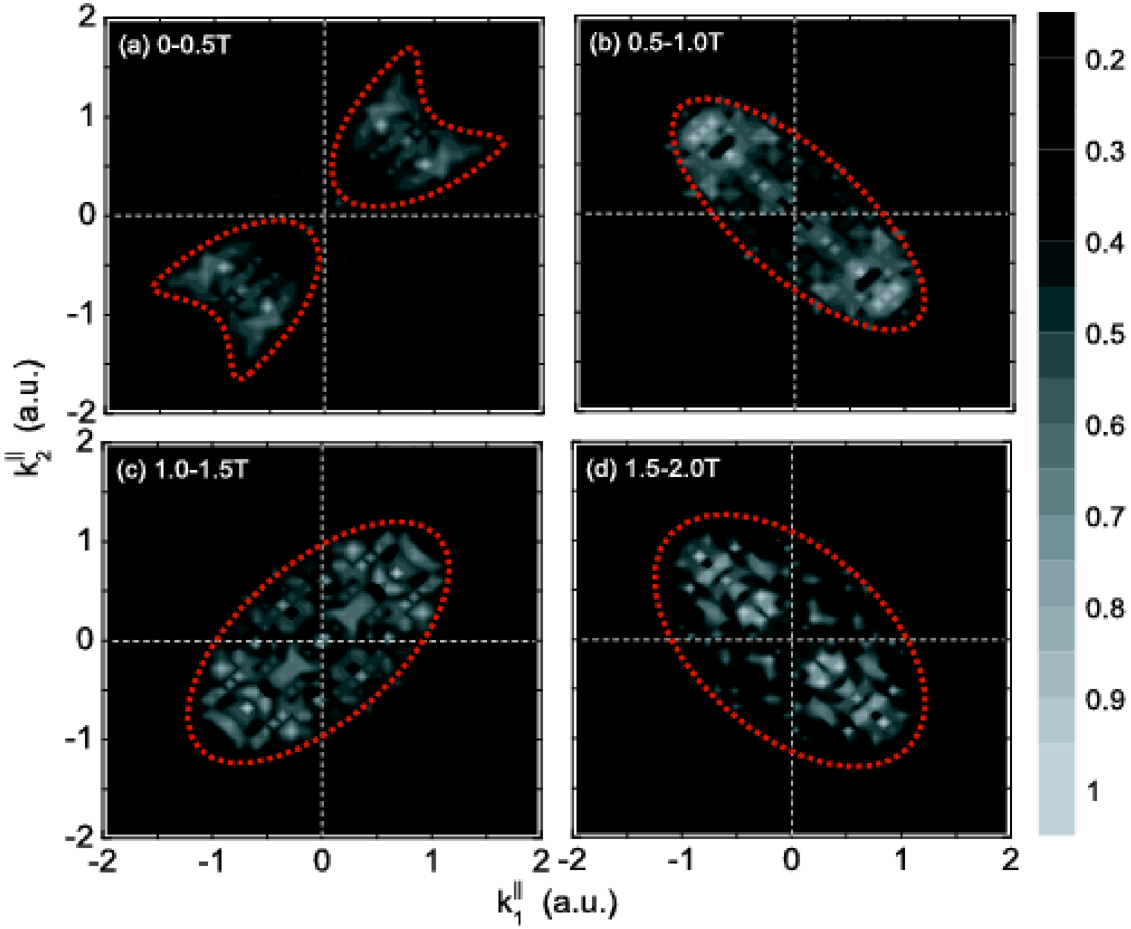
Time Delay



Time delay between double ionization and recollision

1. Statistical result
2. Individual trajectory
3. Effects on the DI dynamics

Momentum correlation: time delay effect



Momentum distributions with certain time delays.

1. (a) (c) time delay of odd half laser cycles, same hemispere emission

   (b) (d) time delay of even half laser cycles, opposite hemispere emission

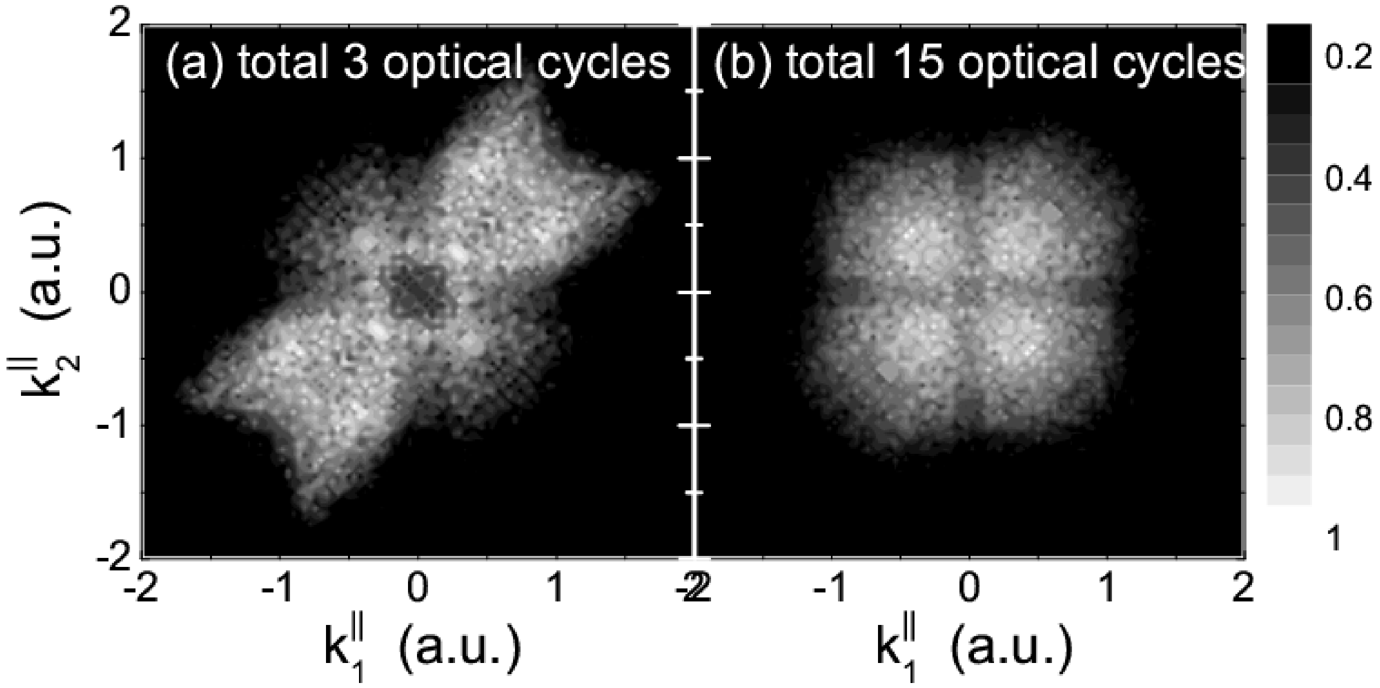
2. (a) fingerlike pattern

   (b)-(d) elliptical pattern

3. As the time delay increase the elliptical pattern becomes fatter and fatter, indicating the vanish of correlation between two electrons.

Phys.Rev.A. 77, 013403, 2008

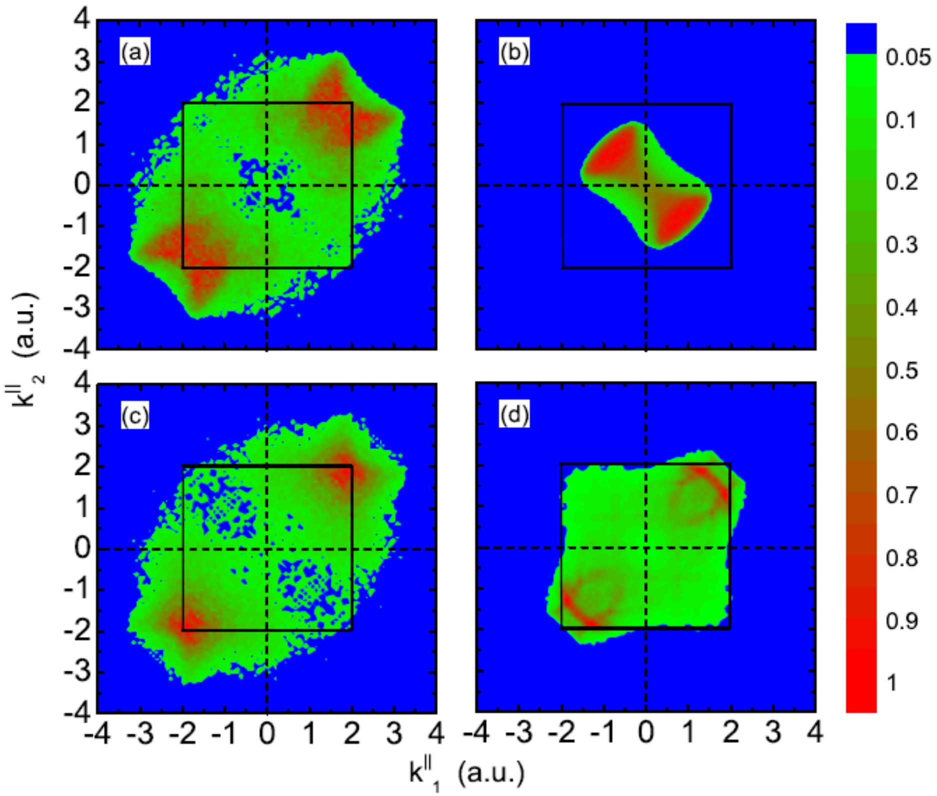
Momentum correlation: laser duration effect



Shorter laser pulse, fingerlike pattern is more pronounced

Longer laser pulse, correlation become blurry

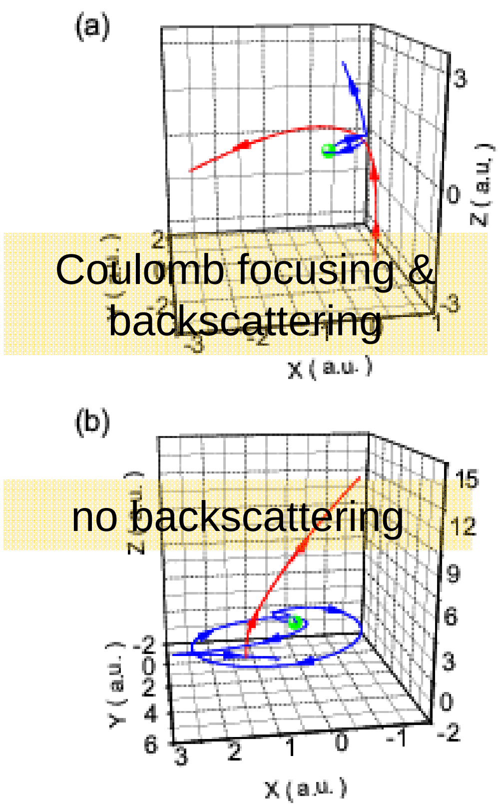
Why finger structure (or V-shaped)? (1)

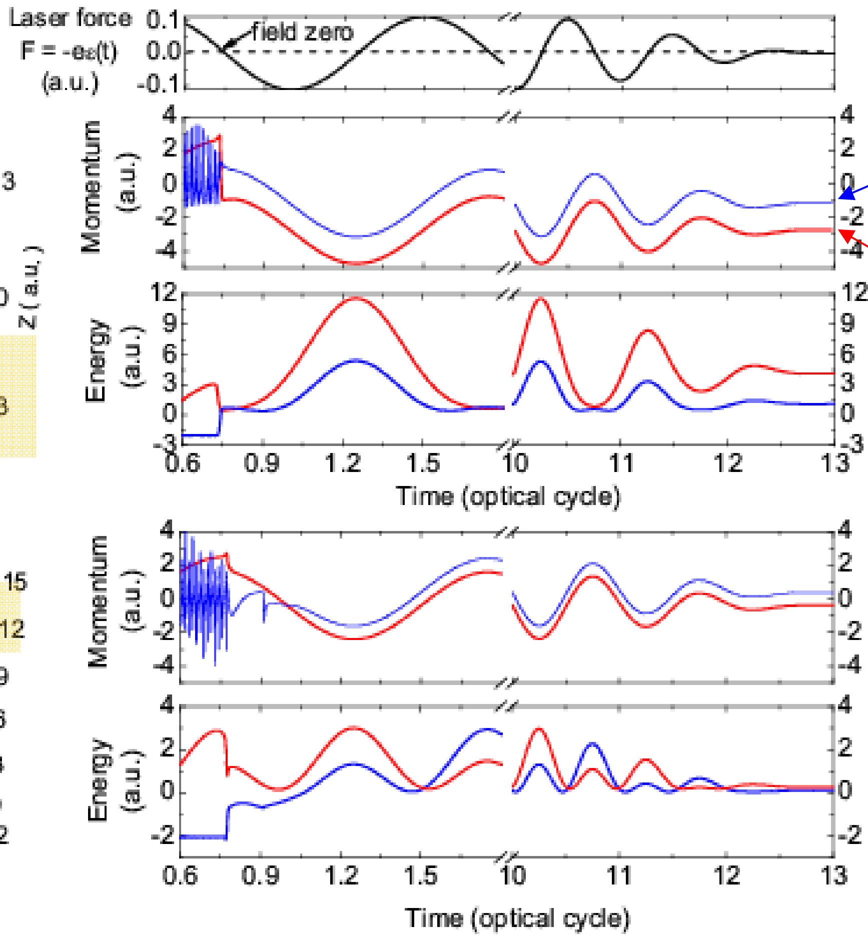


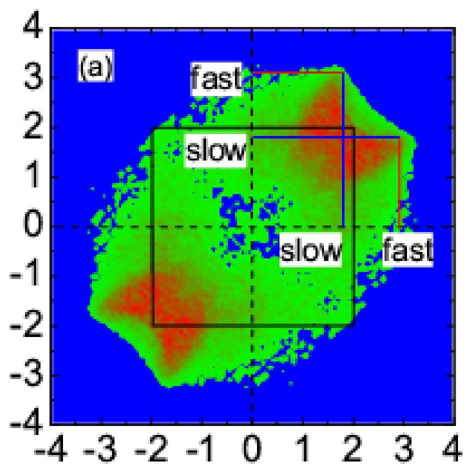
(a) Distribution of correlated electron momenta along the laser polarization for Helium DI irradiated by 800nm, 4.5×10^{14}W/cm^2 laser pulses. The black box indicates the $2\sqrt{U_p}$ boundary of electron momentum. The model calculations under various circumstances yield very different momentum distribution patterns(see text for details): (b) the laser field is removed and the tunneled electrons are replaced by a beam of projectile electrons; (c) the electron-electron Coulombic interaction is replaced with a Yukawa potential; (d) the nuclear Coulomb potential is softened.

The Coulomb-type attraction of nucleus is especially important for the production of fingerlike structure

Why finger structure (or V-shaped)? (2)





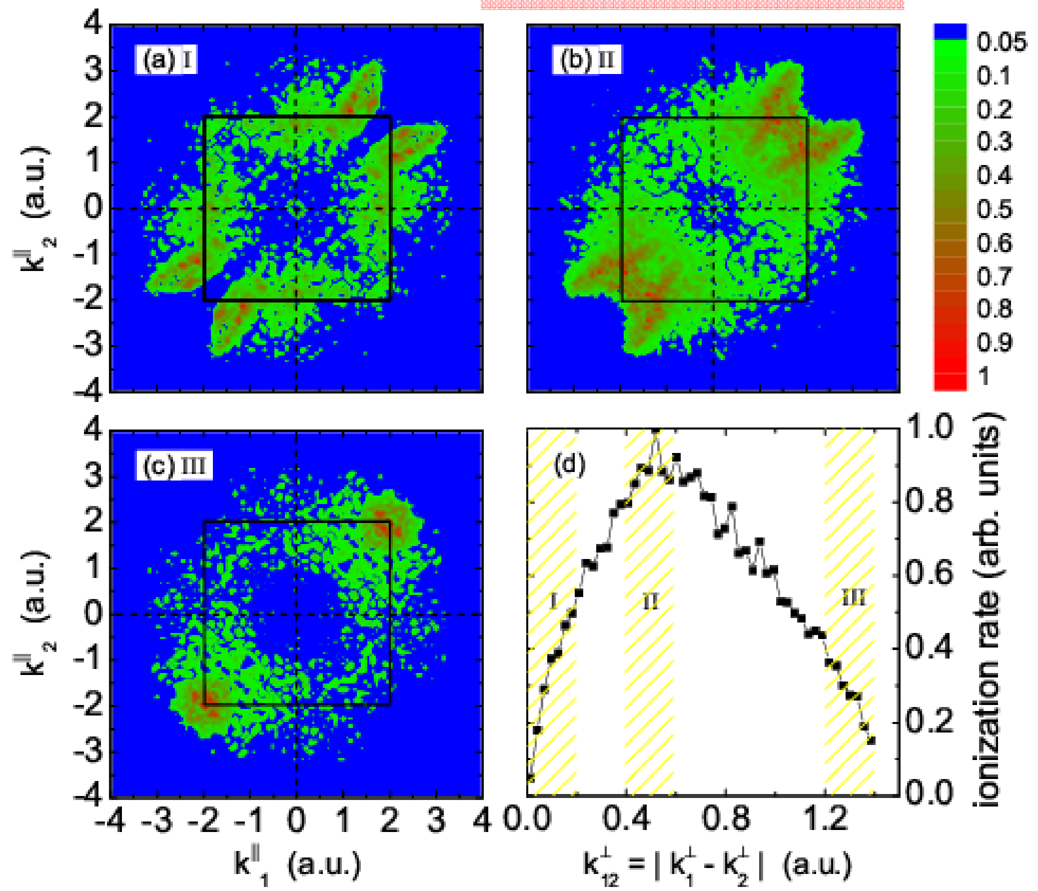


$$v_f = \left| v_0 \pm 2\sqrt{U_p} \right|$$

where v_0 is the speed of the electron at the field zero, and with the + or − chosen based on whether the laser force after the field zero is in the same direction as v_0 or opposite.

Perpendicular effect

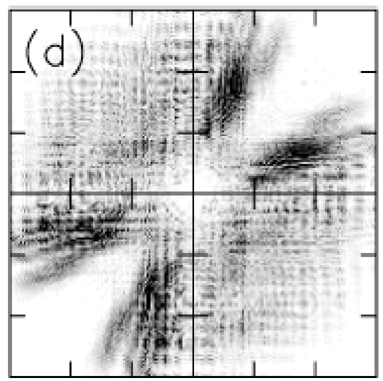
two fingers get close



two fingers disappear

Correlated parallel momentum distributions with additional conditions on the relative perpendicular momentum between two electrons, i.e., for (a) $0 \leqslant k^{\perp}_{12} \leqslant 0.2$, (b) $0.4 \leqslant k^{\perp}_{12} \leqslant 0.6$, (c) $1.2 \leqslant k^{\perp}_{12} \leqslant 1.4$. (d) The overall relative perpendicular momentum distribution.

1D quantum calculation



VOLUME 85, NUMBER 22 PHYSICAL REVIEW LETTERS 27 NOVEMBER 2000

Intense-Field Double Ionization of Helium: Identifying the Mechanism

M. Lein,[1,2] E. K. U. Gross,[2] and V. Engel[1]

Conclusion

1. Set up a semiclassical model for the double ionization problem.

   Advantages:

   ☆Include most of the important effects in DI process, e.g. the quantum tunneling and the Coulomb focusing.

   ☆Underlying mechanisms can be identified with the Classical Trajectory (CT) diagnosis

2. Reproduce and explain many experimental observations.

   ☆ DI ratio

   ☆ Finger-like structure

   …….

Thank You