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ATTOSECOND PHYSICS

UNIT V ABOVE-THRESHOLD IONIZATION OF ATOMS

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Introduction

We consider now the electron initially bound to one atom.
For the hydrogen atom:

$$V(r) = \frac{-e^2}{4\pi\varepsilon_0 r} \quad \text{Potential energy in MKS}$$

$$= -\frac{1}{r} \quad \text{Potential energy in a.u.}$$

The electric force felt by the electron in the first Bohr's orbit is

$$-e\vec{E}_a = -\vec{\nabla}V\Big|_{r=a_0} = \frac{-e^2}{4\pi\varepsilon_0 a_0^2}\hat{r} \Rightarrow E_a = \frac{e}{4\pi\varepsilon_0 a_0^2} = \begin{cases} 5\times10^{11}\,\frac{V}{m} & \text{electric field in MKS} \\ 1 & \text{electric field in a.u.} \end{cases}$$

Ionization condition

It is desirable to have a laser electric field of the same order of the Coulomb electric field

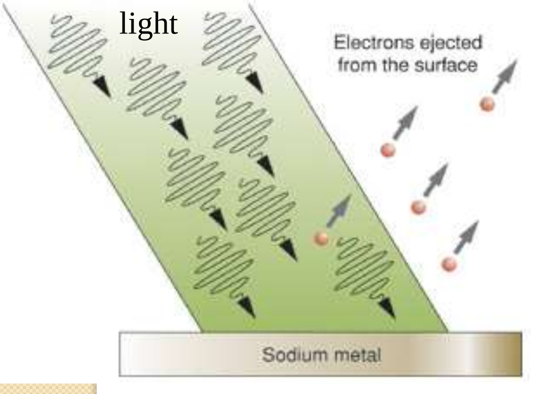
$$I_a = \frac{\varepsilon_0 c E_a^2}{2} = \begin{cases} 3.5\times10^{16}\,\frac{W}{cm^2} \\ \frac{c}{8\pi} = 5.45 \text{ a.u.} \end{cases}$$

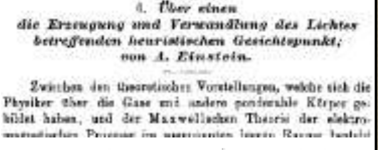
But we observe ionization for $I \geq 10^{10}\,\frac{\text{W}}{\text{cm}^2}$

Photoelectric Effect

A. Einstein, Annalen der Physik **322**, 132 (1905)

Einstein (1879-1955)





6. Über einen
die Erzeugung und Verwandlung des Lichtes
betreffenden heuristischen Gesichtspunkt;
von A. Einstein.

Zwischen den theoretischen Vorstellungen, welche sich die Physiker über die Gase und andere ponderable Körper gebildet haben, und der Maxwellschen Theorie der elektro-

$$K_{\max} = h\nu - W.$$

About a heuristic point of view concerning the production and transformation of light ;

How long do photoelectrons take to exit the metal plate?

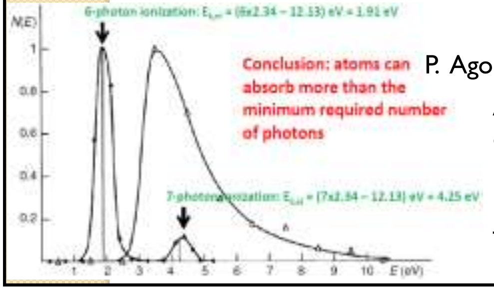
Physicists assumed the effect was instantaneous

Multiphoton Ionization

1930: Maria-Goeppert Mayer performed her PhD in Göttingen, with Max Born as her PhD supervisor, predicting the possibility that **2-photon absorption might be possible**. However, light sources allowing to validate this prediction were unavailable at the time.

In 1963 she became the 2nd female Nobel Laureate in physics, for her work on the nuclear shell model.

Maria-Goeppert Mayer



P. Agostini *et al.*, Phys. Rev. Lett. **42**, 1127 (1979)

Above-Threshold Ionization

6-photon ionization:

Energy spectra of electrons produced by multiphoton ionization of xenon atoms, for two photon energies. Triangles: ω = 1.17 eV; Circles: ω = 2.34 eV;

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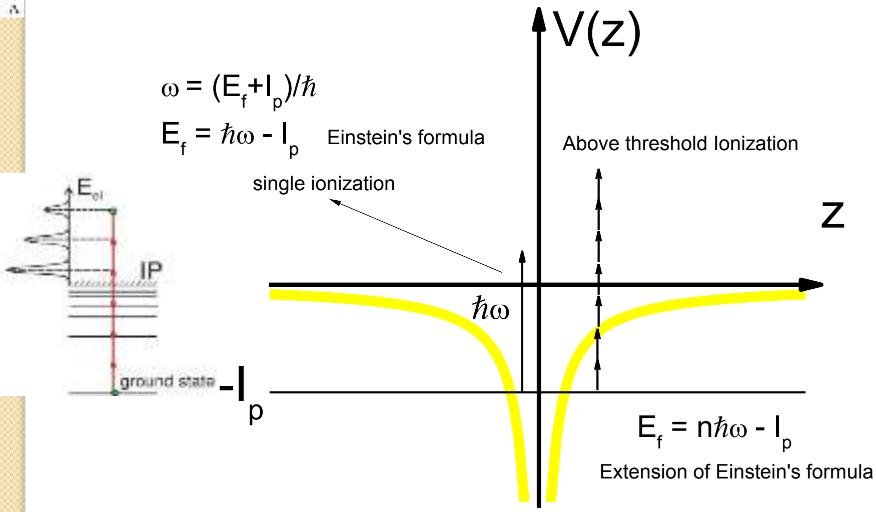
Multiphoton Ionization (ATI)

When lasers are weak and frequencies high, the distortion of the atomic potential due to the external electric field is negligible.

$\omega = (E_f + I_p)/\hbar$

$E_f = \hbar\omega - I_p$ Einstein's formula



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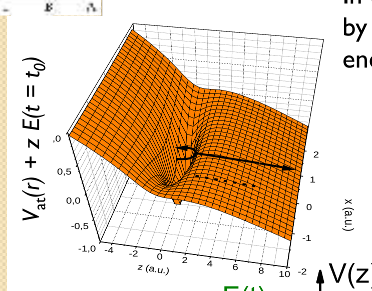
Over the Barrier Ionization

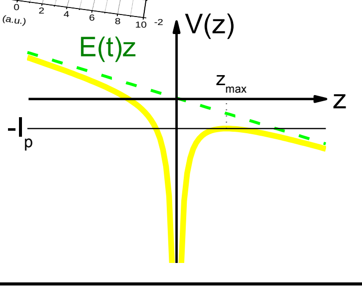
When lasers are strong and/or frequencies low, the distortion of the atomic potential due to the external electric field is appreciable.

In the length gauge, the total potential energy is given by the atomic potential energy added to the potential energy given by the external laser field

$$V(r) = \frac{-e^2}{4\pi\varepsilon_0 r} + eE(t)z \quad \text{(MKS)}$$

$$= -\frac{1}{r} + E(t)z \qquad \text{(a.u.)}$$





The value of the maximum of *V(z)* is:

$$z_{\max} = \frac{1}{\sqrt{|E|}}$$

$$V(z_{\max}) = -\frac{1}{z_{\max}} - |E(t)|\, z_{\max}$$

$$= -\sqrt{|E|} - \sqrt{|E|} = -2\sqrt{|E|}$$

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Over the Barrier Ionization (cont.)

The atom will ionize if the electron energy is higher than the potential barrier:

$$-I_p \geq -2\sqrt{|E|} \quad \Rightarrow \quad I_p \leq 2\sqrt{|E|} \quad \Rightarrow \quad |E| > E_c = \left(\frac{I_p}{2}\right)^2$$

For the hydrogen atom

$$I_p = \tfrac{1}{2} \quad \Rightarrow \quad E_c = \frac{E_a}{16} = 3.1\times 10^{10}\ \frac{\mathrm{V}}{\mathrm{m}}$$

$$\Rightarrow \quad I_c = \frac{I_a}{16^2} = 1.37\times 10^{14}\ \frac{\mathrm{W}}{\mathrm{cm}^2}$$

This provides a good idea about the importance of ionization:

If $I > I_c$ => atomic ionization is important

If $I < I_c$ => atomic ionization is small (but appreciable).

Conclusion: Another mechanism for atomic ionization

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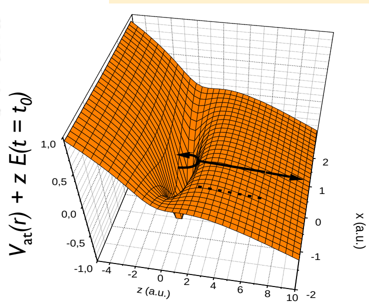
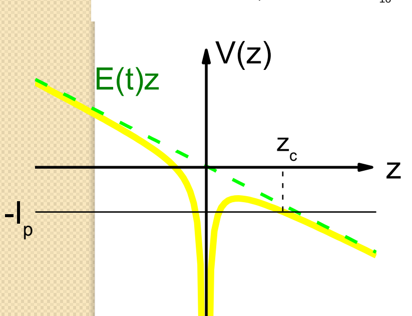
Tunneling Ionization



The electron can cross through the potential barrier by tunnel effect.

The ease (or difficulty) for the electron to tunnel can be defined as the classical time that the electron takes to tunnel through the barrier.

$$t_{\text{tun}} = \frac{\text{"tunneling distance"}}{\text{"tunneling speed"}}$$



According to the Virial theorem, the kinetic energy is minus the total energy:

$$\tfrac{1}{2} m_e v^2 = I_p \quad \Rightarrow \quad v_{\text{tun}} = \sqrt{\frac{2I_p}{m_e}}$$

The tunneling distance is approximately

$$-z_c |E_0| = -I_p \quad \Rightarrow \quad z_c = \frac{I_p}{|E_0|}$$

Tunneling Ionization (cont.)

Therefore, the tunneling time is:

$$t_{\text{tun}} = \frac{z_c}{v_{\text{tun}}} = \frac{I_p / |E_0|}{\sqrt{2 I_p}} = \frac{\sqrt{I_p/2}}{|E_0|}$$

The adiabaticity or Keldysh parameter is defined as the ratio of the tunneling time over half a laser cycle:

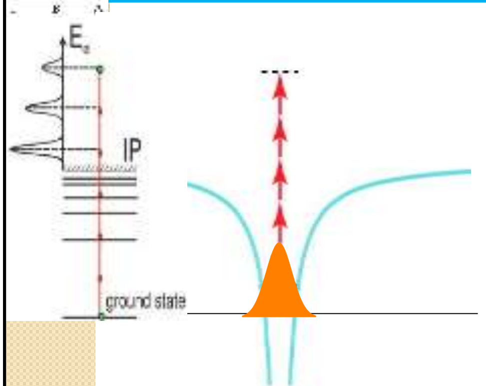
$$\gamma = \frac{\text{"tunneling time"}}{\text{"half cycle"}} = \frac{t_{tun}}{T/2}; \quad T \sim \frac{1}{\omega}$$

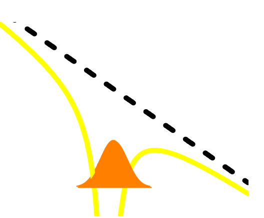
$$= \frac{\sqrt{I_p/2}}{E_0} 2\omega = \sqrt{2I_p}\,\frac{\omega}{E_0} = \sqrt{\frac{I_p}{2U_p}}$$

If $\gamma < 1$ => tunneling regime

PHOTOIONIZATION MECHANISMS (cont.)

Exercise 10: How much is the intensity of a Ti-Sapphire laser (λ = 800 nm) impinging over H so that the Keldysh parameter is γ = 1?





How long does it take for an atom to ionize?

Low intensities: multiphoton regime (γ > 1)

High intensities: tunneling regime (γ < 1)

Tunneling time?

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Theories and Approximations

Semiclassical Model

Born Approximation R-Matrix

Floquet Theory Coulomb-Volkov Approximation

Time-Dependent Schrödinger Equation

Impulse Coulomb-Volkov Approximation Simple Man's Model

Strong-Field Approximation Quantum Orbit Theory

Back-propagation method Lewenstein´s Model

Semiclassical Two-Step Model Quantum Trajectory Monte Carlo

Time-Dependent Density Functional Theory

Three-Step Model

Coulomb-Corrected Strong-Field Approximation

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Simple Man's Model

For the tunneling regime ($\gamma < 1$), we suppose that the ionization is produced in the following way:

- At $t = t_0$ the electron is tunneled out through the barrier. The ionization rate is given by a semiclassical estimation due to Amosov, Delone, and Krainov (ADK) J. Opt. Soc. Am. B **8**, 1207 (1991):

$$w = \left(\frac{3e}{\pi}\right)^{3/2} \frac{Z^2}{3n^{*3}} \frac{2l+1}{2n^*-1} \left[\frac{4eZ^3}{(2n^*-1)n^{*3}|E(t)|}\right]^{2n^*-3/2} \exp\left[\frac{-2Z^3}{3n^{*3}|E(t)|}\right]$$

$$\text{where } n^* = Z\Big/\sqrt{2I_p} \quad (n^* = 1 \text{ for atomic hydrogen})$$

- Right after tunneling (which is considered to be instantaneous at $t = t_0$), the electron appears with zero speed at the origin.

- After the ionization ($t > t_0$), the electron behaves classically driven by the external electric field and the atomic Coulomb potential is neglected (Strong Field Approximation).

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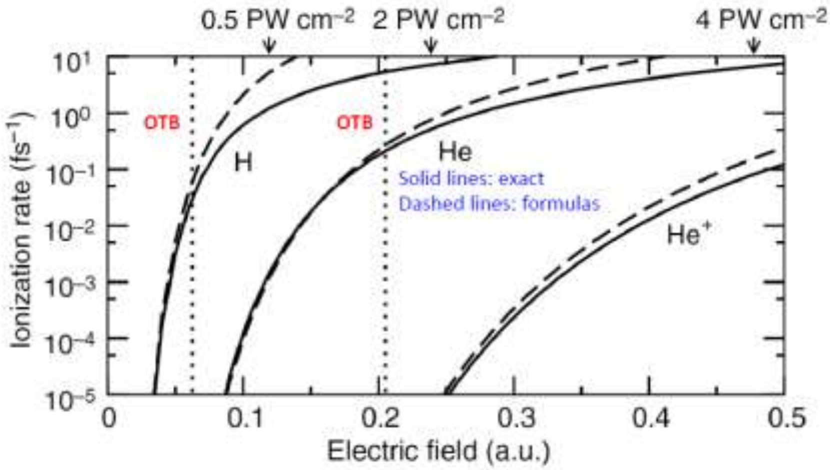
Simple Man's Model (cont.)

Tunneling formulas



C.J. Joachain, N.J. Kylstra and R.M. Potvliege, Atoms in Intense Laser Fields, (Cambridge University Press, 2012)

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Simple Man's Model (cont.)

As an example we will apply the Simple Man's Model to the case of an atom in a linearly polarized sinusoidal field

$$\vec{E} = \hat{z}E_0 \sin \omega t; \quad \hat{z}: \text{ polarization direction}$$

We use atomic units

Lorentz Force: $\ddot{z}(t) = -E_0 \sin \omega t$

$$\dot{z}(t) = \frac{E_0}{\omega}\cos \omega t + v_{\text{drift}} \quad \text{with} \quad v_{\text{drift}} = \frac{-E_0}{\omega}\cos \omega t_0$$

$$z(t) = \alpha_0 \sin \omega t + v_{\text{drift}}(t - t_0) - \alpha_0 \sin \omega t_0$$

kinetic energy: $T_{\text{kin}}(t) = \frac{1}{2}\dot{z}^2(t) = \frac{E_0^{\;2}}{2\omega^2}\cos^2 \omega t + \frac{v_{\text{drift}}^2}{2} + v_{\text{drift}}\frac{E_0}{\omega}\cos \omega t$

time average: $\langle T_{\text{kin}} \rangle = \frac{1}{T}\int_t^{t+T} T_{\text{kin}}(t')dt' = \frac{E_0^{\;2}}{4\omega^2} + \frac{v_{\text{drift}}^2}{2} = U_p + \frac{v_{\text{drift}}^2}{2}$

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Simple Man's Model (cont.)

What do we measure at the detector?

- The detector is at a distance considered infinite in relation to the atomic dimensions.

- The laser pulse adiabatically switches off ➔ the electron does not oscillate any more and only the drift velocity stays on:

$$\boxed{\langle T_{\text{kin}} \rangle = \frac{v_{\text{drift}}^2}{2}}$$

What is the maximum kinetic energy of the electron that we can expect? We must find the ionization times so that $|v_{\text{drift}}|$ is maximized.

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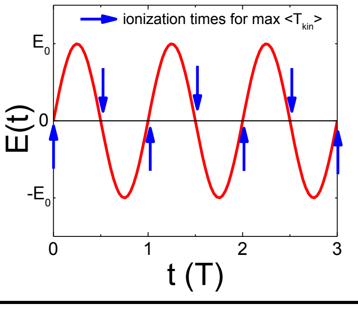
Simple Man's Model (cont.)

$v_{\text{drift}} = \frac{-E_0}{\omega} \cos \omega t_0$ possesses extremes (max and min) for

$\omega t_0 = j\pi, \quad t_0 = jT/2$, that is, every half cycle

$$\Rightarrow \left(\frac{v_{\text{drift}}^2}{2} \right)_{\max} = \frac{E_0{}^2}{2\omega^2} = 2U_p$$

$$\Rightarrow \boxed{\langle T_{\text{kin}}(t) \rangle_{\max} = 2U_p}$$



The electric field is zero when $<T_{kin}>_{max} = 2U_p$

Therefore, there cannot be tunneling ionization and $<T_{kin}>_{max}$ cannot be classically reached.

Keldysh-Faisal-Reiss Theory

We will develop the S-matrix theory and will apply it to the problem of the laser-atom interaction. We consider an atom in the Single-Active Electron (SAE) approximation under the influence of a linearly polarized electric field.

We suppose an interaction Hamiltonian H_{int} delimited in time, we mean, it starts and finishes : $\lim_{t\to\pm\infty} H_{\text{int}} = 0$

$$H = H_0 + H_{\text{int}}$$

$$i\hbar \frac{\partial \Phi}{\partial t} = H_0 \Phi \qquad \text{(with no interaction)} \quad (1)$$

$$i\hbar \frac{\partial \Psi}{\partial t} = H \Psi \qquad \text{(with interaction)} \quad (2)$$

Keldysh-Faisal-Reiss Theory (cont.)

$\Psi_i^{(+)}(t)$ is solution of Eq. (2) with the following boundary conditions

$$\lim_{t\to-\infty} \Psi_i^{(+)}(t) = \Phi_i(t)$$

since H_{int} is not initially acting.

The transition amplitude from one initial state (without interaction) to a final state (without interaction) is $(\Phi_i \to \Phi_f)$:

$$T_{if} = \lim_{t\to+\infty} \left\langle \Phi_f(t) \middle| \Psi_i^{(+)}(t) \right\rangle$$

To eliminate the asymptotic limits we make use of integration by parts:

$$\int_a^b dx\, f(x) g'(x) = f(x)g(x)\Big|_a^b - \int_a^b dx\, f'(x) g(x)$$

Keldysh-Faisal-Reiss Theory (cont.)

$$\int_{-\infty}^{+\infty} dt \left\langle \Phi_f(t) \middle| \dot{\Psi}_i^{(+)}(t) \right\rangle = \left\langle \Phi_f(t) \middle| \Psi_i^{(+)}(t) \right\rangle \Big|_{-\infty}^{+\infty} - \int_{-\infty}^{+\infty} dt \left\langle \dot{\Phi}_f(t) \middle| \Psi_i^{(+)}(t) \right\rangle$$

$$= \left\langle \Phi_f(\infty) \middle| \Psi_i^{(+)}(\infty) \right\rangle - \underbrace{\left\langle \Phi_f(-\infty) \middle| \underbrace{\Psi_i^{(+)}(-\infty)}_{\Phi_i(-\infty)} \right\rangle}_{\delta_{if}} - \int_{-\infty}^{+\infty} dt \left\langle \dot{\Phi}_f(t) \middle| \Psi_i^{(+)}(t) \right\rangle$$

$$\left\langle \Phi_f(\infty) \middle| \Psi_i^{(+)}(\infty) \right\rangle = \delta_{if} + \int_{-\infty}^{+\infty} dt \left[\left\langle \Phi_f(t) \middle| \underbrace{\dot{\Psi}_i^{(+)}(t)}_{\frac{-i}{\hbar} H \Psi_i^{(+)}(t)} \right\rangle + \left\langle \underbrace{\dot{\Phi}_f(t)}_{\frac{i}{\hbar} H_0 \Phi_f(t)} \middle| \Psi_i^{(+)}(t) \right\rangle \right]$$

$$\left\langle \Phi_f(\infty) \middle| \Psi_i^{(+)}(\infty) \right\rangle = \delta_{if} - \tfrac{i}{\hbar} \int_{-\infty}^{+\infty} dt \left[\left\langle \Phi_f(t) \middle| (H - H_0) \middle| \Psi_i^{(+)}(t) \right\rangle \right]$$

$$\left\langle \Phi_f(\infty) \middle| \Psi_i^{(+)}(\infty) \right\rangle = \delta_{if} - \tfrac{i}{\hbar} \int_{-\infty}^{+\infty} dt \left[\left\langle \Phi_f(t) \middle| H_{\text{int}} \middle| \Psi_i^{(+)}(t) \right\rangle \right]$$

Keldysh-Faisal-Reiss Theory (cont.)

For the case of ionization: $(\Phi_f \neq \Phi_i)$

$$T_{if} = \lim_{t \to +\infty} \left\langle \Phi_f(t) \middle| \Psi_i^{(+)}(t) \right\rangle = \left\langle \Phi_f(\infty) \middle| \Psi_i^{(+)}(\infty) \right\rangle$$

$$T_{if} = -\tfrac{i}{\hbar} \int_{-\infty}^{+\infty} dt \left\langle \Phi_f(t) \middle| e\vec{r} \cdot \vec{E}(t) \middle| \Psi_i^{(+)}(t) \right\rangle$$

The quantum mechanics equations of motion are invariant under time inversion:

$$(\Phi_f \to \Phi_i) \qquad T_{fi} = \lim_{t \to -\infty} \left\langle \Psi_f^{(-)}(t) \middle| \Phi_i(t) \right\rangle \qquad \lim_{t \to +\infty} \Psi_f^{(-)}(t) = \Phi_f(t)$$

On equal footing

$$T_{fi} = -\tfrac{i}{\hbar} \int_{-\infty}^{+\infty} dt \left\langle \Psi_f^{(-)}(t) \middle| e\vec{r} \cdot \vec{E}(t) \middle| \Phi_i(t) \right\rangle$$

So far, everything is exact:

$\Phi_{i,f}(t)$ is exactly known (eigenfunction of the atomic Hamiltonian).

$\Psi_{i,f}^{(\pm)}(t)$ is not known (eigenfunction of the full Hamiltonian).

In this way, to compute T_{if} some kind of approximation is necessary.

Keldysh-Faisal-Reiss Theory (cont.)

1) First Born Approximation: Perturbative method

The exact wave function is replaced by the non-distorted solution of the free Hamiltonian H_0. Thus, the laser field is neglected in the final channel.

Post:

$$T_{if} = -\tfrac{i}{\hbar}\int_{-\infty}^{+\infty} dt \left\langle \Phi_f(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \Psi_i^{(+)}(t) \right\rangle \Rightarrow T_{if} = -\tfrac{i}{\hbar}\int_{-\infty}^{+\infty} dt \left\langle \Phi_f(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \Phi_i(t) \right\rangle$$

Prior:

$$T_{fi} = -\tfrac{i}{\hbar}\int_{-\infty}^{+\infty} dt \left\langle \Psi_f^{(-)}(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \Phi_i(t) \right\rangle \Rightarrow T_{fi} = -\tfrac{i}{\hbar}\int_{-\infty}^{+\infty} dt \left\langle \Phi_f(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \Phi_i(t) \right\rangle$$

$$\therefore T_{if} = T_{fi}$$

and therefore the probabilities are the same in the prior and post forms

For the case of the Hydrogen atom:
$$\begin{cases} \Phi_f(t) = \Phi_{\vec{k}}(\vec{r},t) = e^{-i\varepsilon t}\dfrac{e^{i\vec{k}.\vec{r}}}{(2\pi)^{3/2}} D_c(Z,\vec{k},t) \\ D_c(Z,\vec{k},t) = N_T(k)\,{}_1F_1\left(-i\tfrac{Z}{k},1,-ikr-i\vec{k}.\vec{r}\right) \\ N_T(k) = e^{\frac{\pi Z}{2k}}\Gamma(1+i\tfrac{Z}{k}); \quad Z \text{ is the ion charge} \end{cases}$$

This is a good approximation for **weak** external electric fields: $H_{int} << H_0$.

Keldysh-Faisal-Reiss Theory (cont.)

2) Strong Field Approximation (SFA):

The exact wave function is replaced by the solution of the Schrödinger equation in the laser field (Volkov function) neglecting the atomic potential

Prior:
$$T_{fi} = -\tfrac{i}{\hbar}\int_{-\infty}^{+\infty} dt \left\langle \chi_f^{(V)-}(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \Phi_i(t) \right\rangle$$

For the case of the Hydrogen atom:

Volkov state:
$$\chi_f^{V-}(\vec{r},t) = e^{-i\varepsilon_f t}\frac{e^{i\vec{k}_f.\vec{r}}}{(2\pi)^{3/2}} e^{iD^-(\vec{k}_f,\vec{r},t)}$$

$$D^-(\vec{k},\vec{r},t) = \vec{A}(t)\cdot\vec{r} - \vec{k}\int_{+\infty}^{t} dt'\vec{A}(t') - \tfrac{1}{2}\int_{+\infty}^{t} dt'\left[\vec{A}(t')\right]^2$$

A(t): vector potential

This is a good approximation for **strong** external electric fields: $H_{int} >> H_0$.

Keldysh-Faisal-Reiss Theory (cont.)

3) Coulomb-Volkov Approximation (CVA): not perturbative method
The exact wave function is replaced by a compromise of the two former approximations.
We consider the interaction of the active electron with the external laser field and the core potential in a non-perturbative way yet approximated.
We replace the exact solution by the Coulomb-Volkov wave function.

Post: $$T_{if} = -\tfrac{i}{\hbar} \int_{-\infty}^{+\infty} dt \left\langle \Phi_f(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \chi_i^{(CV)+}(t) \right\rangle$$

Prior: $$T_{fi} = -\tfrac{i}{\hbar} \int_{-\infty}^{+\infty} dt \left\langle \chi_f^{(CV)-}(t) \middle| e\vec{r}\cdot\vec{E}(t) \middle| \Phi_i(t) \right\rangle$$

$$\therefore T_{if} \neq T_{fi}$$

and therefore the probabilities are not the same in the prior and post forms

Coulomb-Volkov state: $$\chi_f{}^{(CV)-}(\vec{r},t) = \Phi_{\vec{k}}(\vec{r},t) e^{iD^-(\vec{k},\vec{r},t)}$$
$$= \chi_f{}^{(V)-}(\vec{r},t) D_c(Z,\vec{k},0)$$

This is a good approximation for **intermediate** external electric fields: $H_{int} \sim H_0$.

Keldysh-Faisal-Reiss Theory (cont.)

$$D_c(Z,\vec{k},t) \xrightarrow[Z\to 0]{} 1$$
$$\chi_f{}^{(CV)-}(\vec{r},t) \xrightarrow[Z\to 0]{} \chi_f{}^{(V)-}(\vec{r},t)$$

CV → SFA

4) Eikonal-Volkov distorted wave Approximation (EVA): not perturbative method.
It corresponds to the inclusion of the Coulomb phase accumulated in straight trajectories, neglecting the deflection on the core field.

$$D_c(Z,\vec{k},t) \xrightarrow[r\to\infty]{} \exp\left[i\frac{Z}{k} \ln\left(\vec{k}\cdot\vec{r} + kr\right)\right]$$

This is a good approximation for **strong** external electric fields but it is more elaborated than the first Born approximation.

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Analytical Solution of the SFA

Lewenstein model:
It solves the TDSE within the SFA and many other assumptions.
The result is rather simple and for sinusoidal pulses is analytical.

We will arrive to the same result of the Lewenstein model starting from the continuum distorted wave SFA:

$$T_{if} = -i\int_{-\infty}^{+\infty} dt \left\langle \chi_f^{V(-)}(t) \middle| zE(t) \middle| \Phi_i(t) \right\rangle$$

$$T_{if} = -i\int_{0}^{\tau} dt\, E(t) d^*(\vec{k}+\vec{A}(t)) \exp[-iS(t,\tau)] \quad \text{pulse starts at 0 and ends at } \tau$$

$$d^*(\vec{v}) = \langle \vec{v} | z | i \rangle \quad \text{atomic dipole element}$$

$$S(t,\tau) = \int_t^{\tau} dt' \left\{ \frac{\left[\vec{k}+\vec{A}(t')\right]^2}{2} + I_p \right\} \qquad \text{classical action (see Volkov states)}$$

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Analytical Solution of the SFA (cont.)

Exercise 11: Calculate the classical action for a sinusoidal linearly polarized fiel

$$\vec{E}(t) = E_0 \cos(\omega t)\hat{z}$$

Solution:

$$S(t,\tau) = -\left[\frac{k^2\tau}{2} + \frac{E_0^2}{2\omega^2}\left(\frac{\tau}{2} + \frac{\sin 2\omega\tau}{4\omega}\right) + \frac{k_z E_0}{\omega^2}\cos\omega\tau + I_p\tau\right] +$$
$$+\left[\frac{k^2 t}{2} + \frac{E_0^2}{2\omega^2}\left(\frac{t}{2} + \frac{\sin 2\omega t}{4\omega}\right) + \frac{k_z E_0}{\omega^2}\cos\omega t + I_p t\right]$$

We define $S(t,\tau) = S(t) - S(\tau)$, where

$$S(t) = \left[\frac{k^2 t}{2} + \frac{E_0^2}{2\omega^2}\left(\frac{t}{2} + \frac{\sin 2\omega t}{4\omega}\right) + \frac{k_z E_0}{\omega}\cos\omega t + I_p t\right]$$

Analytical Solution of the SFA

I suppose a flattop pulse with adiabatic switch on and switch off

$$\vec{F}(t) = F_0 \cos(\omega t)\hat{z} \quad \Rightarrow \vec{A}(t) = \frac{F_0}{\omega}\sin(\omega t)\hat{z}$$

$$S(t,\tau) = S(\tau) - S(t)$$

$$S(t) = at + b\cos(\omega t) + c\sin(2\omega t)$$

$$a = \frac{k^2}{2} + I_p + U_p$$

$$b = \alpha k_z, \qquad \alpha = F_0/\omega^2$$

$$c = -\frac{U_p}{2\omega}, \qquad U_p = (F_0/2\omega)^2$$

Periodicity properties:

$T = 2\pi/\omega$

$$\begin{cases} S(t) - at = S(t+jT) - a(t+jT) \\ \Rightarrow S(t+jT) = S(t) + ajT \\ \vec{d}\left[\vec{k} + \vec{A}(t+jT)\right] = \vec{d}\left[\vec{k} + \vec{A}(t)\right] \end{cases}$$

Analytical Solution of the SFA

pulse with N cycles $\tau = NT$

$$T_{if} = -i\exp[-iS(\tau)]\int_0^{NT} dt\,E(t)d^*(\vec{k} + \vec{A}(t))\exp[iS(t)]$$

$$= -i\exp[-iS(\tau)]\sum_{j=0}^{N-1}\int_{jT}^{(j+1)T} dt\,E(t)d^*(\vec{k} + \vec{A}(t))\exp[iS(t)]$$

performing the transformation $t = t' + jT$

$$T_{if} = -i\exp[-iS(\tau)]\sum_{j=0}^{N-1} e^{iajT}\int_0^T dt'\,E(t')d^*(\vec{k} + \vec{A}(t'))\exp[iS(t')]$$

Geometrical sum

$$\sum_{j=0}^{N-1} r^j = \frac{1-r^N}{1-r}$$

$$\sum_{j=0}^{N-1} e^{iajT} = \frac{1-e^{iaNT}}{1-e^{iaT}} = \frac{e^{iaNT/2}\left(e^{-iaNT/2} - e^{iaNT/2}\right)}{e^{iaT/2}\left(e^{-iaT/2} - e^{iaT/2}\right)}$$

$$= e^{ia(N-1)T/2}\frac{\sin(aNT/2)}{\sin(aT/2)}$$

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Analytical Solution of the SFA

$$T_{if} = -i\exp[-iS(\tau)]e^{ia(N-1)T/2}\frac{\sin\left(aNT/2\right)}{\sin\left(aT/2\right)}I(\vec{k})$$

$$I(\vec{k}) = \int_{jT}^{(j+1)T} dt' E(t') d^*(\vec{k}+\vec{A}(t'))\exp[iS(t')]$$

The important physical magnitude is the probability:

$$\boxed{\frac{dP}{d\vec{k}} = \left|T_{if}\right|^2 = \underbrace{\left|\frac{\sin\left(aNT/2\right)}{\sin\left(aT/2\right)}\right|^2}_{B(k)} \underbrace{\left|I(\vec{k})\right|^2}_{F(\vec{k})}}$$

Equation of the diffraction grating in the time domain

Intercycle factor

Intracycle factor

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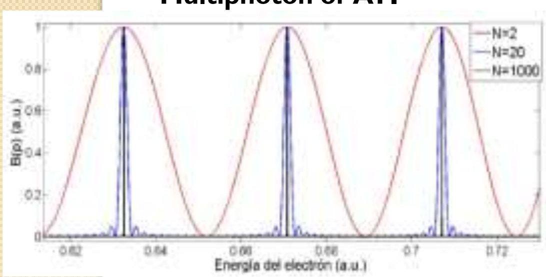
Analytical Solution of the SFA (cont.)

Intercycle interference

$$\max: \frac{aT}{2} = \frac{\pi}{\omega}\left[\varepsilon + U_p + I_p\right] = \pi n \quad \Rightarrow \quad \boxed{\varepsilon_n = n\hbar\omega - I_p - U_p}$$

Generalized Einstein's formula

Multiphoton or ATI



Uncertainty Principle:

$$\Delta\varepsilon\,\tau = \frac{\omega}{N}\frac{2\pi N}{\omega} = 2\pi = h$$

$$\Delta\varepsilon\,\tau \sim \hbar$$

1D SMM: $F_0 = 0.05$ ($I = 8.8 \times 10^{13}$ W/cm^2), $\omega = 0.05$ ($\lambda = 911$ nm), $I_p = 0.5$

$$\underbrace{\left[\frac{\sin\left(NaT/2\right)}{\sin\left(aT/2\right)}\right]^2}_{B(k)} \xrightarrow[N\to\infty]{} \sum_n \delta(\varepsilon - \varepsilon_n)$$

Energy conservation

Analytical Solution of the SFA (cont.)

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In order to calculate the time integral we use the **Saddle-Point Approximation:**

$$I(\vec{k}) = \int_0^T dt\, E(t) d^*(\vec{k} + \vec{A}(t)) \exp[iS(t)]$$

Stationary phase or steepest descent method [Arfken, Mathematical Methods for Physicists, Academic Press, Inc (1985) pg. 428]:

$$I(s) = \int_C g(z) \exp[sf(z)] dz \approx \frac{\sqrt{2\pi}\, g(z_0) \exp[sf(z_0)]}{\left| sf''(z_0) \right|^{1/2}}$$

$$\frac{df(z_0)}{dz} = 0$$

In our case:

$$g(z) = E(t) d^*(\vec{v}(t))$$

$$sf(z) = iS(t)$$

$$sf''(z_0) = i\frac{\partial^2 S(t)}{\partial t^2} = i\frac{\partial}{\partial t}\left[\frac{v(t)^2}{2} + I_p\right] = i\vec{v}(t)\frac{\partial \vec{v}(t)}{\partial t} = -i[\vec{k} + \vec{A}(t)]\vec{E}(t)$$

$$z_0 = t_{SP};$$

Analytical Solution of the SFA (cont.)

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$$\boxed{T_{if} = -i \sum_{SP} \left\{ \frac{2\pi E(t_{SP})}{\left| \vec{k} + \vec{A}(t_{SP}) \right|} \right\}^{1/2} d^*(\vec{k} + \vec{A}(t_{SP})) \exp[iS(t_{SP})]}$$

Ionization times:

$$\left.\frac{\partial S(t)}{\partial t}\right|_{t=t_{SP}} = \frac{\left[\vec{k} + \vec{A}(t_{SP})\right]^2}{2} + I_p = 0 \qquad \text{SPA}$$

$$= k + A(t_{SP}) = 0 \qquad \text{SMM}$$

The electron escapes the atom with initial null velocity: $v(t_{SP}) = 0$

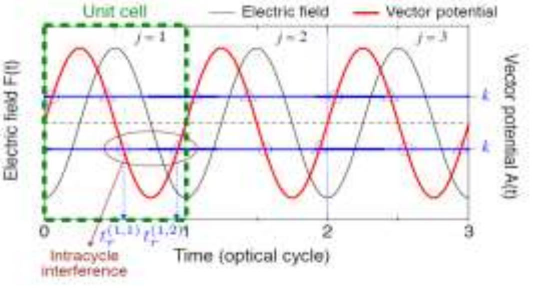
Sinusoidal field:

$$E(t) = -E_0 \cos(\omega t)$$

$$A(t) = \frac{E_0}{\omega} \sin(\omega t)$$

$$t_r^{(1)} = \frac{1}{\omega} \sin^{-1}\left[-\frac{\omega k}{E_0}\right]$$

$$t_r^{(2)} = \frac{2\pi}{\omega} - t_r^{(1)}$$



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Analytical Solution of the SFA (cont.)

$$I(\vec{k}) = -i\sum_{SP}\left\{\frac{2\pi E(t_{SP})}{\left|\vec{k}+\vec{A}(t_{SP})\right|}\right\}^{1/2} d^*(\vec{k}+\vec{A}(t_{SP}))\exp[iS(t_{SP})]$$

$$I(\vec{k}) = \Gamma^{1/2}(\vec{k})\left(e^{iS(t_r^{(1)})}+e^{iS(t_r^{(2)})}\right)$$

$$= \Gamma^{1/2}(\vec{k})e^{i\bar{S}}\left[e^{i\frac{\Delta S}{2}}+e^{-i\frac{\Delta S}{2}}\right]$$

$$= 2\Gamma^{1/2}(\vec{k})e^{i\bar{S}}\cos\frac{\Delta S}{2}$$

$$\bar{S} = \frac{S(t_r^{(1)})+S(t_r^{(2)})}{2}$$

$$\Delta S = S(t_r^{(1)})-S(t_r^{(2)})$$

The accumulated action between the two ionization times is ΔS.

$$\boxed{\frac{dP}{d\vec{k}} = \left|T_{if}\right|^2 = 4\Gamma(\vec{k})\underbrace{\left[\frac{\sin(NaT/2)}{\sin(aT/2)}\right]^2}_{B(k)}\underbrace{\cos^2\left(\frac{\Delta S}{2}\right)}_{F(\vec{k})}}$$

intercycle ***intracycle***

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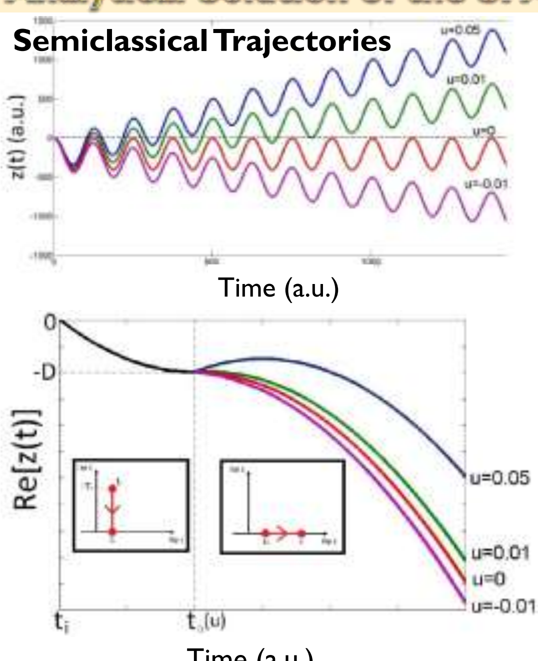
Analytical Solution of the SFA (cont.)

Semiclassical Trajectories



$u = \omega t_0$

Analytical Solution of the SFA (cont.)

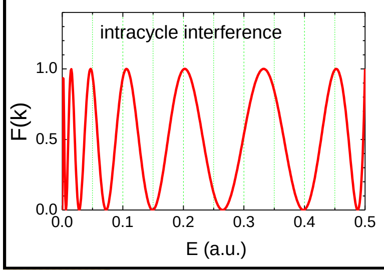
We now analyze the intracycle factor:

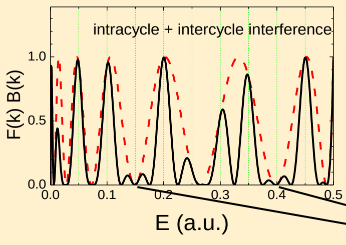
$$F(\vec{k}) = \cos^2\left(\frac{\Delta S}{2}\right)$$

Time double-slit experiment

where ΔS : accumulated action between $t_r^{(1)}$ and $t_r^{(2)}$

1D SMM: $F_0 = 0.05$ ($I = 8.8\times10^{13}$ W/cm^2), $\omega = 0.05$ ($\lambda = 911$ nm), $I_p = 0.5$

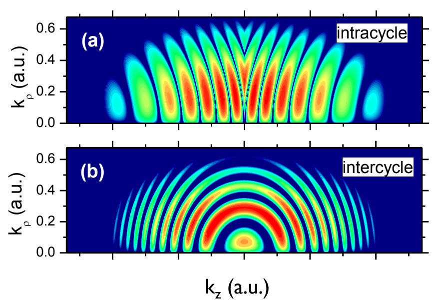




Frustrated orders

Analytical Solution of the SFA (cont.)

SPA: Doubly-differential momentum distributions $F_0 = 0.0675$, $\omega = 0.05$, $I_p = 0.5$



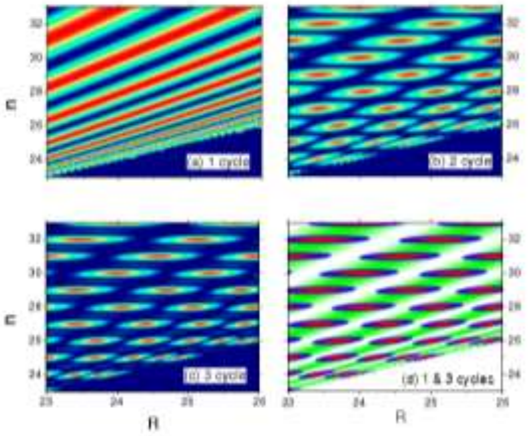
Analytical Solution of the SFA (cont.)

How does the photoelectron spectrum change with wavelength?
2D interferograms

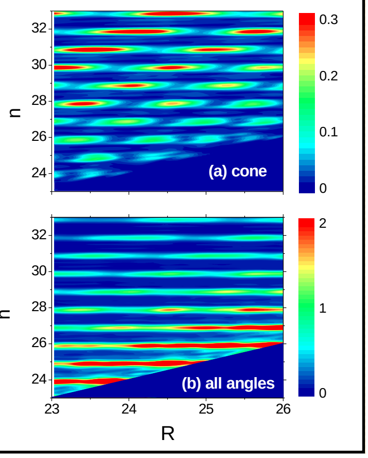
$$R = \frac{I_p + U_p}{\omega} \qquad n = R + \frac{E}{\omega}$$

$F_0 = 0.0675$, $I_p = 0.5$

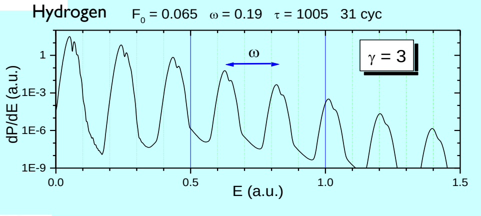
SMM

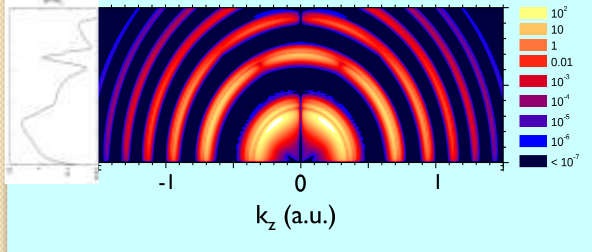


TDSE



Above Threshold Ionization Results (cont.)





$I = 1.5 \times 10^{14}\,W/cm^2$
$\lambda = 230$ nm

- ATI peaks

$$E_n = n\hbar\omega - I_p - U_p$$

$$U_p = \frac{E_0^2}{4\omega^2}$$

- 2d: iso-energy circles

$$k_\rho{}^2 = k_x{}^2 + k_y{}^2$$
$$k_\rho{}^2 + k_z{}^2 = 2E_n$$

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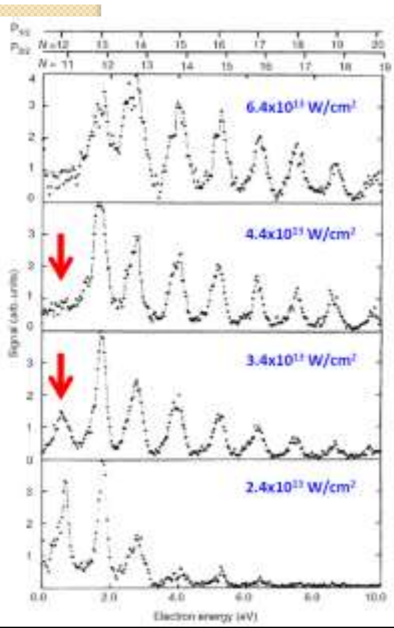
Above Threshold Ionization Results (cont.)



Ionization of Xe using 10 ns long, 1064 nm laser pulses

Measured ATI spectra as a function of intensity show a suppression of the lowest order ATI peak.

This shows that the magnitude of the laser electric field has a non-trivial influence on the ATI peak distribution (--> non-perturbative)

P. Kruit et al., Phys. Rev. A 28, 248 (1983)

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Above Threshold Ionization Results (cont.)

PHYSICAL REVIEW A 87, 013421 (2013)

Classical-quantum correspondence in atomic ionization by midinfrared pulses: Multiple peak and interference structures

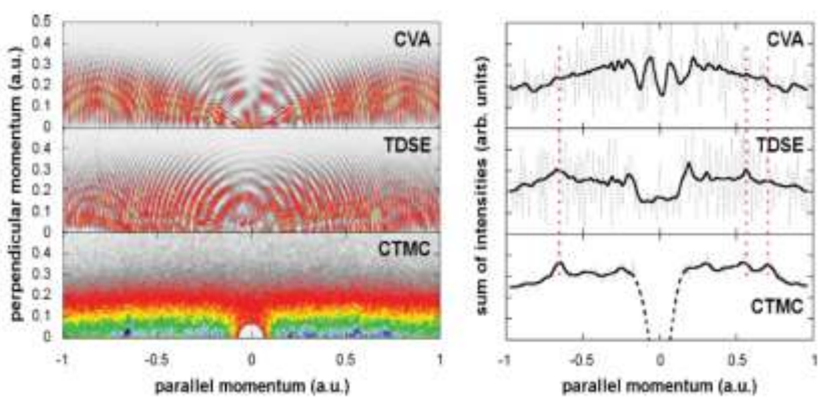

FIG. 1. (Color online) Momentum distributions (linear color scale) after interaction of a strong ($I = 10^{14}$ W/cm^2) midinfrared ($\lambda = 3200$ nm) laser pulse with a hydrogen atom. The distributions were calculated using the CVA (top), full quantum simulation (middle), and CTMC (bottom); the total pulse duration is eight cycles. Left: two-dimensional distributions; right: projection onto $p_\parallel$.

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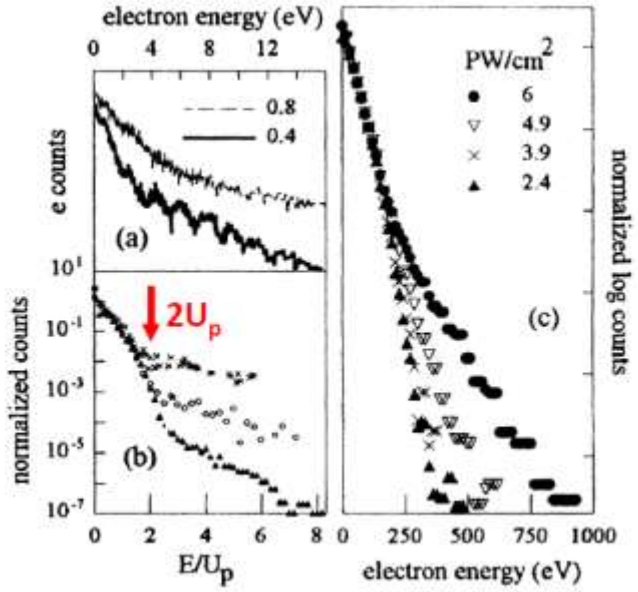
Above Threshold Ionization Results (cont.)



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

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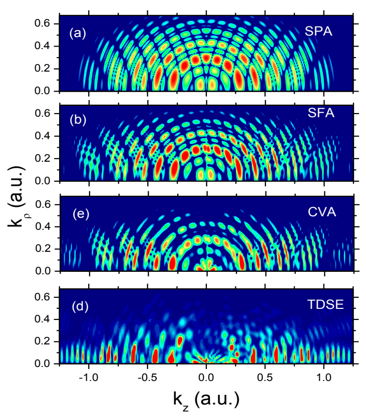
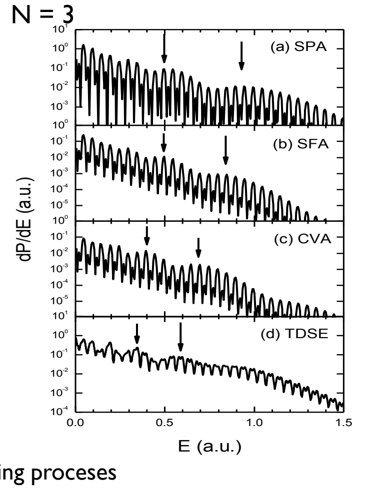
Above Threshold Ionization Results (cont.)

Comparison among different computation methods

E_0 = 0.0675, ω = 0.05 (trapezoidal pulse with half a cycle ramp on and off)

N = 3





TDSE considers
- rescattering proceses
- depletion of the ground state
- excited bound states

Nucl. Instr. Meth. B **279**, 24 (2012)

Above Threshold Ionization Results (cont.)

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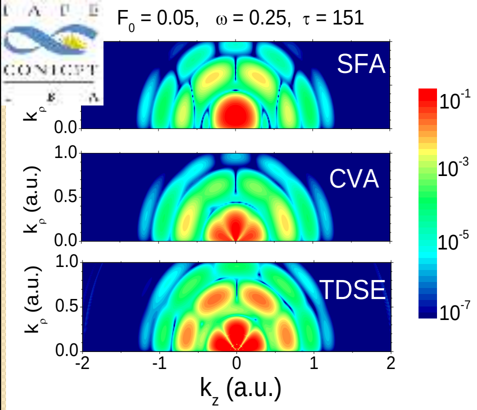

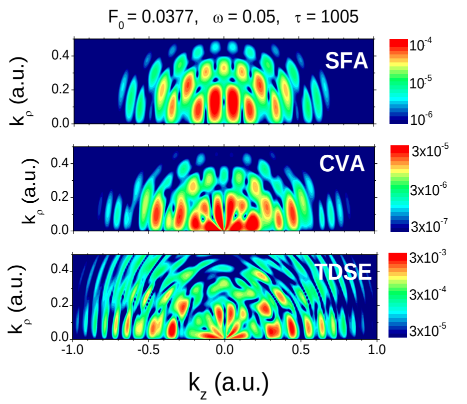

Coulomb potential → Formation of the bouquet

SFA is accurate for short-range potentials (negative ions)

Above Threshold Ionization Results (cont.)

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Photodetachment of negative ions

Reichle *et al*, PRA **68**, 063404 (2003)

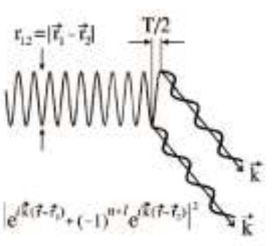


FIG. 10. Schematic picture of the origin of interference in electron emission from two laser induced point sources.

Two-slit interference:

n: minimum number of photons to reach the continuum
l: initial state angular momentum

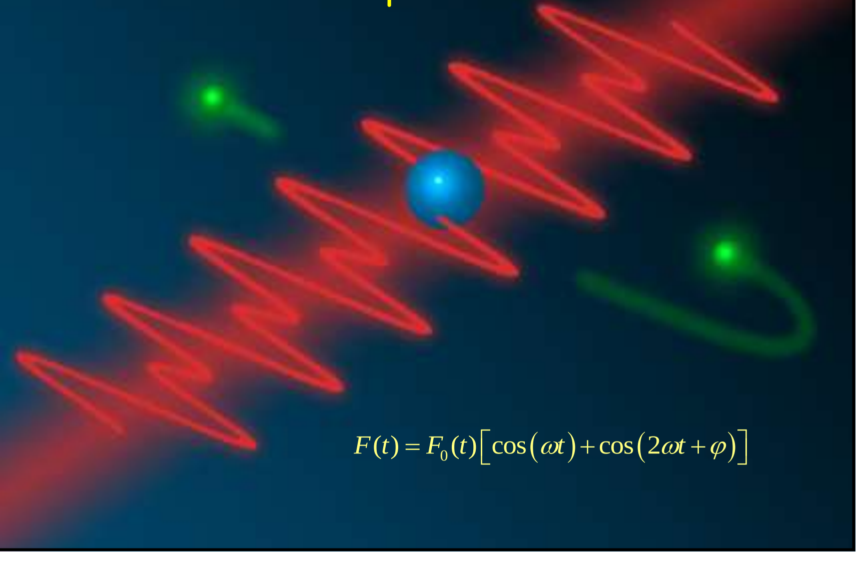
Dominant angular momentum near threshold:
***n+l* is even → s-wave**
***n+l* is odd → p-wave**

Wigner threshold law

Classical trajectories (negative ions): wiggling around straight lines

Photoionization by two-color lasers

Control of sub-cycle interference patterns with a sculpted laser field



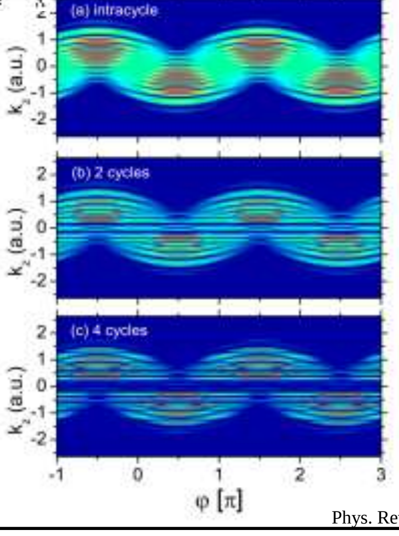
$$F(t) = F_0(t)\left[\cos(\omega t) + \cos(2\omega t + \varphi)\right]$$

Photoionization by two-color lasers

Two-color Laser Theory: Single ionization of H

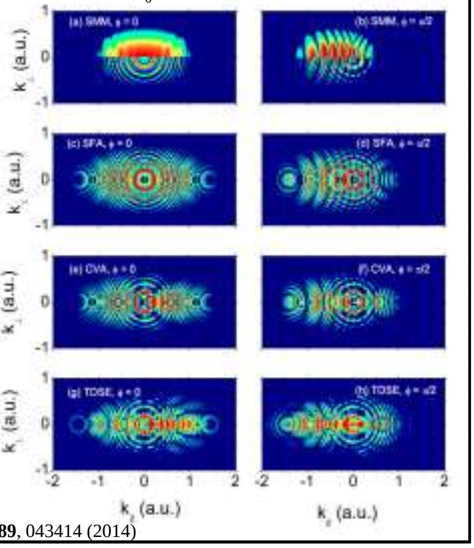
SMM

$F_0 = 0.075 \quad \omega = 0.057$



$$F(t) = F_0(t)\left[\cos(\omega t) + \cos(2\omega t + \varphi)\right]$$

$F_0 = 0.053 \quad \omega = 0.057$



Phys. Rev. A **89**, 043414 (2014)

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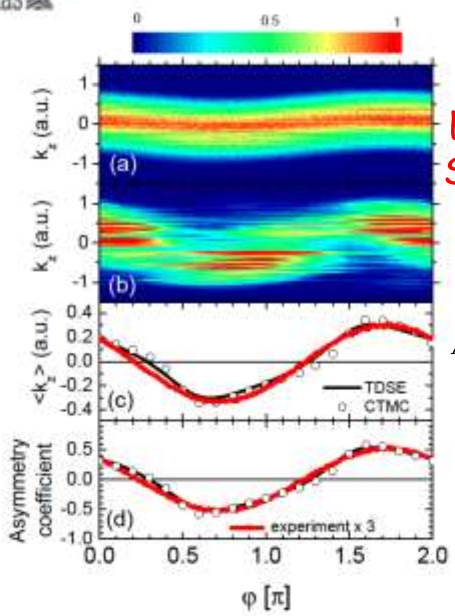
Photoionization by two-color lasers



Two-color Laser Experiment & Theory: Single ionization of He

$$A(\varphi_{CE}) = \frac{W_{\varphi_{CE}}(0^o) - W_{\varphi_{CE}}(180^o)}{W_{\varphi_{CE}}(0^o) + W_{\varphi_{CE}}(180^o)}$$

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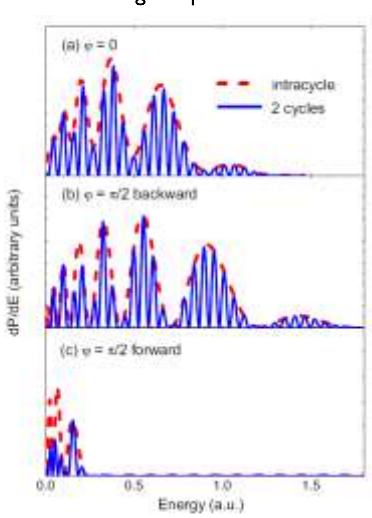
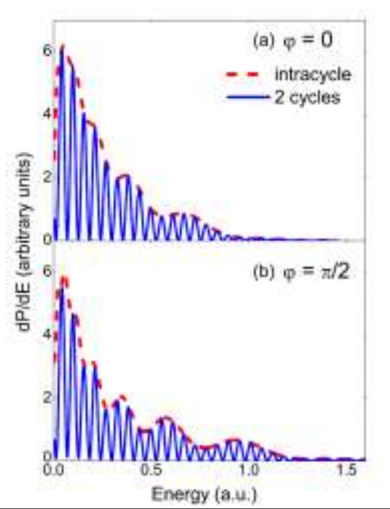
Photoionization by two-color lasers

Along the polarization axis

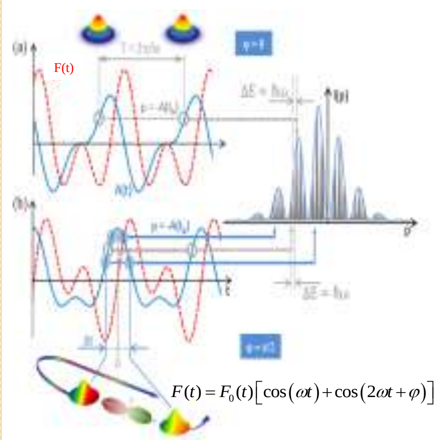


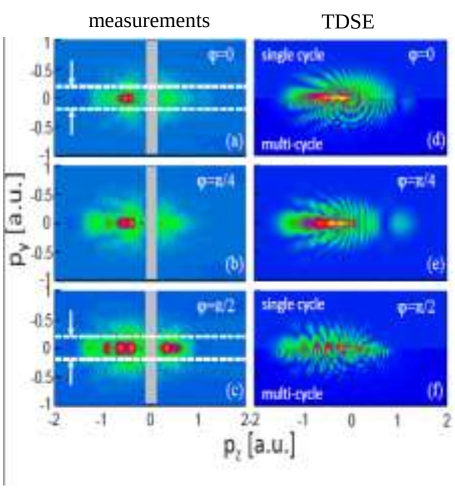
Angle integrated



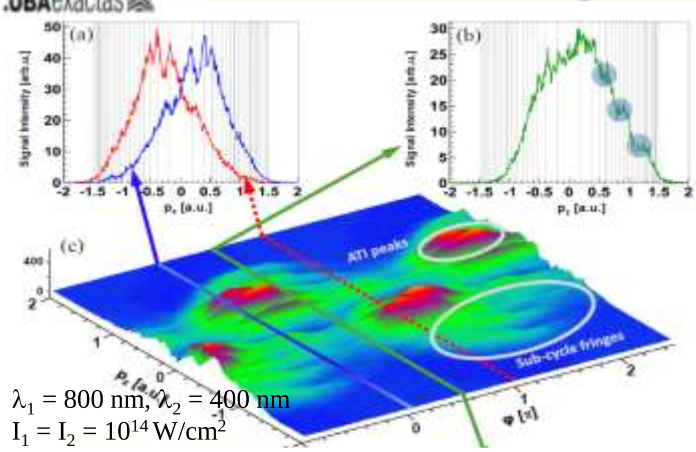
Photoionization by two-color lasers

Two-color Laser Experiment & Theory: Single ionization of He





Photoionization by two-color lasers



(a) Measured ion momentum spectra of He^+ along the laser polarization direction for $\varphi = 0$ (blue) and π (red). The thin vertical lines mark the position of the intercycle peaks.

(b) Same as (a) but for $\varphi = \pi/2$. The blue shaded dots mark the sub-cycle interference patterns.

(c) Intercycle and sub-cycle interference fringes extracted from the measured ion momentum spectra as a function of the longitudinal momentum p_z and the relative phase φ.

(d) Same as (c) but calculated using the TDSE. The upper half of the diagram ($p_z > 0$) shows the simulated spectrum for a single-cycle pulse, the lower half ($p_z < 0$) for a multi-cycle pulse.

(e) Comparison of normalized spectra for $\varphi = \pi/2$, calculated using the TDSE for a multi-cycle pulse (gray line) and single-cycle pulse (red line) with the measured spectrum (blue line).

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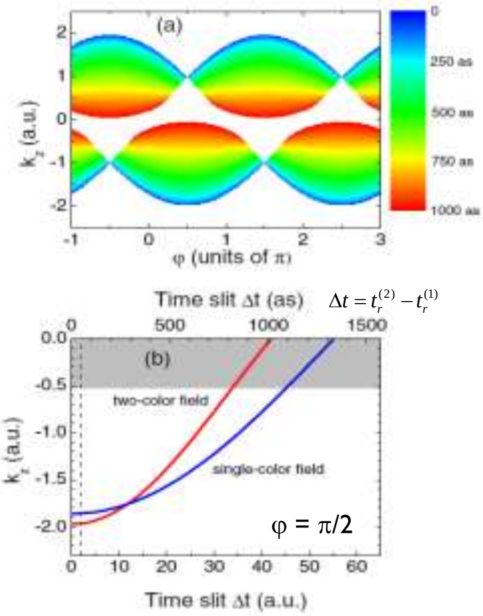
Photoionization by two-color lasers



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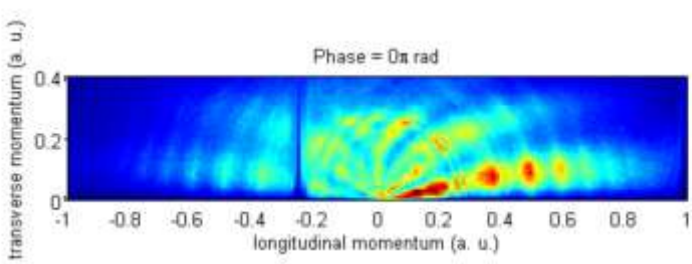
Photoionization by two-color lasers

Experiment MPI in Heidelberg:
Single ionization of Ar

$I_1 = 2.4 \times 10^{14}$ W/cm^2, $\lambda_1 = 800$ nm
$I_2 = 2.0 \times 10^{13}$ W/cm^2, $\lambda_2 = 400$ nm



Phys. Rev. A (2015, in press)

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Photoionization by two-color lasers

Experiment MPI in Heidelberg:
Single ionization of Ar

$I_1 = 10^{14}$ W/cm^2, $\lambda_1 = 800$ nm
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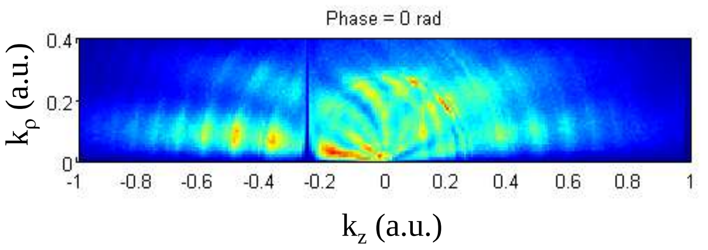

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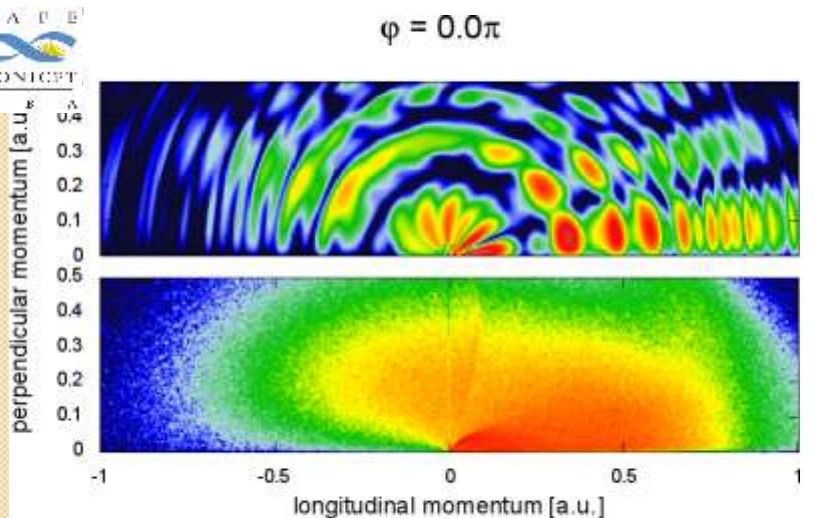
Photoionization by two-color lasers



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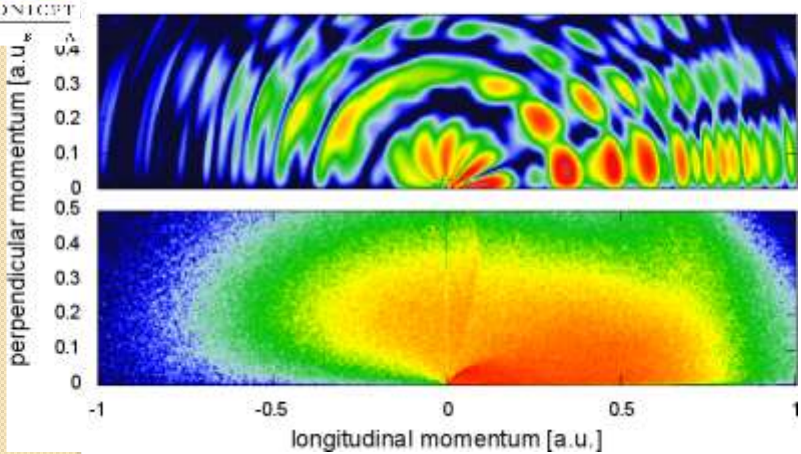
Photoionization by two-color lasers

$\varphi = 0.0\pi$



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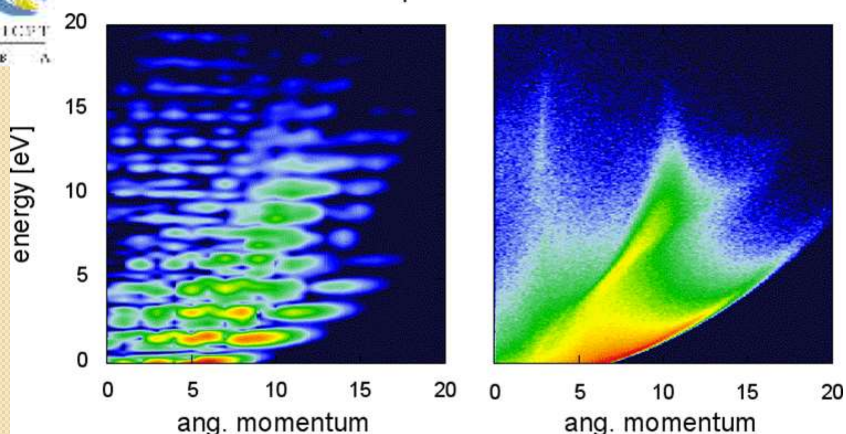
Photoionization by two-color lasers

$\varphi = 0.0\pi$



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Photoionization by two-color lasers