Time-Dependent Method in Laser Material Interactions

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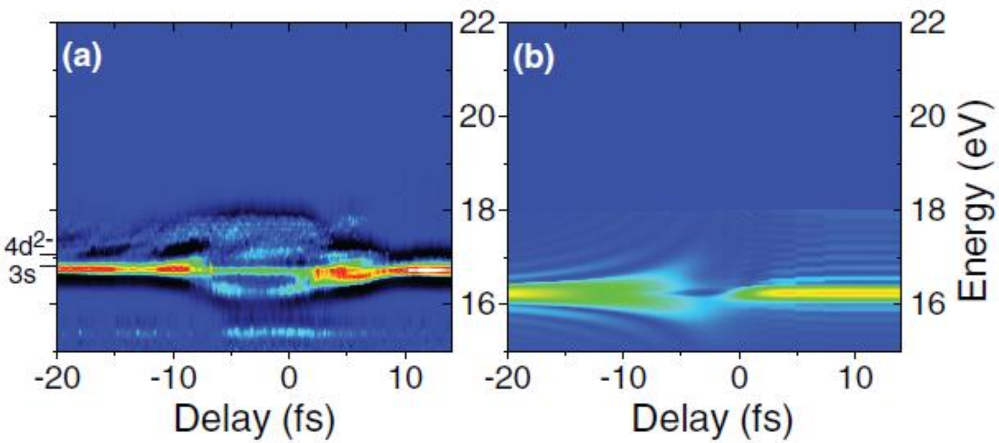
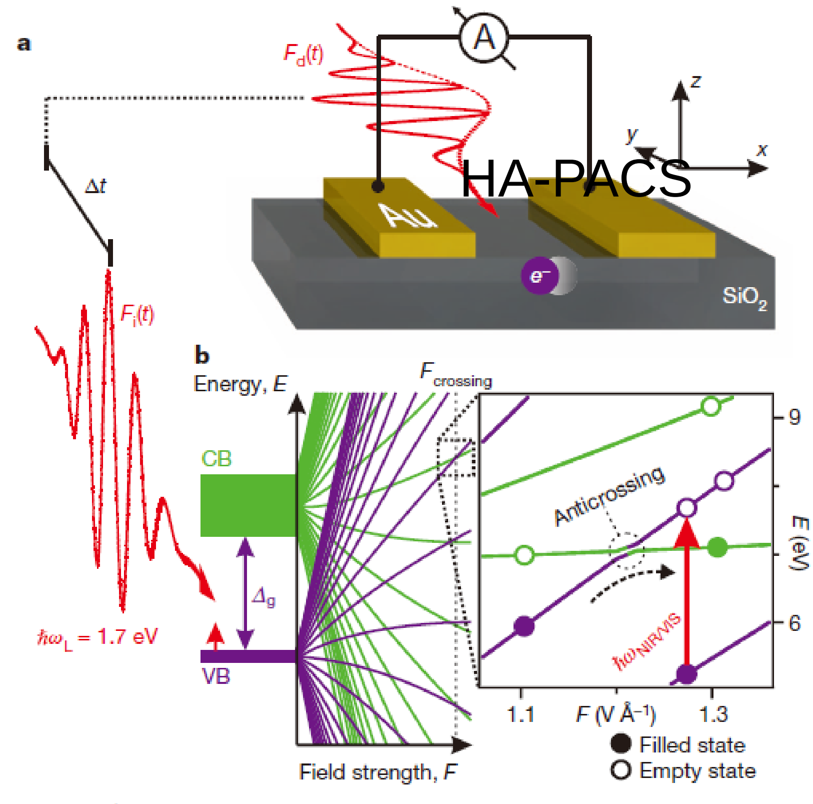
Outline:

- What we are working on
  - ***Understand*** the mechanism of laser-material interactions
  - ***Control*** material properties in an ultra-short time scale
- Computational Methods
  - Working Equations: Time-dependent Schrodinger equation (PDE)
  - Many-electron effect: Model potential, Density-functional theory
- Examples
  - Mechanism of atomic ionization in mid-infrared Laser field
  - Control transparency of a material in attosecond domain
  - Others
- Future Plan
  - Develop a numerical tool for a many-electron system
  - Investigate dynamics in a many-electron system

Our Research Goals: Control



PRA **87** (2013) 063413.



Insulator ➔ conductor (1 fs)

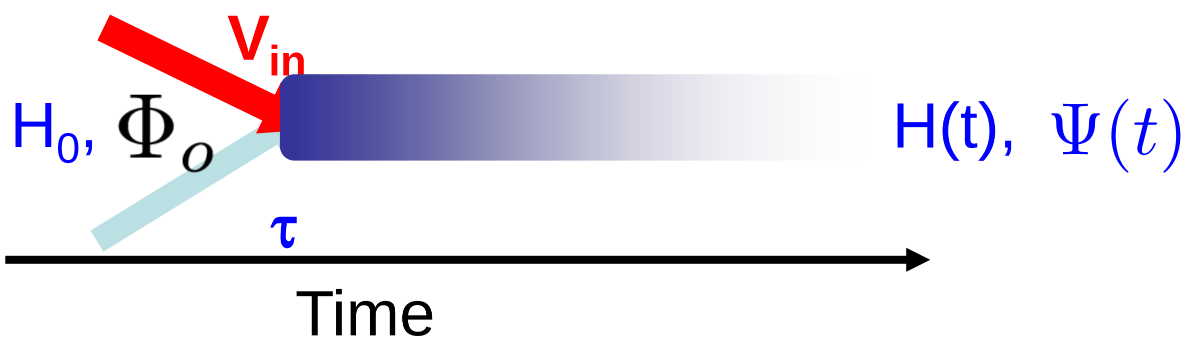
A. Schiffrin, *et. al.*, Nature, (2012)

TDSE in differential form

$$i\frac{\partial}{\partial t}\Psi(t) = H(t)\Psi(t) \quad \text{with} \quad \Psi(t=-\infty) = \Phi_0$$

TDSE in integral form

$$\begin{aligned}\Psi(t) &= -i\int_{-\infty}^{t} e^{-i\int_{\tau}^{t} H(t')dt'} V_{in}(\tau) e^{-iH_0\tau}\Phi_0 d\tau + e^{-iH_0 t}\Phi_0 \\ &= -i\int_{-\infty}^{t} U(t,\tau)V_{in}(\tau)U_0(\tau,-\infty)\Phi_0 d\tau + U_0(t,-\infty)\Phi_0\end{aligned}$$

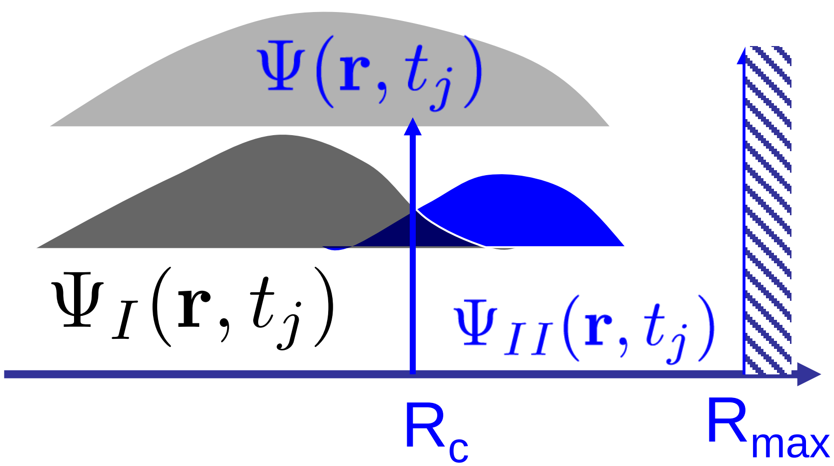


Theoretical Method: Boundary

Dynamics-related wavefunction

$$\Psi(t) = -i\int_{-\infty}^{t} U(t,\tau)V_{in}(\tau)U_0(\tau,-\infty)\Phi_0 d\tau + U_0(t,-\infty)\Phi_0$$

Repartition of the wavefunction $\Psi(\mathbf{r},t_j) = \Psi(\mathbf{r},t_j)(1-f(r)) + \Psi(\mathbf{r},t_j)f(r)$



$\Psi_I(\mathbf{r},t_j)$ Space

$\Psi_{II}(\mathbf{r},t_j) \rightarrow \Psi_{II}(\mathbf{p},t_j)$

Momentum

[Chelkowski & Bandrauk, IJQC **60** (1996) 1685.]

Theoretical Method: Time Propagator I

Second order split-operator-method in the energy representation

$$\Psi(t+\Delta t) = U(t+\Delta t, t)\Psi(t) = e^{-iH\Delta t}\Psi(t)$$
$$\approx e^{-iH_0\Delta t/2} e^{-iV(t)\Delta t} e^{-iH_0\Delta t/2}\Psi(t) + O(\Delta t^3)$$

[Chem. Phys. **217** (1997) 119]

Discretize space in pseudo-spectral grid:

$$H_0 = H^0_{r_i,r_j}(\ell), \quad \Psi(r_i,\theta) = \sum_\ell R_\ell(r_i) Y_{\ell,m}(\hat{\mathbf{r}}), \quad \Psi(r_i,\ell) = R_\ell(r_i)$$

Time-propagation → vector, matrix operations → blas

Easy refactor to modern computers, GPU cublas, MIC ??

Theoretical Method: Time Propagator I

Time propagators:

$$U(t+\Delta t, t) = e^{-iH_o\Delta t/2} e^{-iV_{in}(t)\Delta t} e^{-iH_o\Delta t/2}$$ numerical

$$U_v(t, t_i) = e^{-i\int_{t_i}^{t} (\mathbf{p}-\mathbf{A}(t'))^2/2dt'}$$ analytical

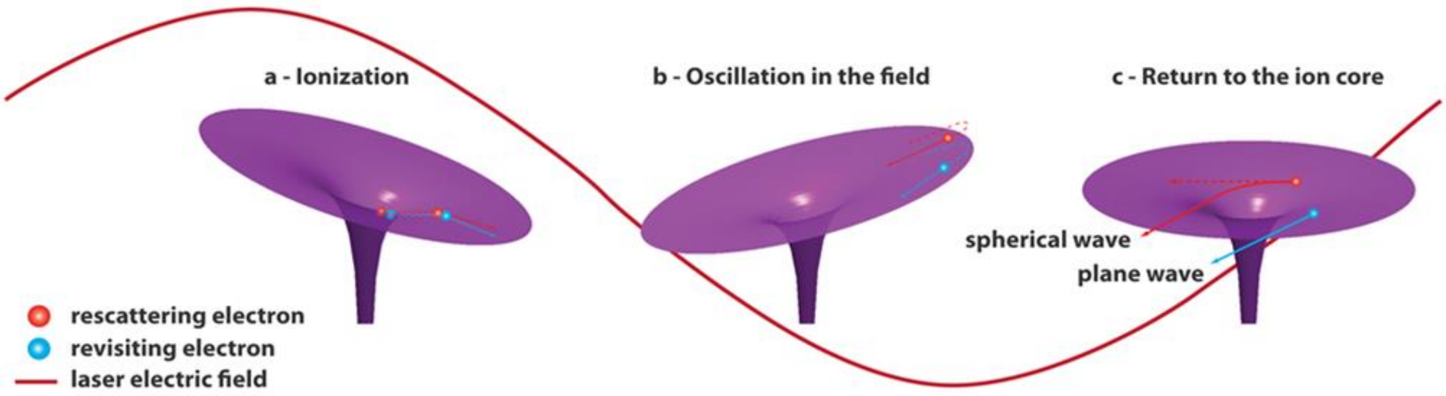
$$U_0(t, t_i) = e^{-iE_o(t-t_i)}$$ analytical

$$\Psi(t) = \Psi(\mathbf{r}, t) + \Psi_c(\mathbf{p}, t)$$

Example 1: Mechanism of ATI in mid-IR Field

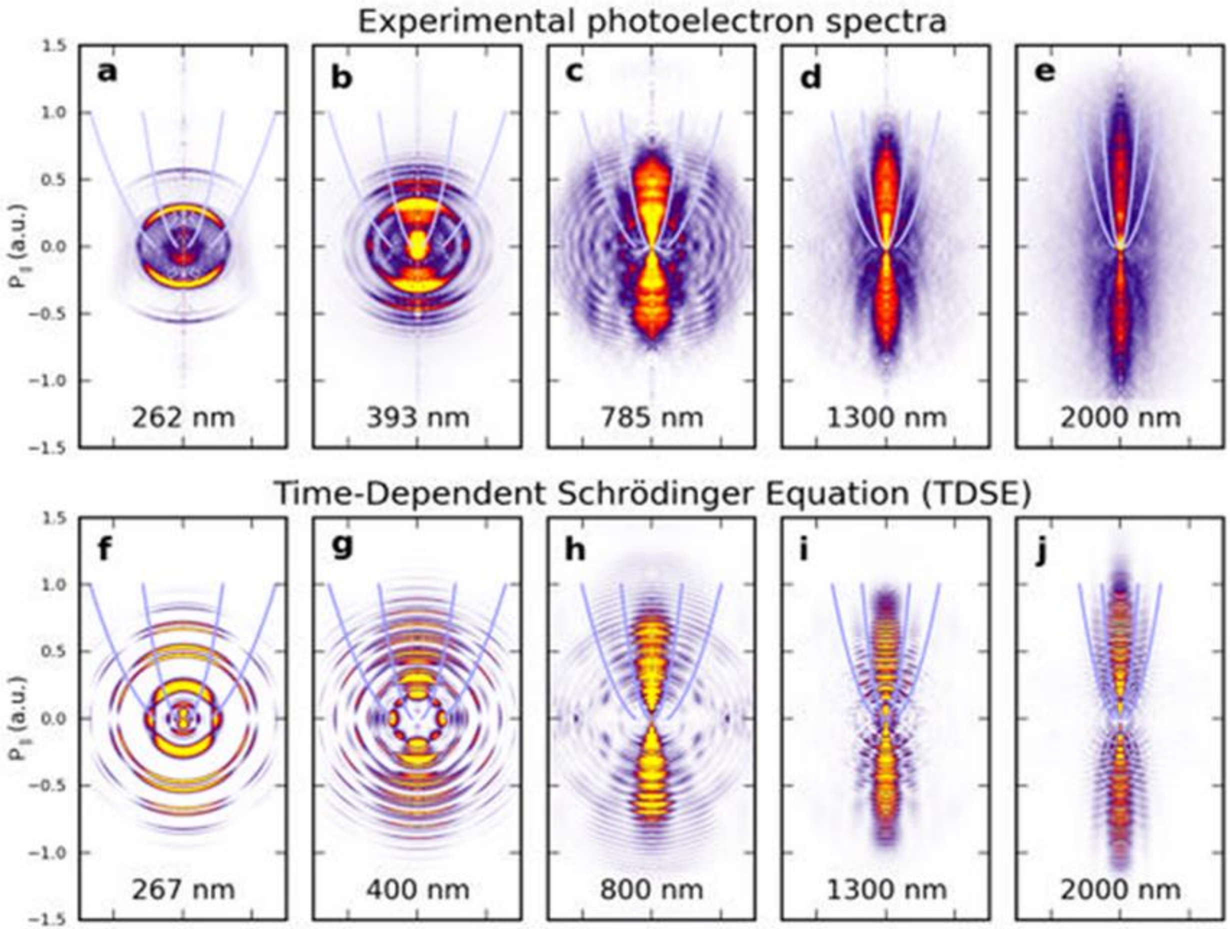
ATI: above-threshold ionization

Physical Process: $A + n\,h\nu \rightarrow A^{+} + e$



- understand the structure:
  interference between the returning and rescattering electrons
- Information encoded in the structure:
  inner-work: how the electron interacts with the parent core

Example 1: Mechanism of ATI in mid-IR Field

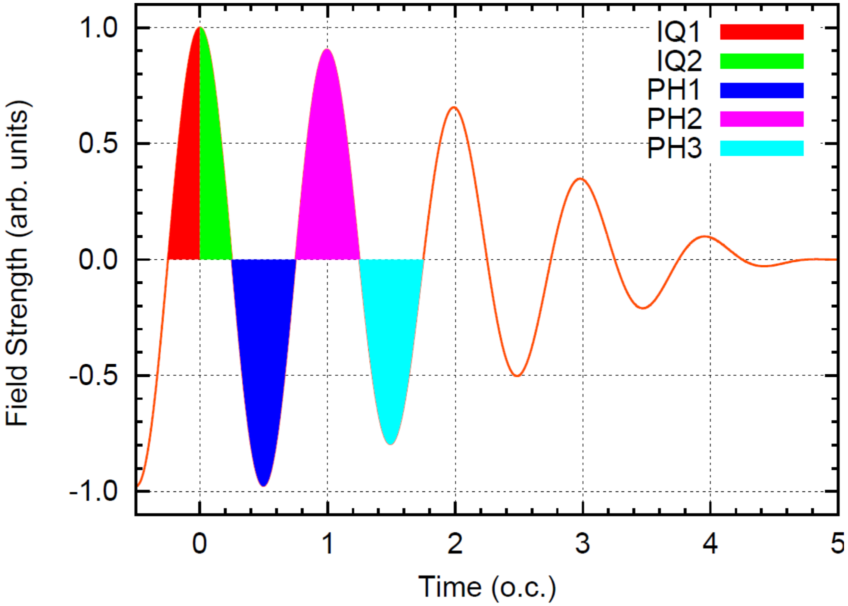


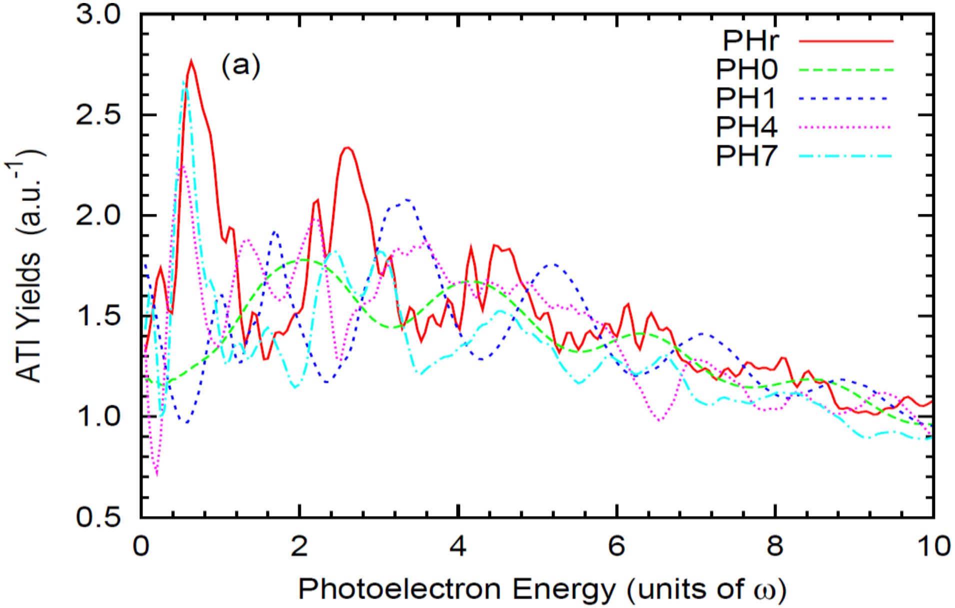
PRL 109 (2012) 073004.

HA-PACS: 5 nodes, 20 hrs

Separate the tunnel ionization from propagation:

$$\Psi_k = U(\infty, t_{k+1}) \int_{t_k}^{t_{k+1}} U(t_{k+1}, t) V_{ext}(t) e^{-iH_0 t} \Psi_0 dt$$





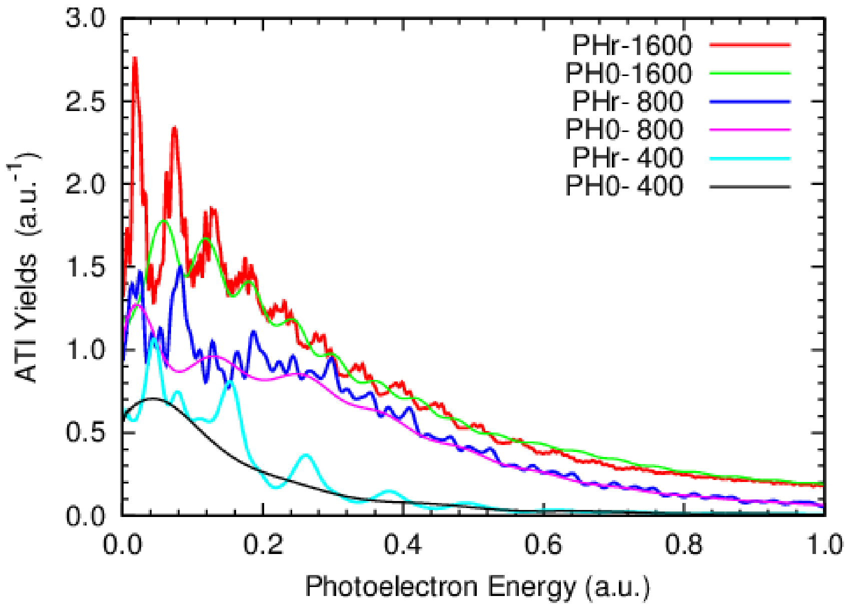
Low Energy Structure comes from multiple re-scattering.

Example 1: Mechanism of ATI in mid-IR field

Multiple re-scattering also exists for other wavelengths.
But no experimental report on LES
in 800 nm or shorter. Why?

TABLE I. The ratio of the tunnel ionization probability to the total ionization probability ionized in a half cycle. ($I_0 = 10^{14}$ W/cm^2).

	0.5 I_0	1.0 I_0	1.5 I_0	2.0 I_0
400 nm	0.055	0.391	0.614	0.727
800 nm	0.570	0.812	0.931	0.969
1600 nm	0.970	0.995	1.000	1.000

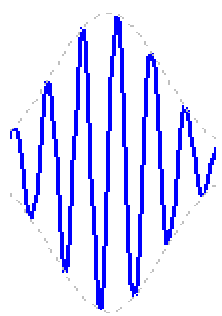


Two conditions to observe LES:

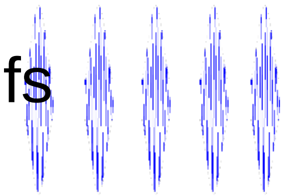
- multiple scattering (exist for all laser wavelengths)
- tunnel ionization (dominant only for longer laser wavelengths)

Physical Processes

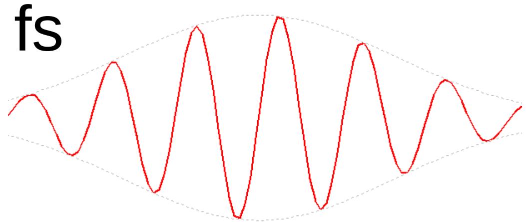
SAP: 0.3 fs

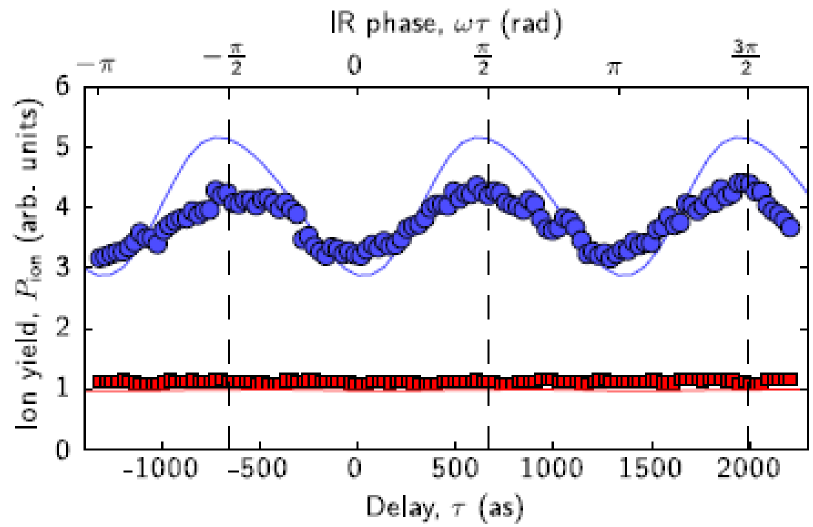


EUV: APT: 10 fs



IR field: 45 fs



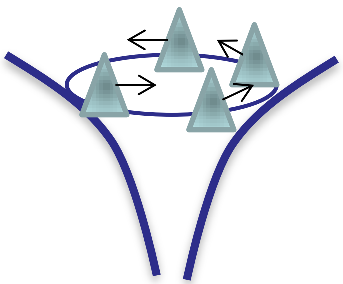


Johnsson *et al.*, PRL **99**, 233001 (2007)

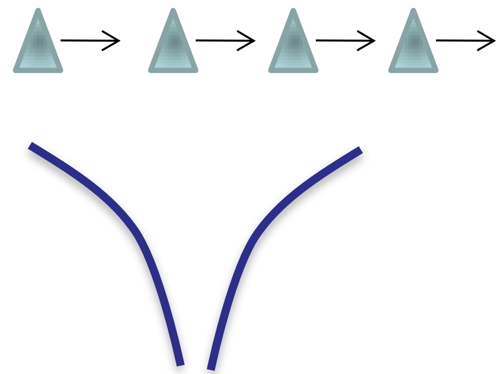
Oscillating or Not ?

Observation: Ionization yield depends on the time-delay for He, not for Ar.

Example 2: Interference between Different APs



He atoms



Ar atoms

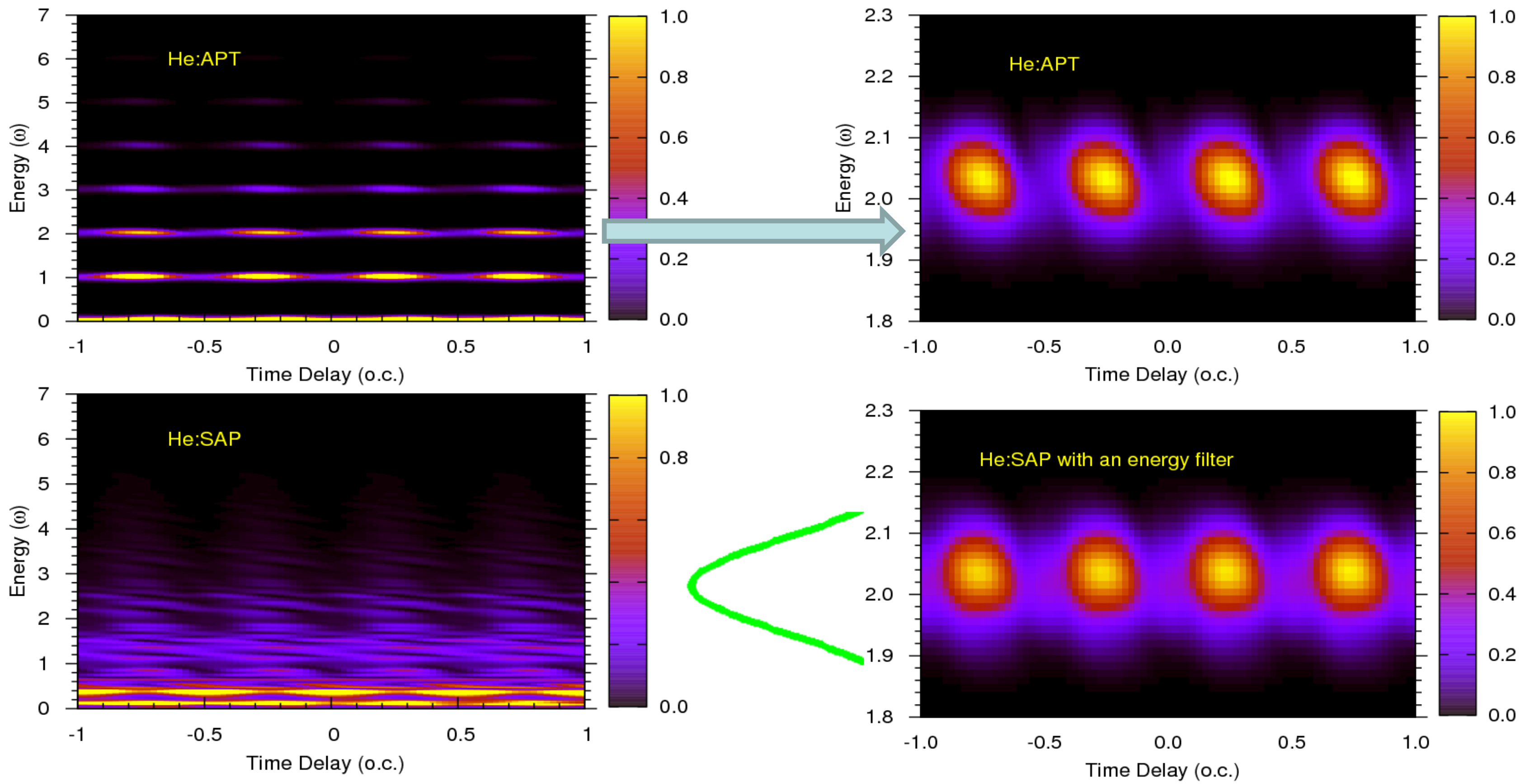
Predictions: total yields

	He	Ar
APT	Yes	No
SAP	No	No

For a given ATI energy

	He	Ar
APT	Yes	?
SAP	?	?

Example 2: Other Evidences



For a specified ATI peak, oscillation *always* exists.

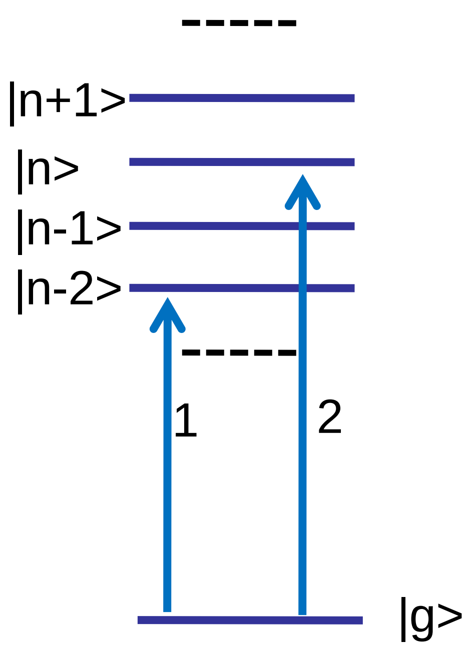
Conclusion: New mechanism is needed!

Example 2: Mechanism

Our proposed mechanism: [PRA 81 (2010) 021404(R)]

- IR field dressed atomic states -- Floquet states
- XUV excites an atom to a Floquet state
  through different sidebands.

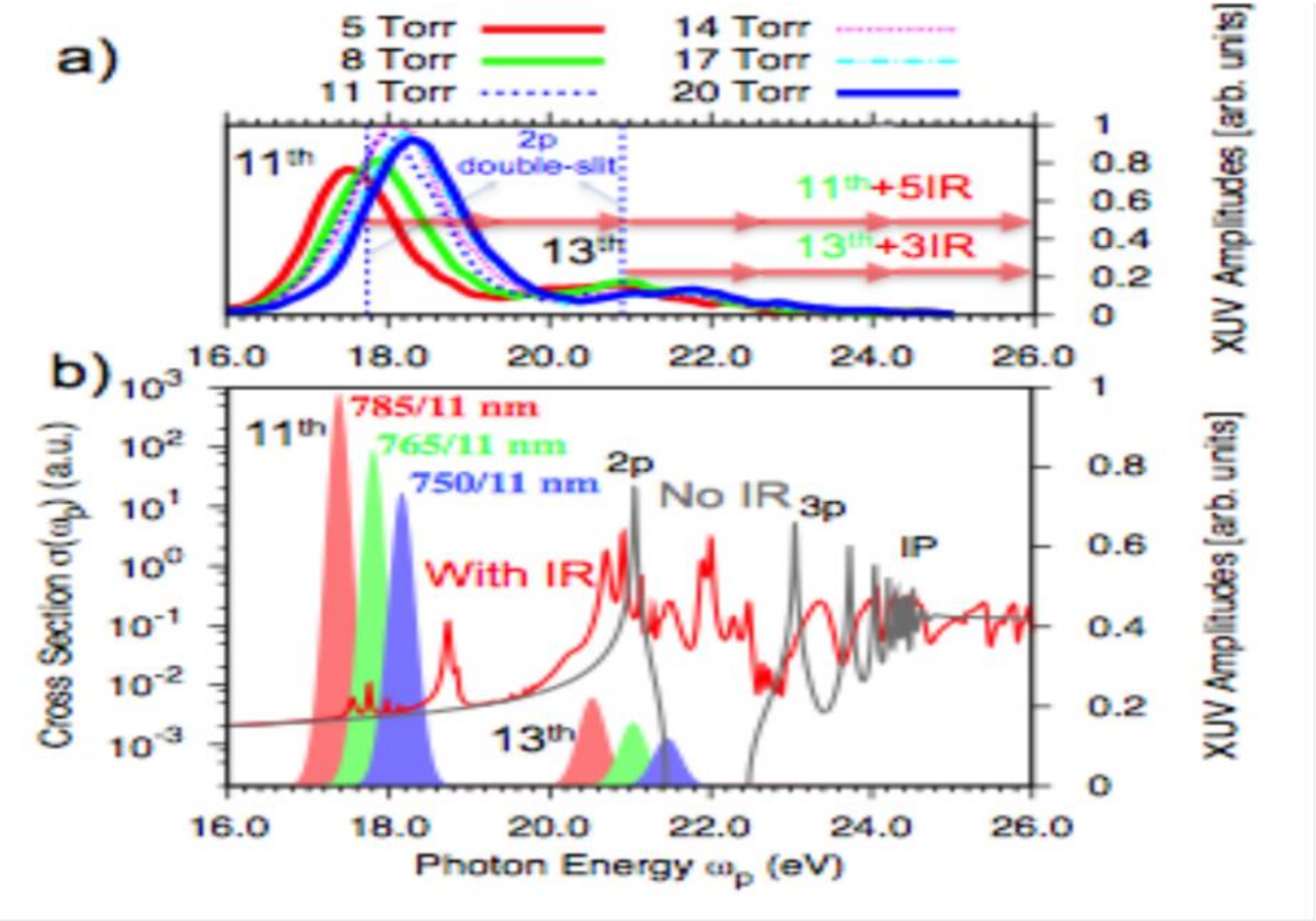
$$\Psi_\alpha(\mathbf{r},t) = e^{-i\epsilon_\alpha t} \sum_{n=-\infty}^{\infty} e^{-i\ n\omega t} \phi_{\alpha,n}(\mathbf{r})$$



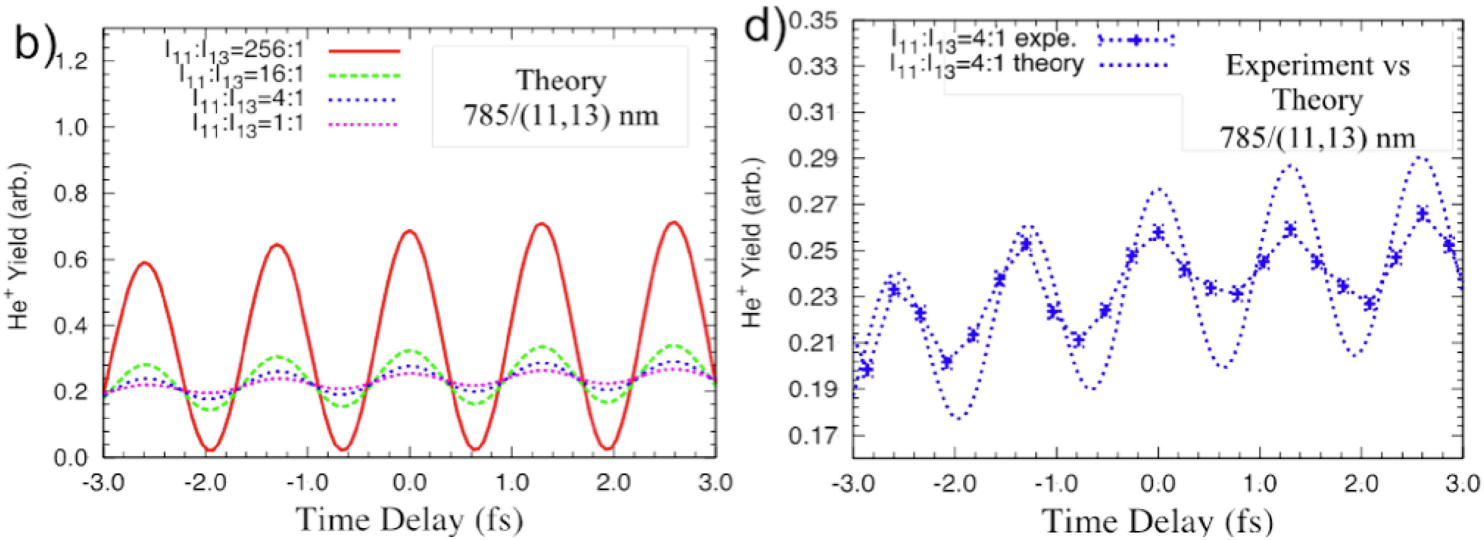
- The interference between the transitions to different side bands results the oscillation.

$$P(t_d) \propto \sum_\alpha |M_1^\alpha F_1 + M_2^\alpha F_2 e^{-i2\omega t_d}|^2$$

Example 2: IR Assisted Photoionization



Example 2: IR Assisted Photoionization



Generate a super-fast optical switch

Other Works

- Visualization of multiple-scattering in ATI spectra
  PRL **108** (2012) 193002.
- ✓ Control Molecular Dynamics from fs to as,
  PNAS, **111** (2014) 912.
- ✓ Attosecond-Resolved Evolution of a Laser-Dressed Helium Atom
  PRL **108** (2012) 193002.
- ✓ Laser enabled Auger decay in rare-gas atoms,
  PRL **106** (2011) 053002.
- ✓ Attosecond Streaking in the Low-Energy Region as a Probe of Rescattering, PRL **107** (2011) 183001.

- Breit interaction on dielectronic recombination in HCI
  PRL **108** (2012) 073002.
- Anomalous bumpy structures in the capture cross-section of antiprotons by helium atoms, PRL **101** (2008) 163201.

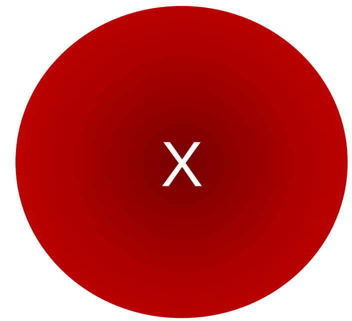
Future Plan: Limitation of the Present Method

Spherical coordinate: waste too much grid point

$$N \propto \lambda^3$$

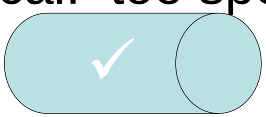
$$z_{max} \propto E_0 \cdot \lambda^2$$

$$U_p \propto E_0^2 \cdot \lambda^2$$

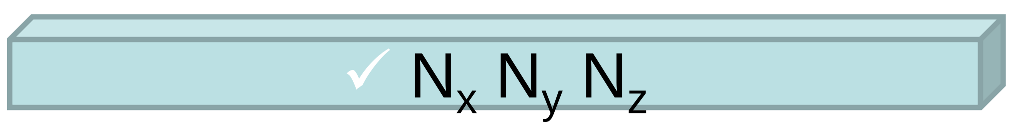


E0 = 0.1: 800 nm -> 30. a.u. 10,000 nm -> 4,000 a.u.

cylindrical: too specified (symmetry)



Cartesian: works for atoms, molecules and clusters



50x50x10,000 → GPU 5GB

Future Plan: Develop a New Method

✧ Develop a general method to solve TDSE using split-operator-method in Cartesian coordinate + FFT

$$\Psi(t+\Delta t) = e^{-iH\Delta t}\Psi(t) \approx e^{-iT\Delta t/2} e^{-iV(t)\Delta t} e^{-iT\Delta t/2}\Psi(t)$$

one time-step propagation: 4x 3DFFT + 3x Multiplications $N \ln N$

- eigen-value problems: $\Delta t \ \rightarrow \ -i\Delta t$
  sub-space → time-propagation → eigenvalue → time-propagation..
  $$\{e^{-H\tau}\psi_i(0)\} \rightarrow \{\psi_i(\tau)\} \rightarrow \{\langle\psi_i|H|\psi_j\rangle\} \ \rightarrow \cdots\cdots$$
- real time-dependent problems.
  $$\{\psi_i(t)\} \ \rightarrow \ \{\psi_i(t+\Delta t) = e^{-iH(t)\Delta t}\psi_i(t)\} \ \rightarrow \ \{\psi_i(t+\Delta t)\}$$
  $$\{\psi_i(t+\Delta t)\} \ \rightarrow \ \{\rho(t+\Delta t)\} \ \rightarrow \ \{V(t+\Delta t)\}$$
- one time-step for 64x64x4096 0.6 sec (8-core Sandy Bridge)

Future Plan: Status

✧ Eigen-value problems: tested examples:

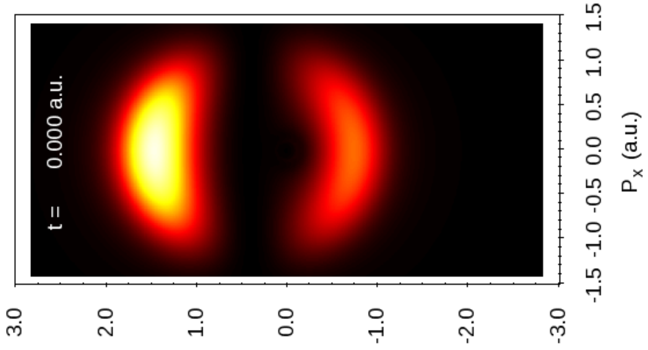
- harmonic potential: ✓
- H atoms with atomic model-potential: ✓
- H_2^+ molecular ions with atomic model-potential: ✓
- H_2 molecules with atomic model-potential: ✓

✧ Extend the above method to a ***many-electron*** system using DFT

- Poisson Equation: FFT-3D $\rho(\mathbf{r}) \rightarrow V_c(\mathbf{r})$ ✓
- Exchange-correlation potential: $\rho(\mathbf{r}) \rightarrow V_{ex}(\mathbf{r})$ almost
- Propagation in atomic pseudo-potential: not yet
- Boundary condition and physical insights: not yet

Future Plan: Study Dynamic Process

- Study the dynamics processes of a material in a intense laser pulse (from 200 nm to 10,000 nm)
  - Extract information from measurements.
  - Use the information for molecular holography.
  - Generate Coherent X-rays in the water window. (below carbon K edge at 4.37 nm)
  - Search a way to control a quantum process in ultra-short time scale (femto or atto-s)
  - And more ….



Acknowledgments

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• Centre for Quantum Dynamics	Griffith University
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