Time-Dependent Calculations of Strong Field Ionization and High Harmonics Generation

Anatoli Kheifets and Igor Ivanov

THE AUSTRALIAN NATIONAL UNIVERSITY

Igor Bray

Centre for Antimatter-Matter Studies

KITP 2009

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Interplay between strong field effects and many-electron correlations

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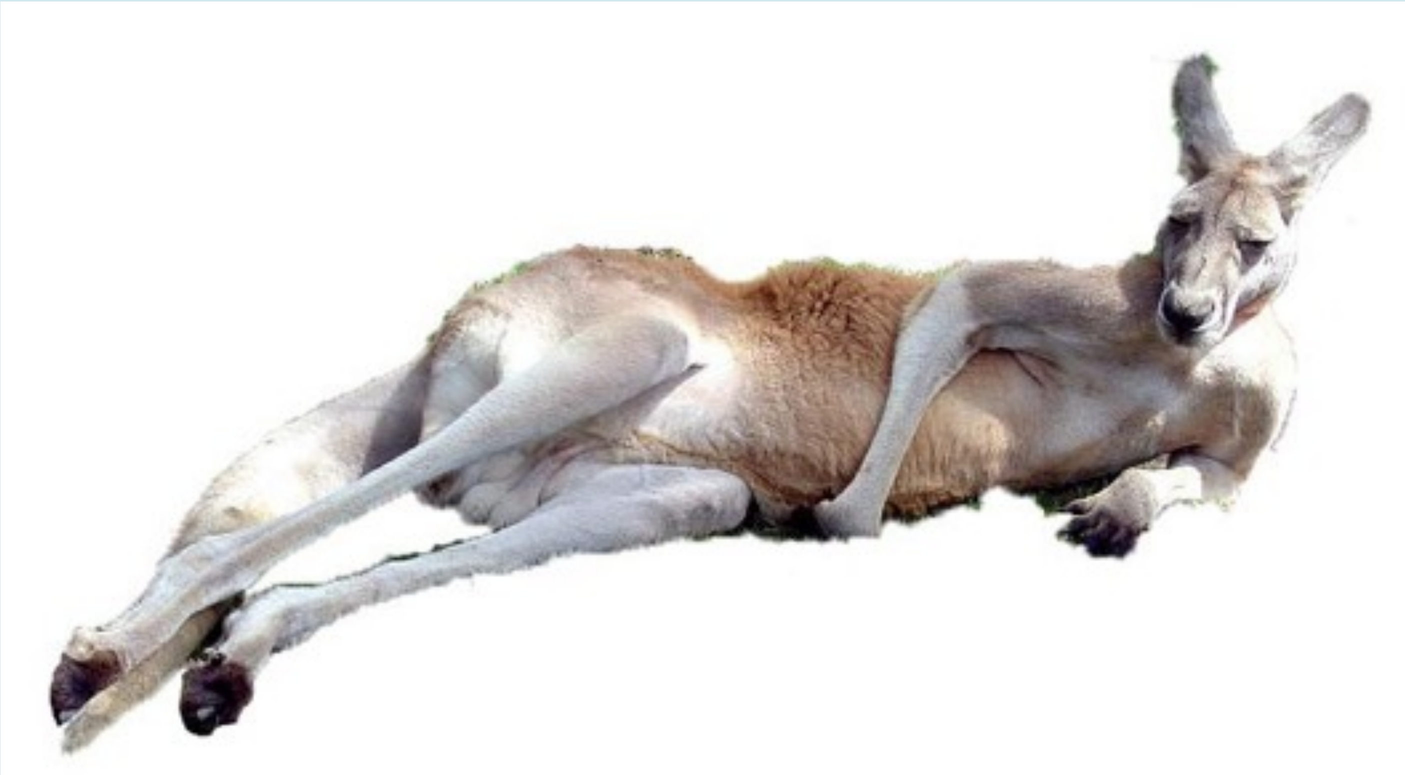

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    - FEL's (FLASH, SCSS, LCLS etc)

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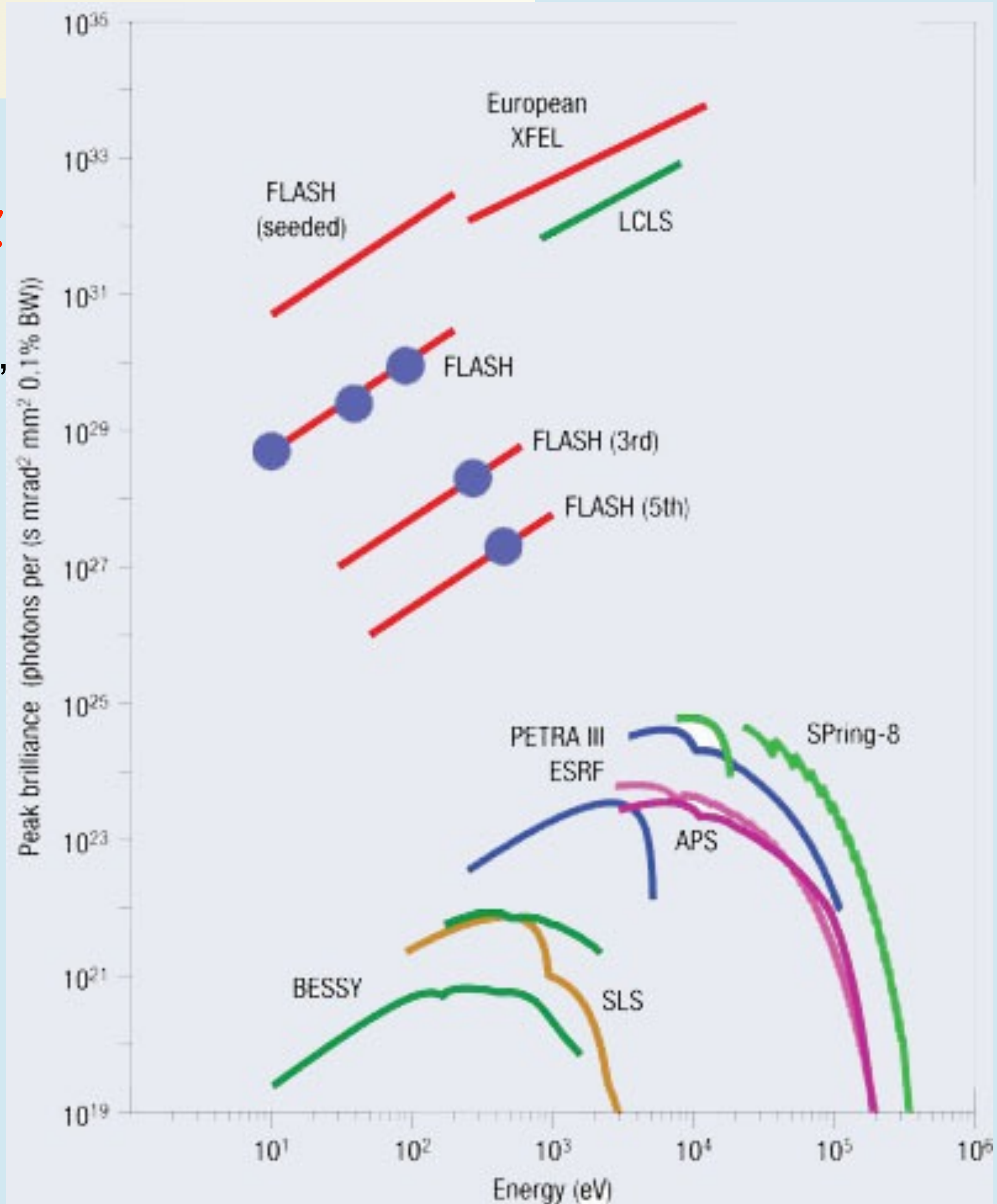

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  - Fundamental importance
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    - *Coherent, compact, intense, tunable, source of XUV radiation*
    - *Neutral gases or ionized plasmas of noble gas atoms*

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  - Race for the "table-top synchrotron"
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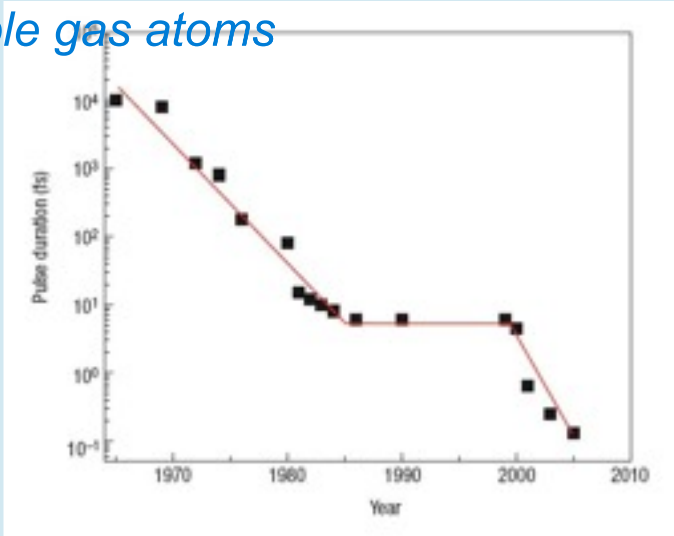
    - *Neutral gases or ionized plasmas of noble gas atoms*
- Attosecond control
  - *Steering of coherent wave packets.*
  - *Time-resolved atomic physics*



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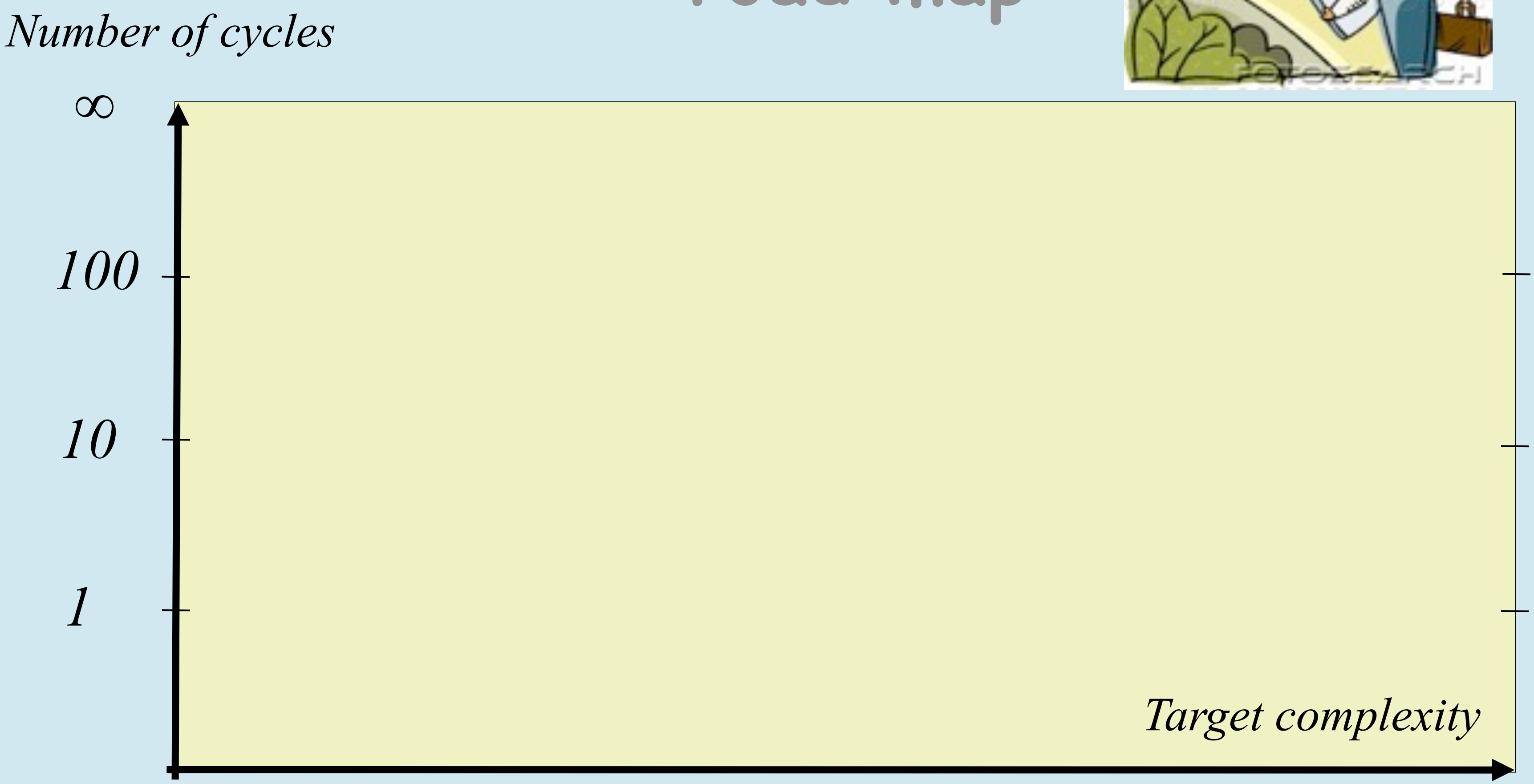
Time-dependent theory road map



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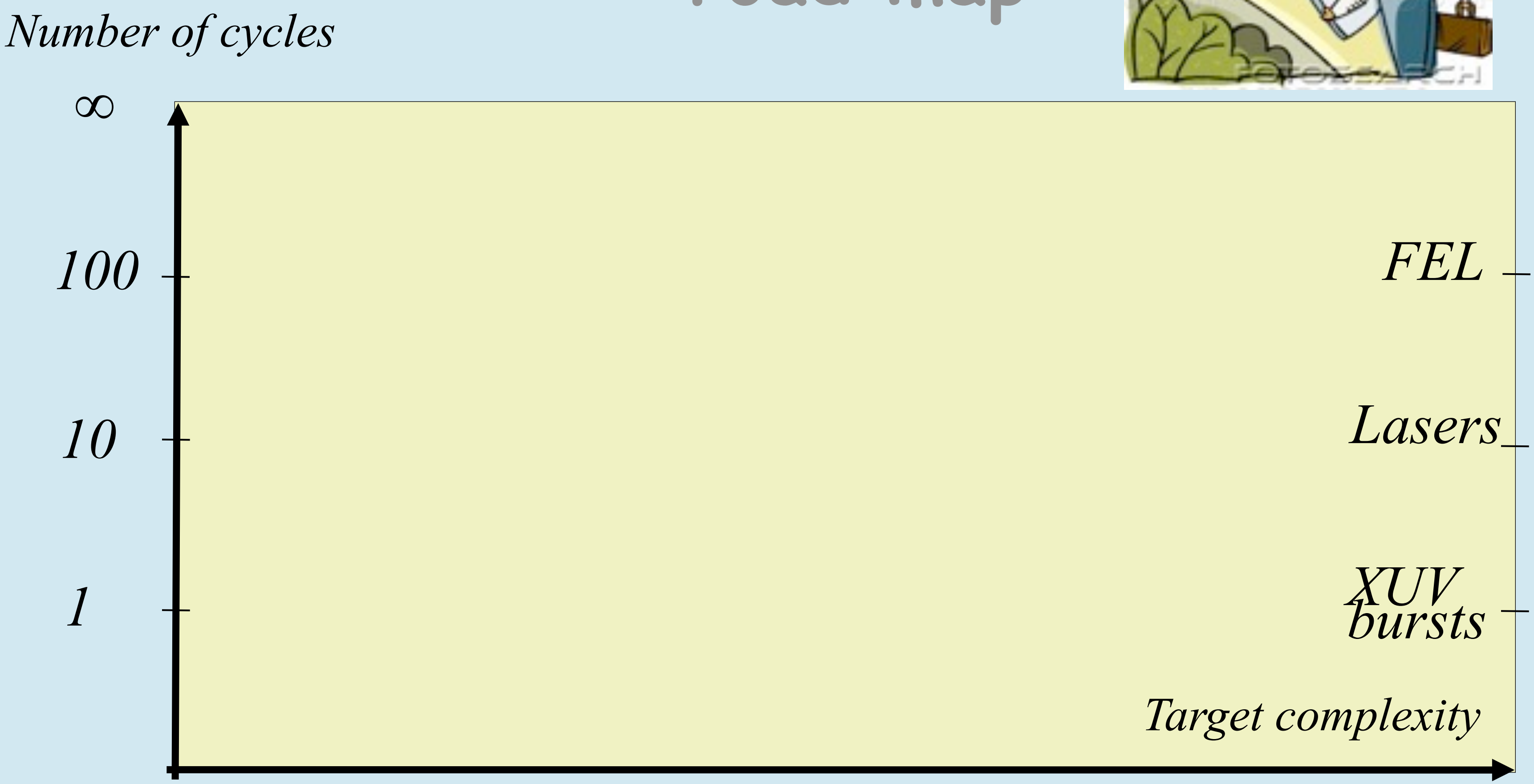

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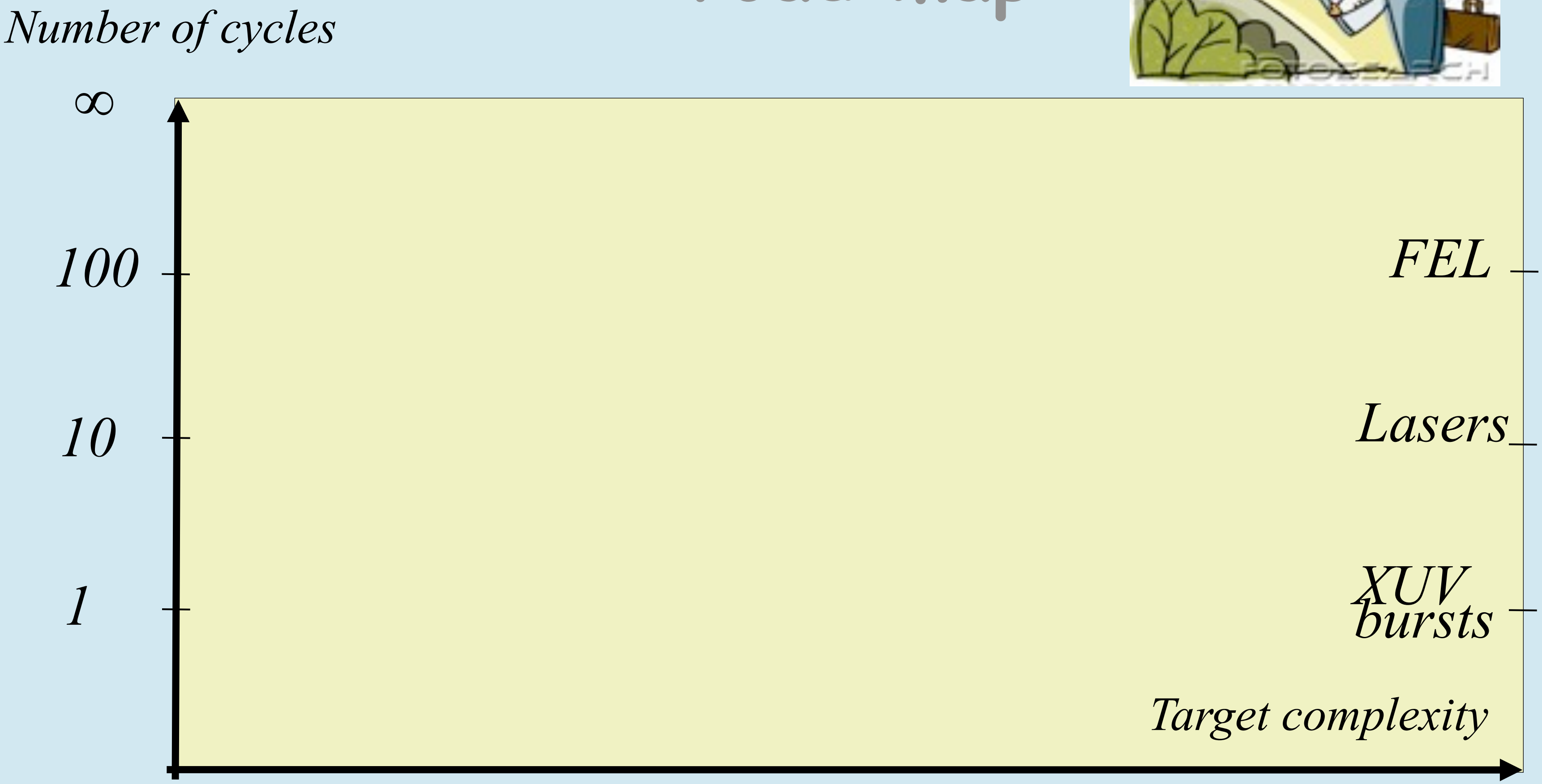

One-electron targets
Li, K, Rb

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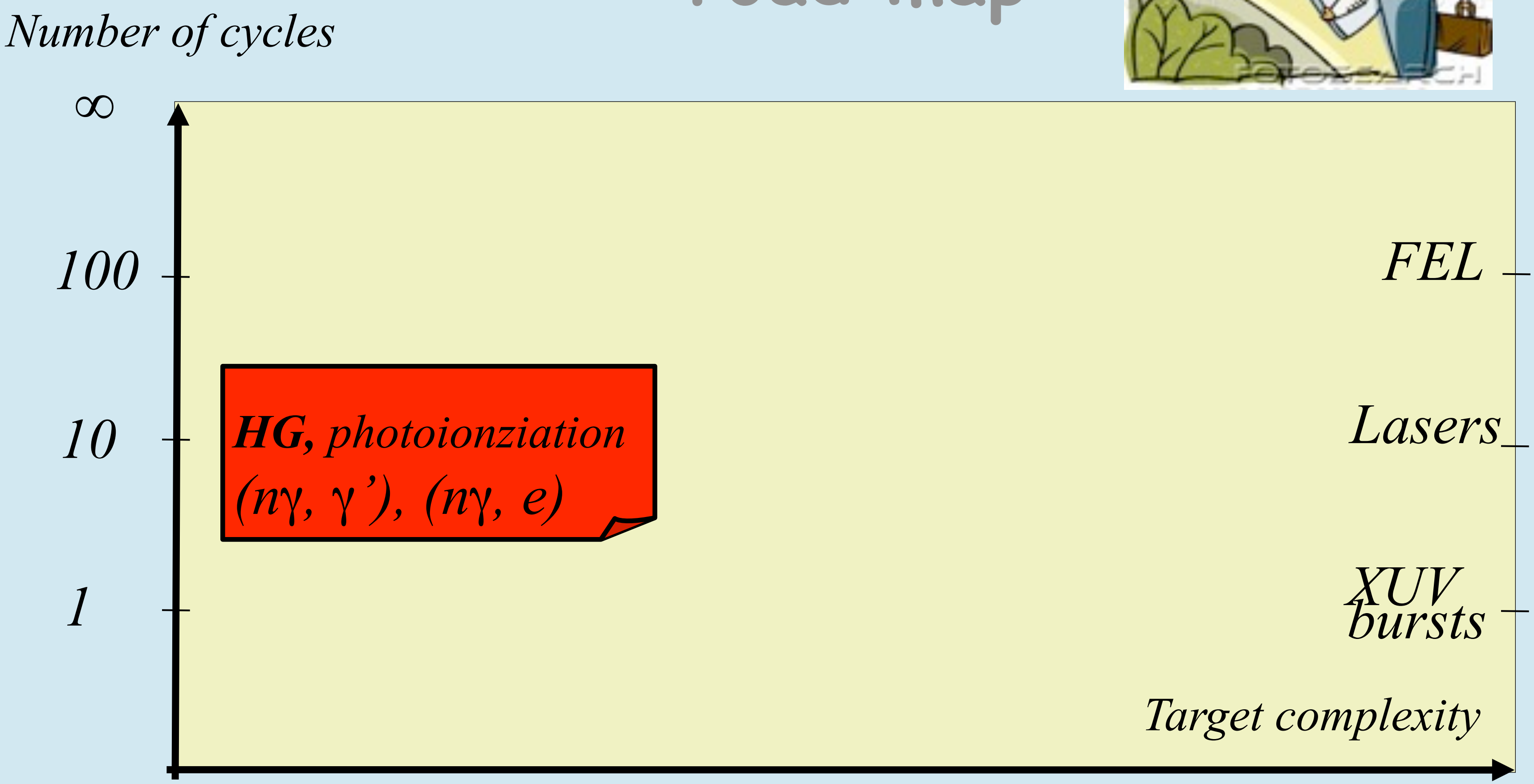

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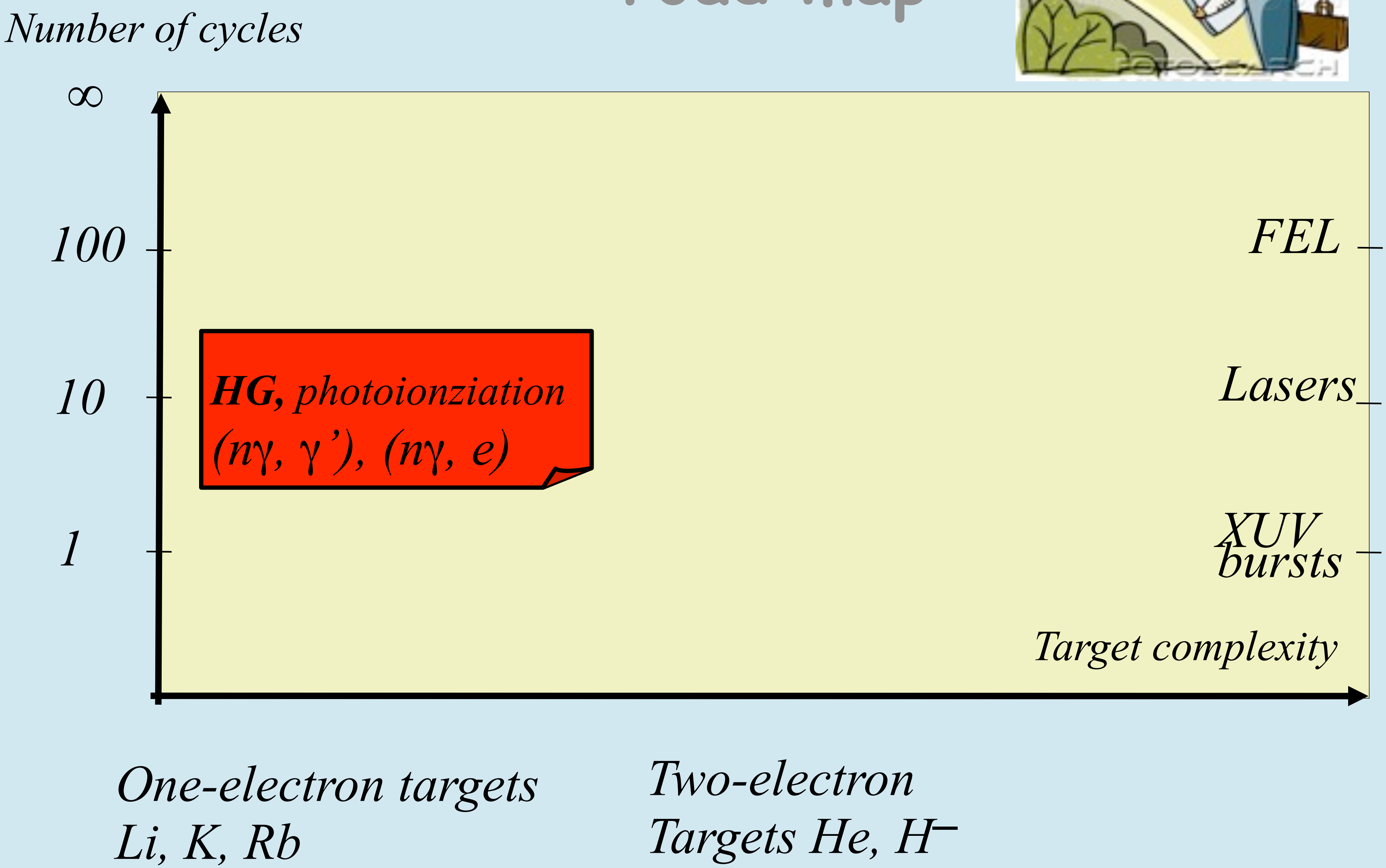

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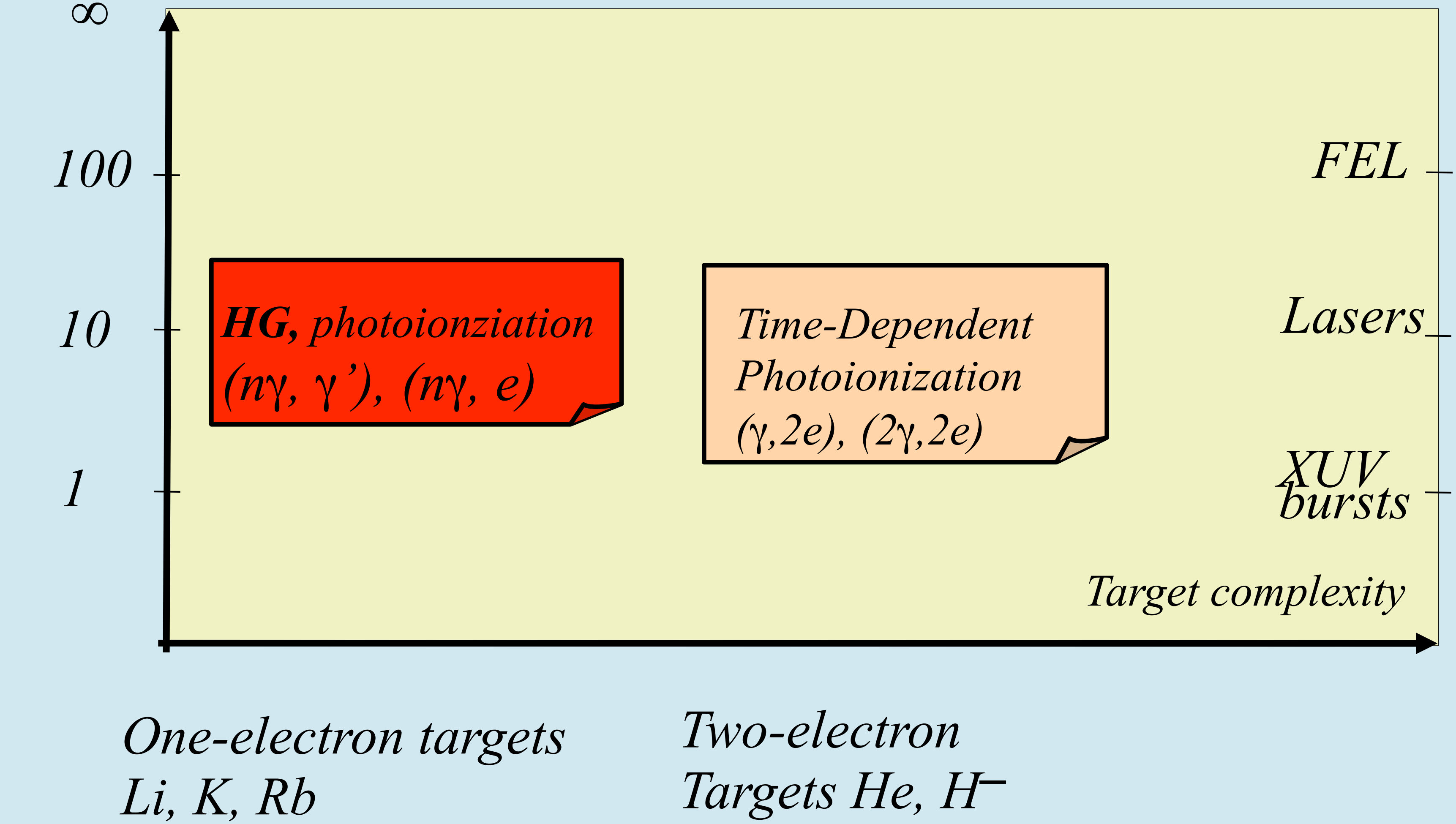

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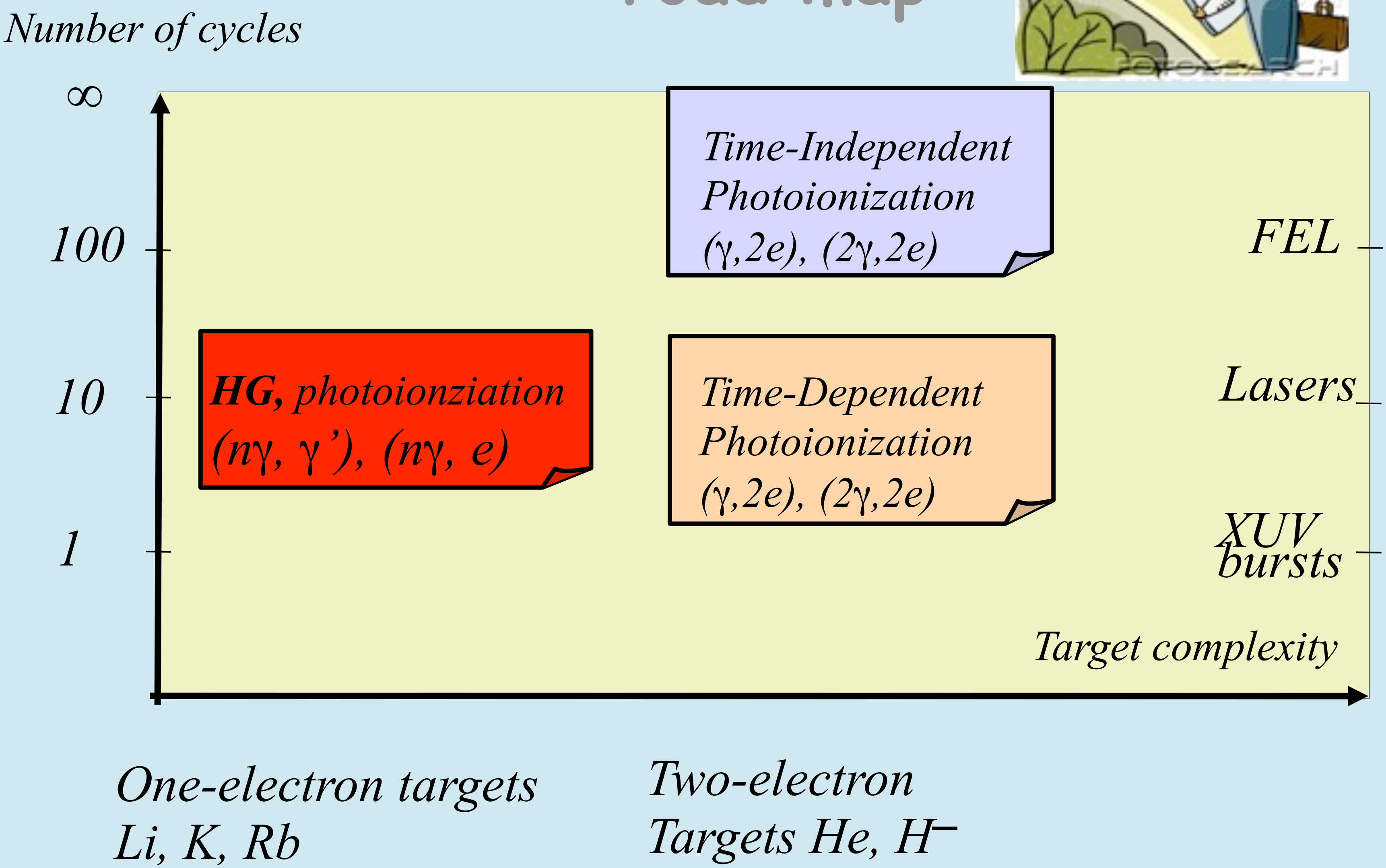

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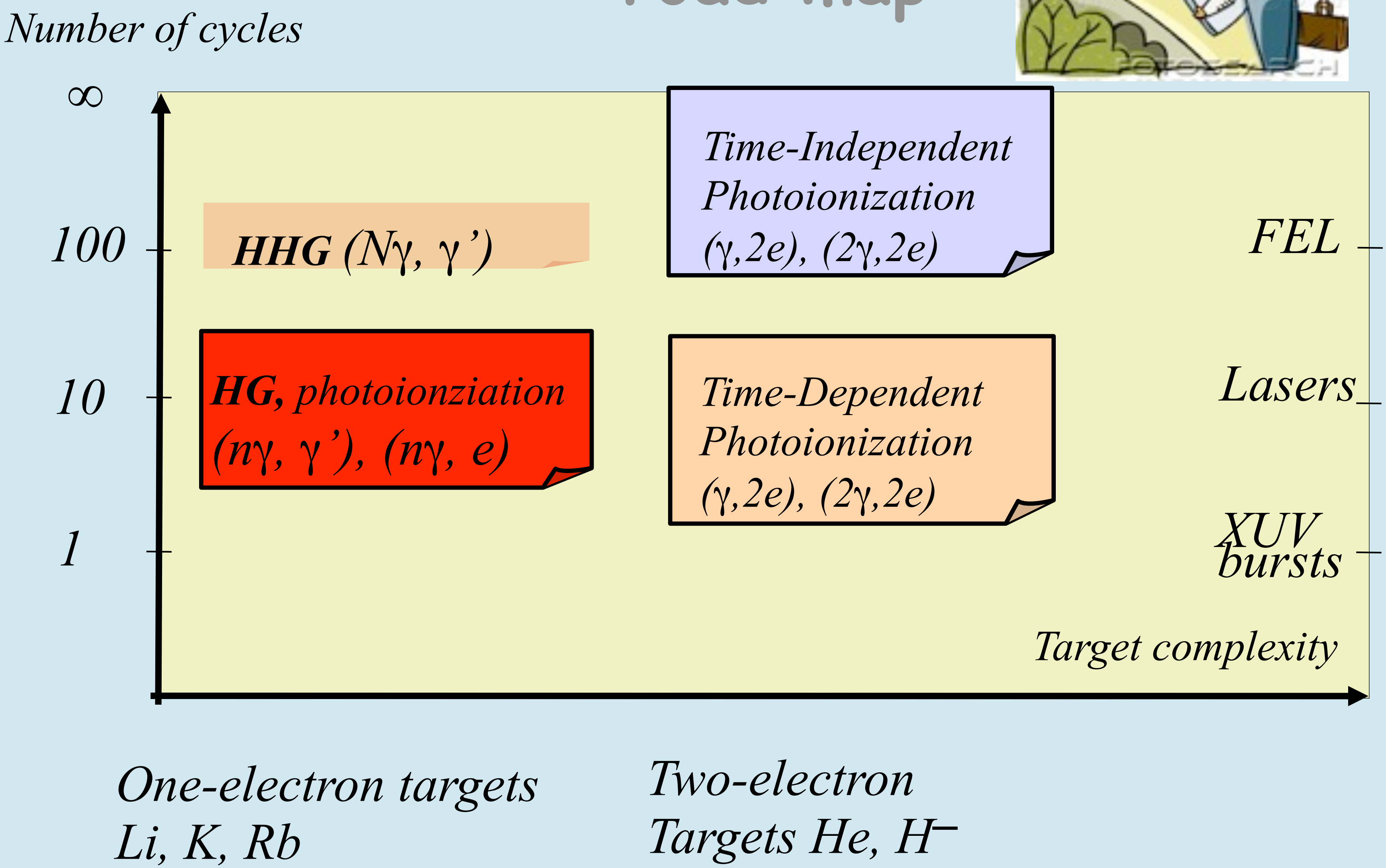

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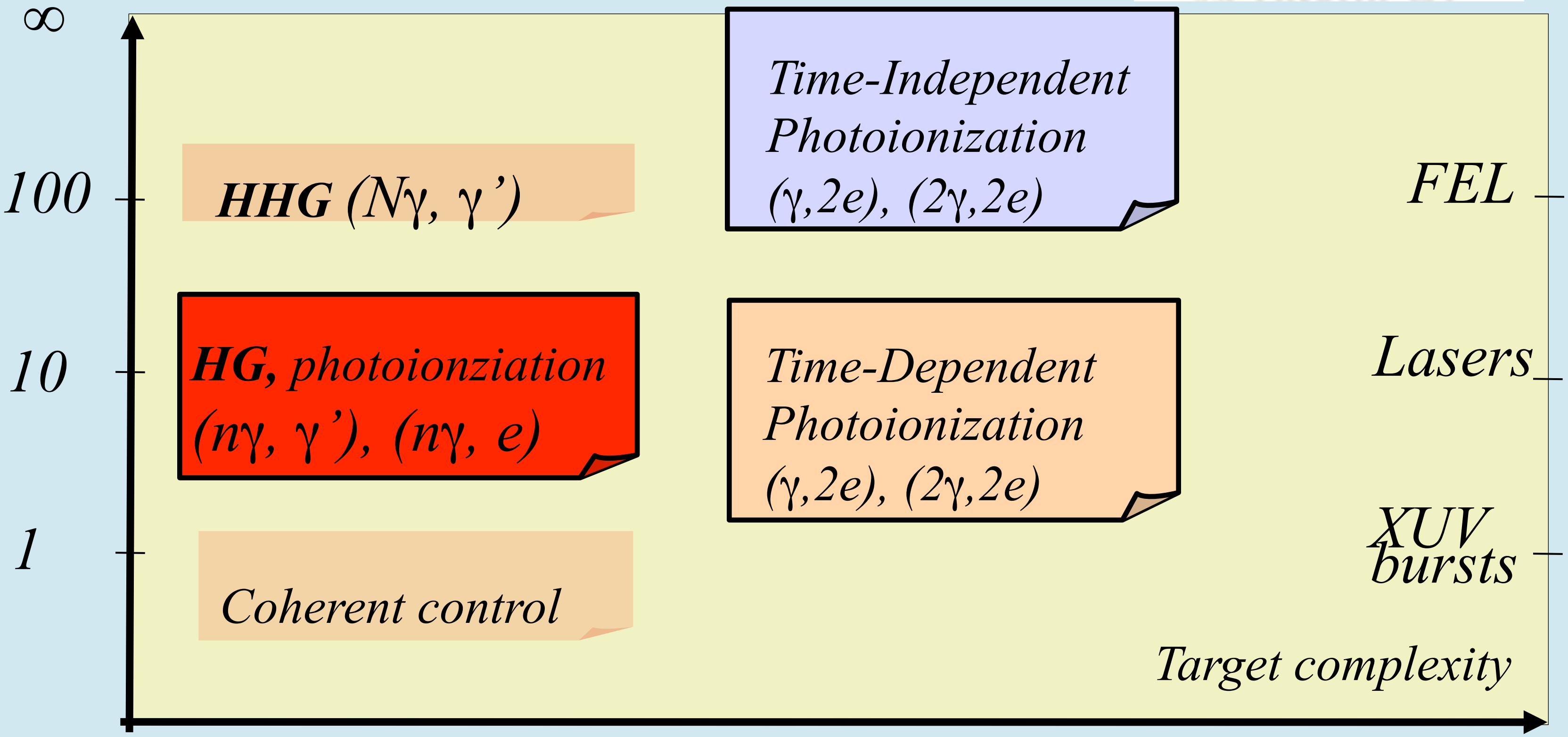

One-electron targets Li, K, Rb

Two-electron Targets He, H^-

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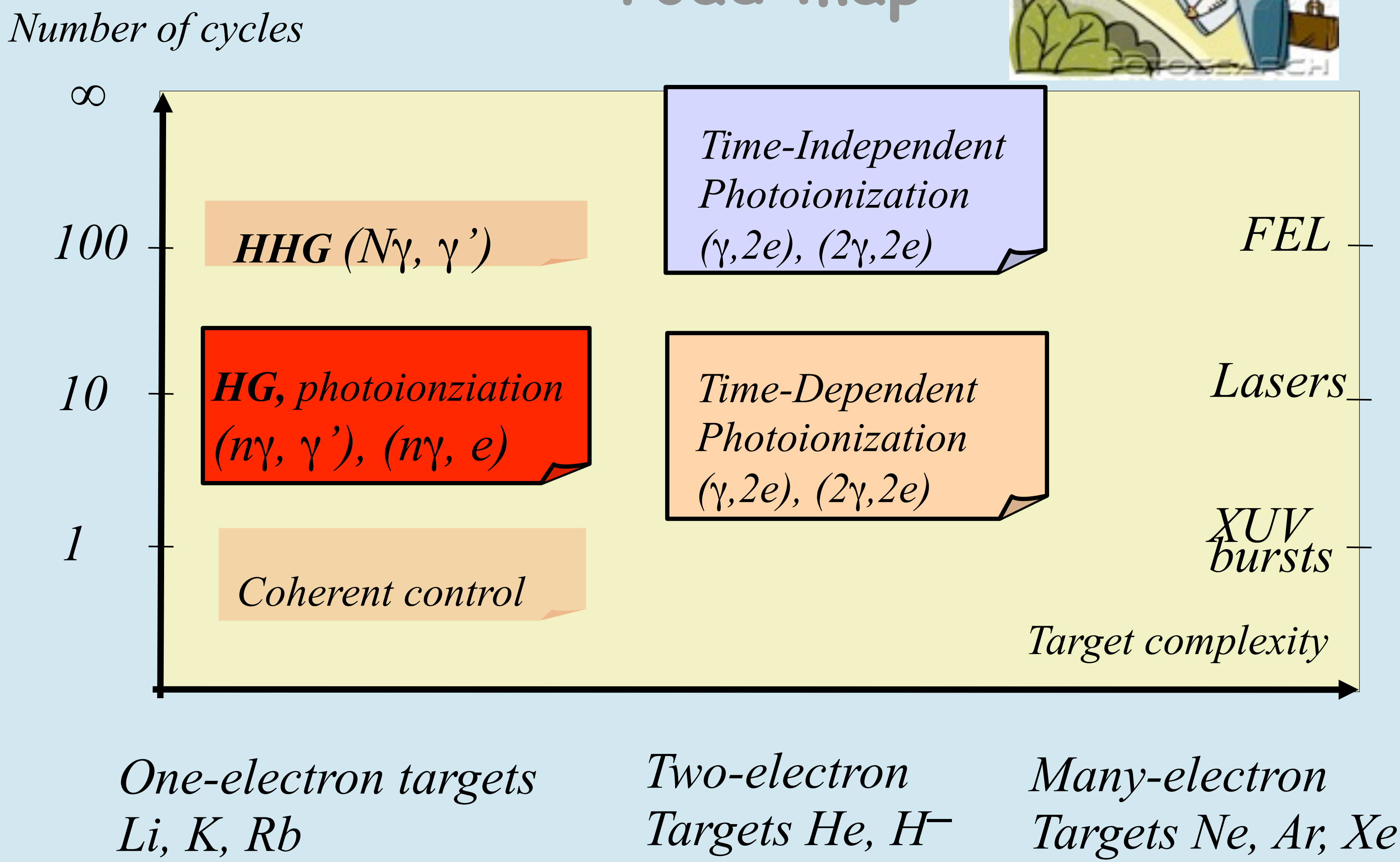

Outline

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- Theoretical model
  - Time-dependent Schrödinger equation

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    - *Pseudostates vs continuum states*

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    - *Optimal control*

Theoretical Model

Theoretical Model

Field on:

Theoretical Model

Field on:

Time-Dependent Schrödiger equation $i\frac{\partial \Psi}{\partial t} = \hat{H}\Psi$

$$\hat{H} = \frac{\boldsymbol{p}_1^2}{2} + \frac{\boldsymbol{p}_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|} + \boldsymbol{D}\cdot\boldsymbol{F}_{\mathrm{AC}} \cos \omega t$$

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Solution:

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = \sum_{j \equiv \left\{ \begin{smallmatrix} n_1 n_2 N \\ l_1 l_2 \ L \end{smallmatrix} \right\}} a_j(t)\, \phi^N_{n_1 l_1}(r_1) \phi^N_{n_2 l_2}(r_2)\, |l_1(1) l_2(2) L\rangle$$

Pseudostate basis: $\langle \phi_i^N | \hat{H} | \phi_j^N \rangle = E_i \delta_{ij}$

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Matrix notations:

$$i\boldsymbol{R}\cdot\dot{\boldsymbol{a}}(t) = \boldsymbol{H}\cdot\boldsymbol{a}(t)\,, \quad \text{with intial condition} \quad \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t = 0) = \Psi_0(\boldsymbol{r}_1, \boldsymbol{r}_2)$$

Theoretical Model

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Field off:

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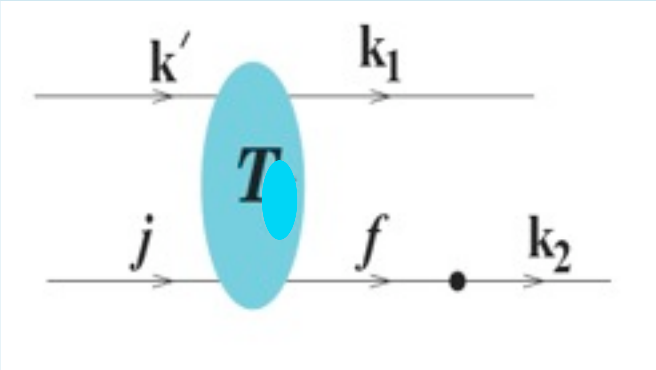
Field off:

CCC expansion for two-electron continuum



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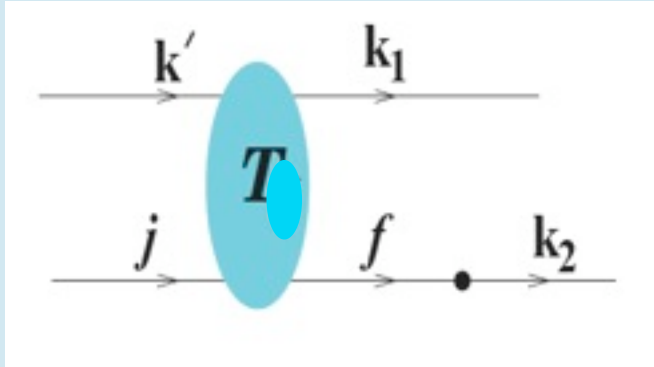

$$\Psi_{\boldsymbol{k}_1\boldsymbol{k}_2}(\boldsymbol{r}_1,\boldsymbol{r}_2)=\Psi_{\boldsymbol{k}_1 f}(\boldsymbol{r}_1,\boldsymbol{r}_2)\langle\boldsymbol{k}_2|f\rangle \quad , \quad \epsilon_f = k_2^2/2$$

$$\Psi_{\boldsymbol{k}f}(\boldsymbol{r}_1,\boldsymbol{r}_2)=\chi_{\boldsymbol{k}}(\boldsymbol{r}_1)\phi_f(\boldsymbol{r}_2)+\sum_{\boldsymbol{k}'j}\frac{\langle\boldsymbol{k}f|T_J|\boldsymbol{k}'j\rangle}{E-k'^2/2-\varepsilon_j+i0}\,\chi_{\boldsymbol{k}'}(\boldsymbol{r}_1)\phi_j(\boldsymbol{r}_2)$$

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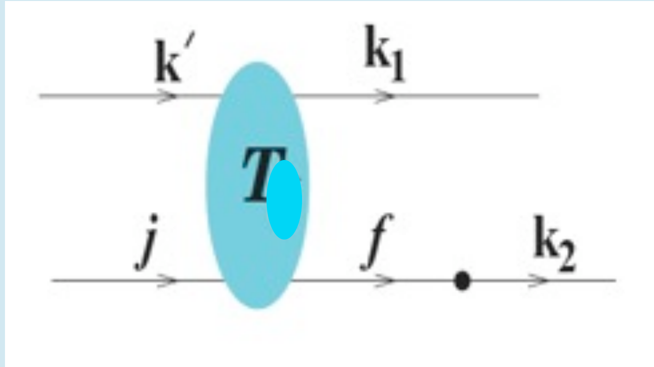

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Coulomb wave

Friday, May 15, 2009

Theoretical Model

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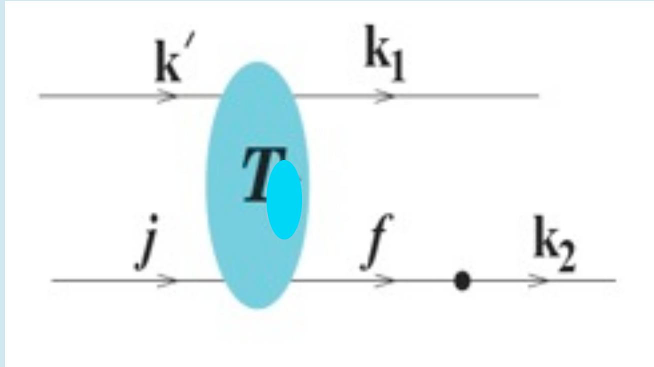

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Pseudostate

Friday, May 15, 2009

Theoretical Model

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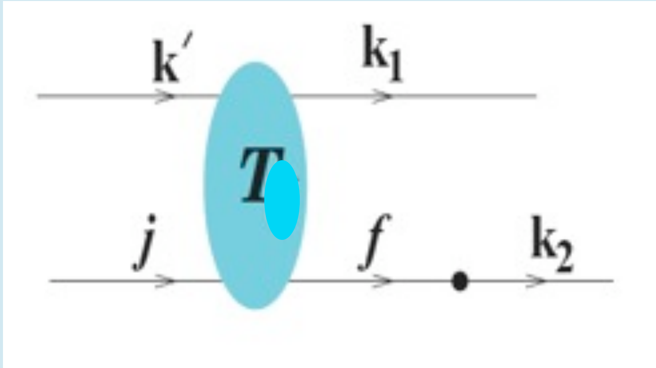

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Coulomb wave

Pseudostate

Fully differential cross-section

$$\frac{d\sigma}{d\boldsymbol{k}_1 d\boldsymbol{k}_2} = \left|\langle \Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t=T)|\Psi_{\boldsymbol{k}_1\boldsymbol{k}_2}(\boldsymbol{r}_1,\boldsymbol{r}_2)\rangle\right|^2 \quad T = N\frac{2\pi}{\omega} \quad , \quad N \gg 1$$

Theoretical Model

Theoretical Model

Harmonics Generation:

Theoretical Model

Harmonics Generation:

Harmonics spectrum

$$|d(\omega)|^2 = \left| \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} e^{-i\omega t} \, d(t) \, dt \right|^2$$

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Expectation value of dipole operator

$$\begin{aligned} d(t) &= \langle \Psi(t)|z|\Psi(t)\rangle = \int d\boldsymbol{r} \, dt \; \Psi(\boldsymbol{r},t) \; (\boldsymbol{r} \cdot \boldsymbol{e})\Psi(\boldsymbol{r},t) \\ &= \sum_{n,m} z_{mn} \exp[-i(m-n)\omega t] = \sum_{N} z_N \exp[-i(2M+1)\omega t] \end{aligned}$$

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Parity conservation $\Pi = -1 = (-1)^{2M+1}$

Integrated TPDI Cross-section of He

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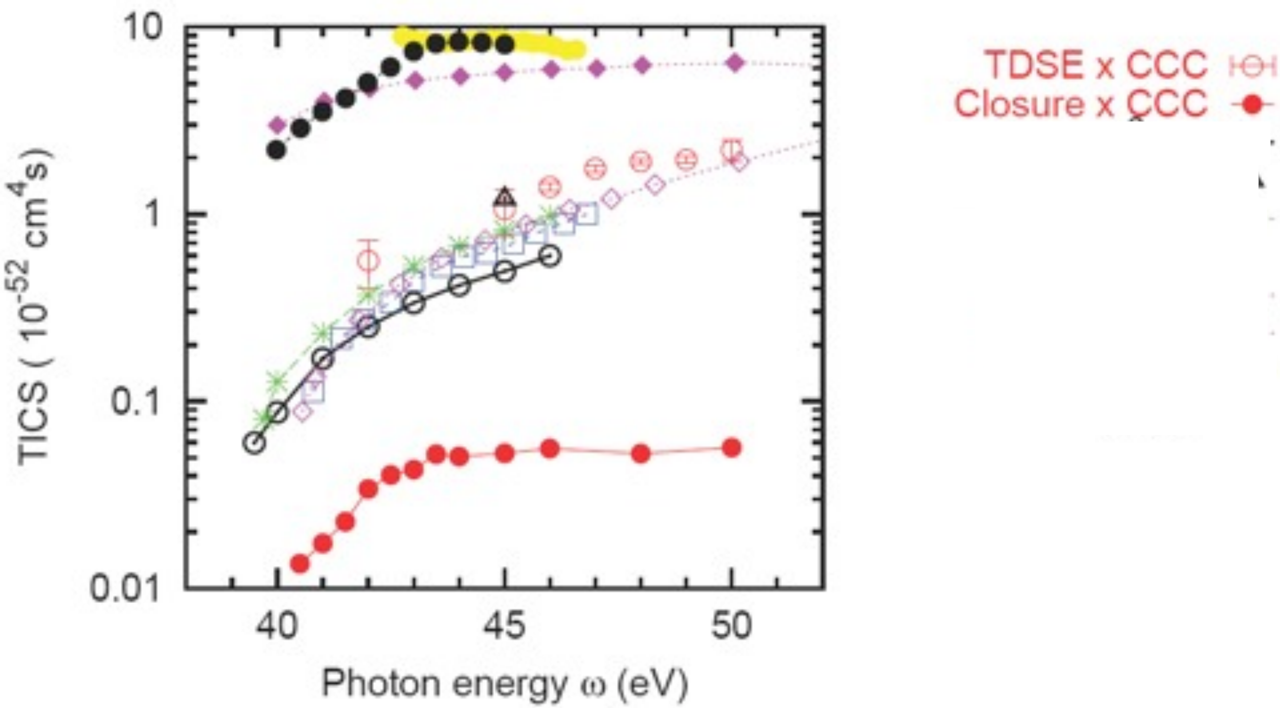

3.5 x 10^{14} W/cm^2

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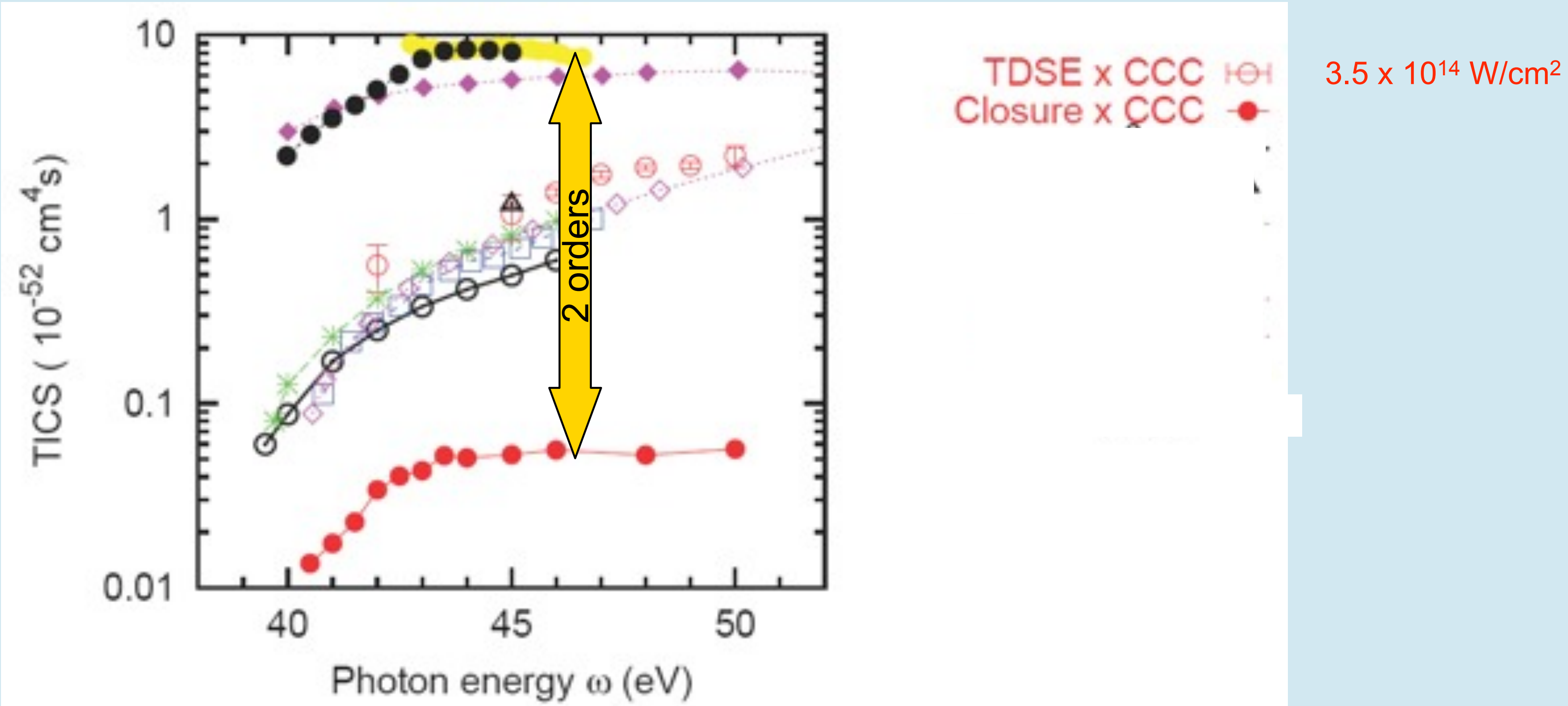

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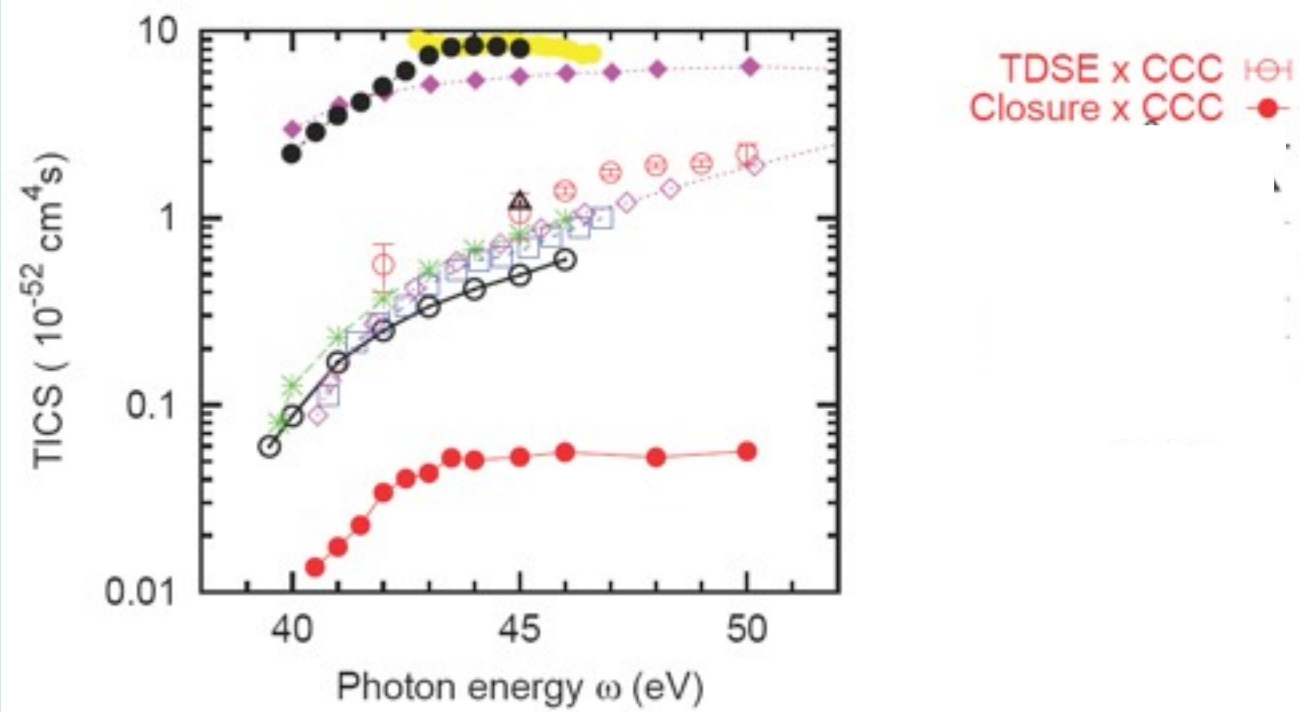

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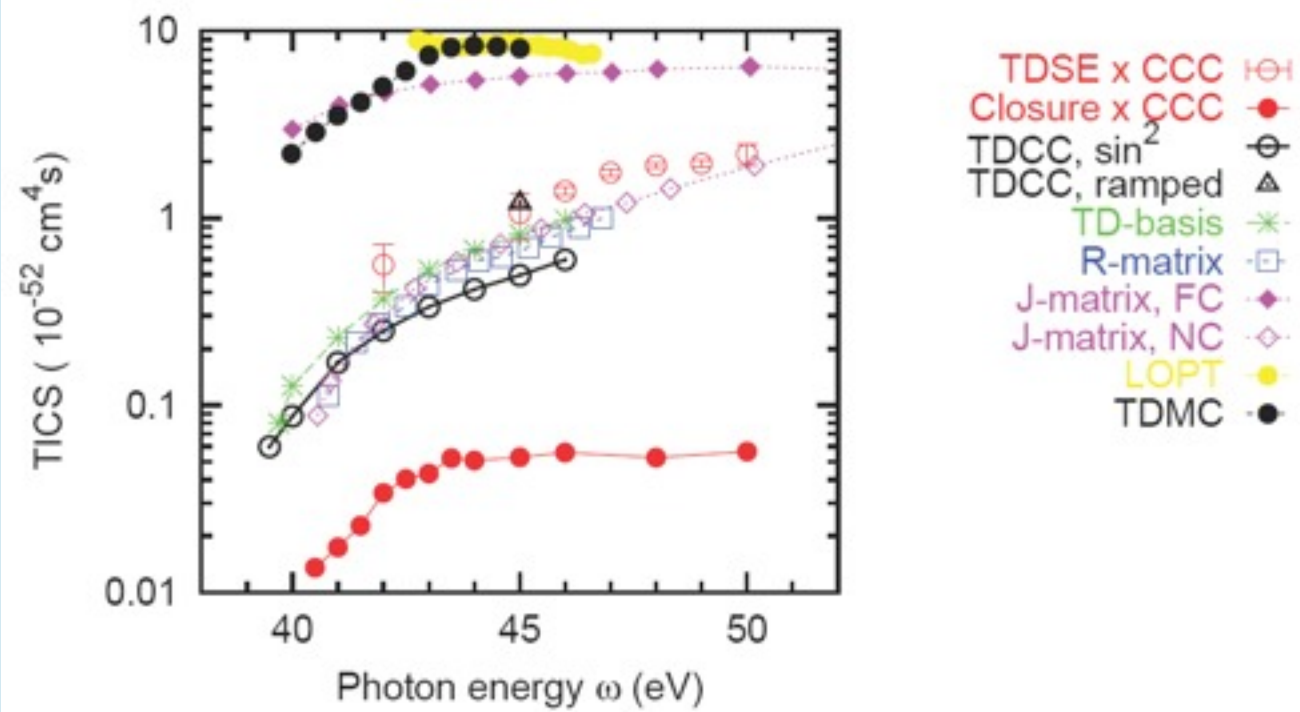

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TDCC $\sin^2$	Hu S X, Colgan J and Collins L A 2005 *J. Phys. B* **38**, L35
TDCC ramped	Colgan J and Pindzola L A 2002 *Phys. Rev. Lett.* **88**, 173002
TD-basis	Piraux B, Bauer J, Laulan S and Bachau H 2003 *Eur. Phys. J. D* **26**, 7
R-matrix	Feng L and van der Hart H W 2003 *J. Phys. B* **36**, L1
J-matrix	Foumouo et al. 2006 *Phys. Rev. A* **74**,063409
LOPT	Nikolopoulos and Lambropoulos 2001 *J. Phys. B* **34**,545
TDMC	Nikolopoulos and Lambropoulos 2007 *J. Phys. B* **40**,1347

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Theory vs. experiment:

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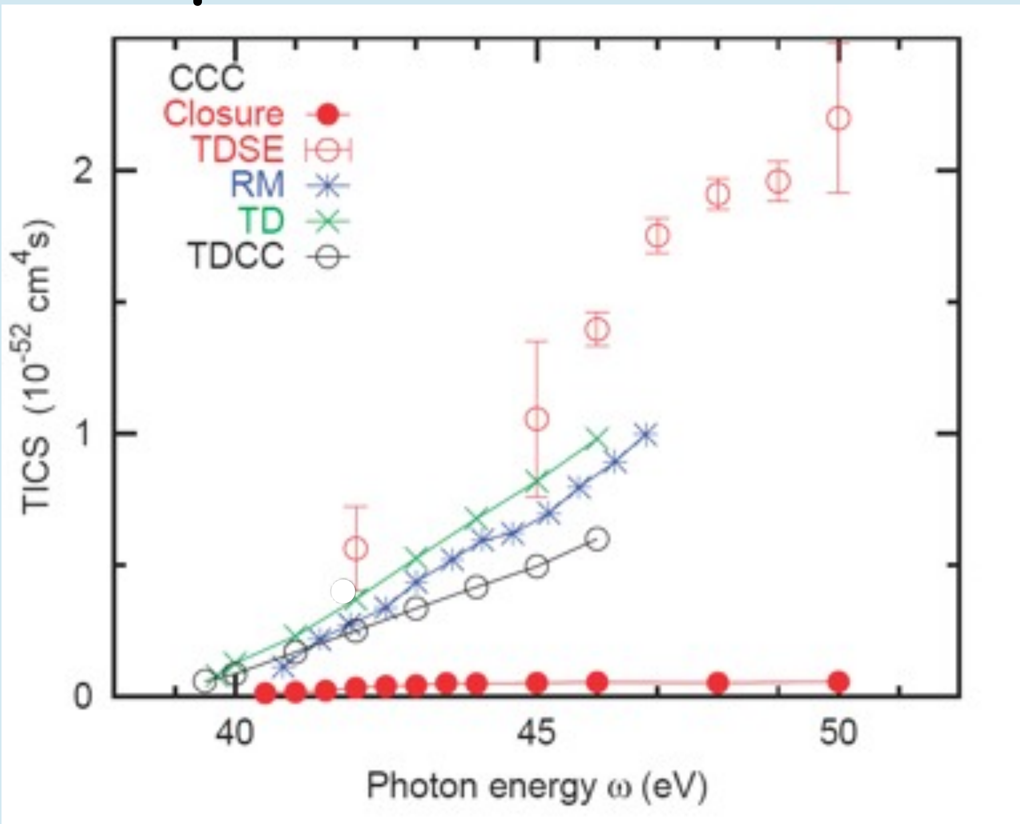

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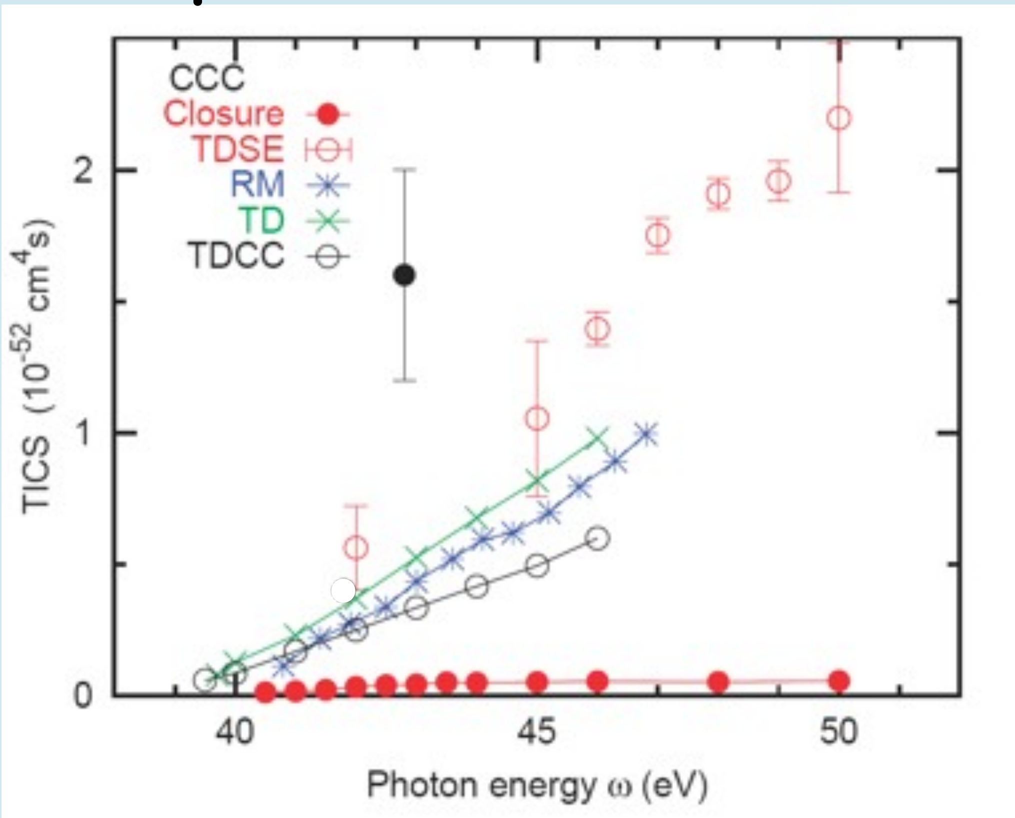

FEL @ 42.8 eV $1.6 \pm 0.6 \times 10^{-52}$ cm^{4}s Sorokin et al. 2007 PRA **75**, 051402

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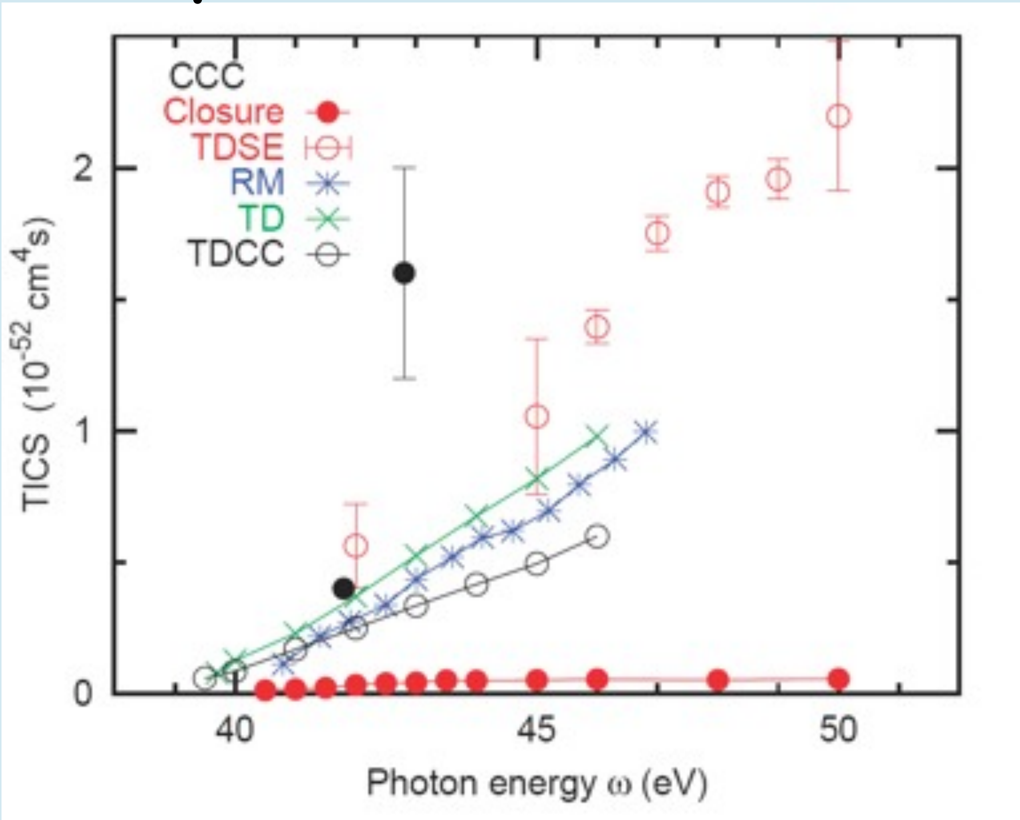

FEL @ 42.8 eV	$1.6\pm0.6 \times 10^{-52}$ cm^{4}s	Sorokin et al. 2007 PRA **75**, 051402
HHG @ 41.8 eV	0.4×10^{-52} cm^{4}s	Hasegawa et al. 2005 PRA **71**, 023407

Lithium atom in intense laser field

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Michael Schurike, Max-Planck-Institut für Kernphysik, Heidelberg, ICPEAC 2009

Lithium atom in intense laser field

	1st IP [eV]	I_{OBI} [W/cm^2]	γ	U_p [eV]
Li(2s)	5.39	$3.38 \cdot 10^{12}$	5.29	0.20
Li(2p)	3.54	$6.28 \cdot 10^{11}$	9.76	0.037
He	24.59	$1.46 \cdot 10^{15}$	0.53	86.1
Ne	21.57	$8.65 \cdot 10^{14}$	0.65	50.5
Ar	15.76	$2.47 \cdot 10^{14}$	1.0	14.6
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Low over-the-barrier intensities as compared to noble gases, absence of ponderomotive effects.

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High Keldysh-parameter γ, i.e. deep within multiphoton regime, but not perturbative.

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Ne	21.57	$8.65 \cdot 10^{14}$	0.65	50.5
Ar	15.76	$2.47 \cdot 10^{14}$	1.0	14.6
Xe	12.13	$8.66 \cdot 10^{13}$	1.5	5.11

Low over-the-barrier intensities as compared to noble gases, absence of ponderomotive effects.

High Keldysh-parameter γ, i.e. deep within multiphoton regime, but not perturbative.

Only 4 (2s) / 3 (2p) Ti:Sa-Photons required to ionize. Low non-linearity ($\sigma \propto I^N$).

Michael Schurike, Max-Planck-Institut für Kernphysik, Heidelberg, ICPEAC 2009

Photoelectron momentum distribution

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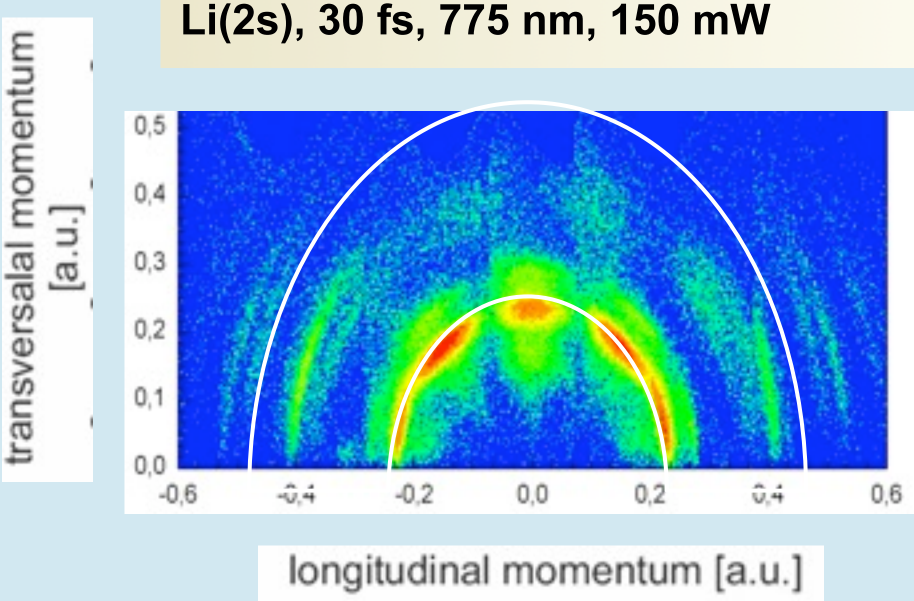
Experiment:
Li(2s), 30 fs, 775 nm, 150 mW



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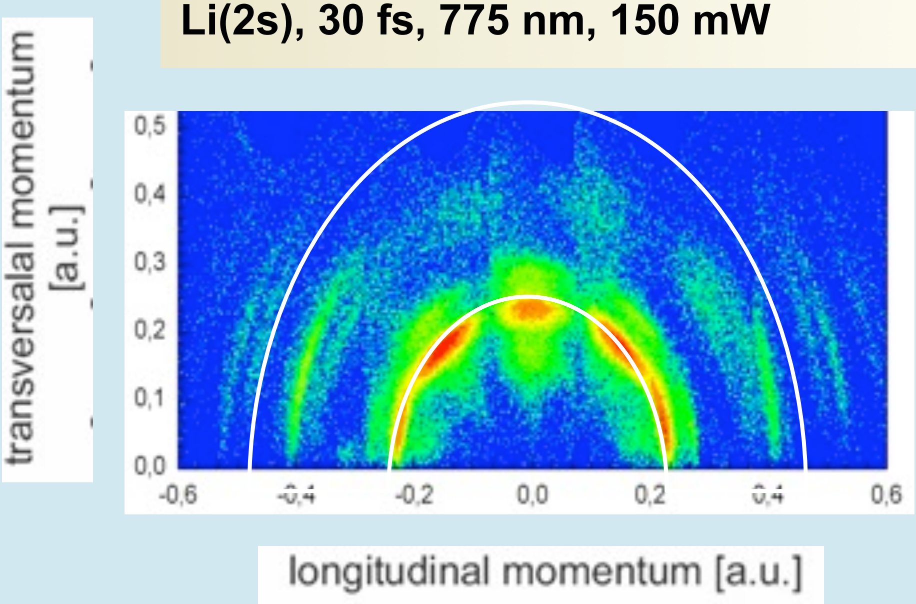

Momentum space multi-photon rings:

$$P_x^2+P_z^2=N\omega - IP,\ N\geq 4 \text{ for Li 2s}$$

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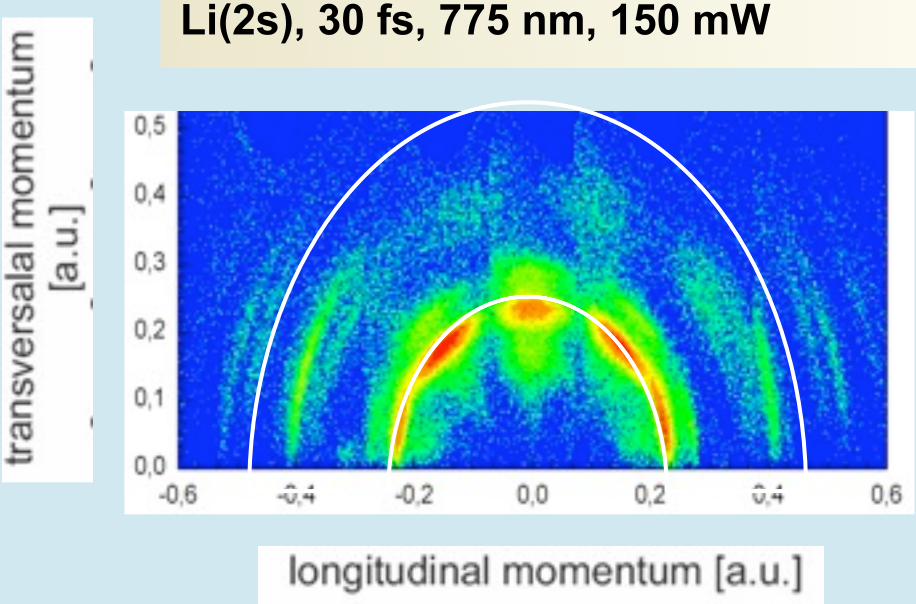
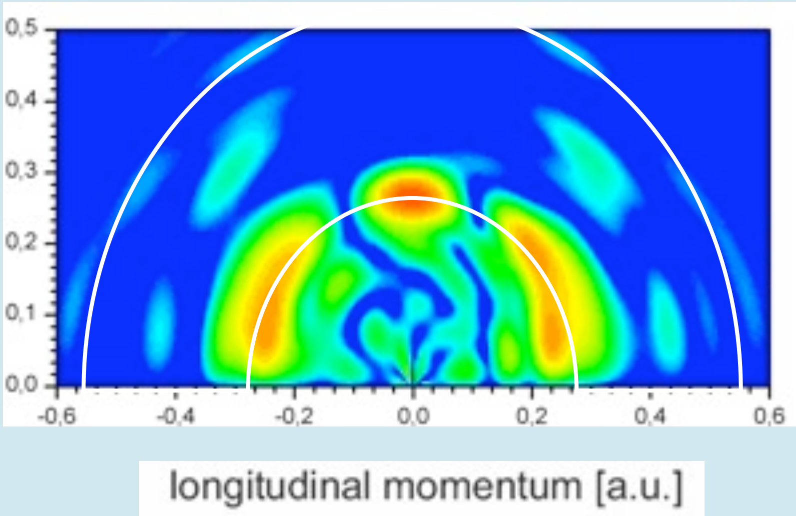
Li(2s), 30 fs, 775 nm, 150 mW

Calculation:
Li(2s), 25 fs, 800 nm, 3×10^{11} W/cm^2





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Resonant Enhancement of Harmonics Generation

KITP 2009

PRL 94, 113906 (2005) PHYSICAL REVIEW LETTERS week ending 25 MARCH 2005

Enhanced High Harmonic Generation from an Optically Prepared Excited Medium

KITP 2009

PRL **94,** 113906 (2005) PHYSICAL REVIEW LETTERS week ending 25 MARCH 2005

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We investigate high harmonics generated from rubidium atoms irradiated simultaneously by an intense 3.5 μm fundamental field and a weak cw diode laser. When $5p$, $5d$, and $4d$ excited states are populated through cascade excitation or deexcitation, orders-of-magnitude increases in harmonic yield as compared with the ground state are observed.

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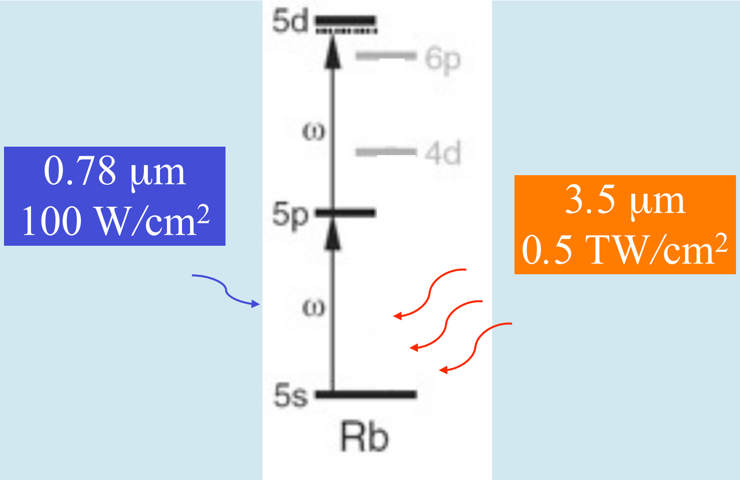

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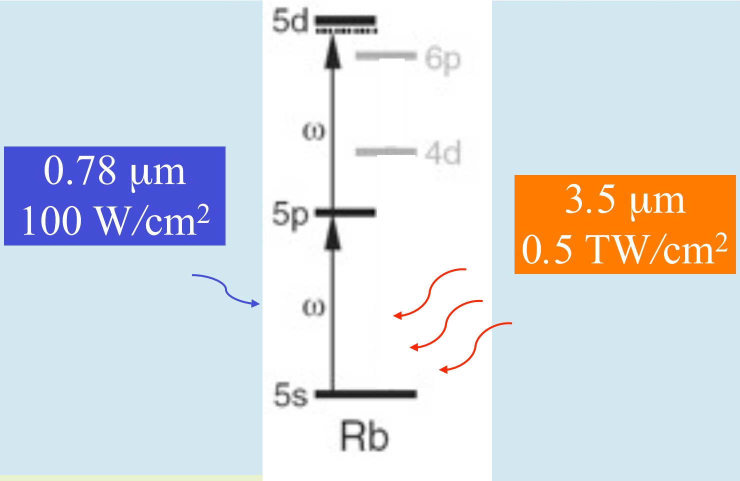

Scaled approach to HHG:

$$N_{cut\text{-}off}=(Ip+3.17Up)/\omega \gg 1$$

$$\gamma = (Ip/Up)^{1/2} \approx 1$$

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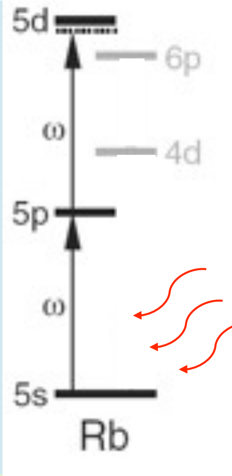
0.78 μm
100 W/cm^2



3.5 μm
0.5 TW/cm^2

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From noble gases in NIR

To alkaline metals in MIR

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Harmonic spectrum of Li @ 330 fs, 3500 nm

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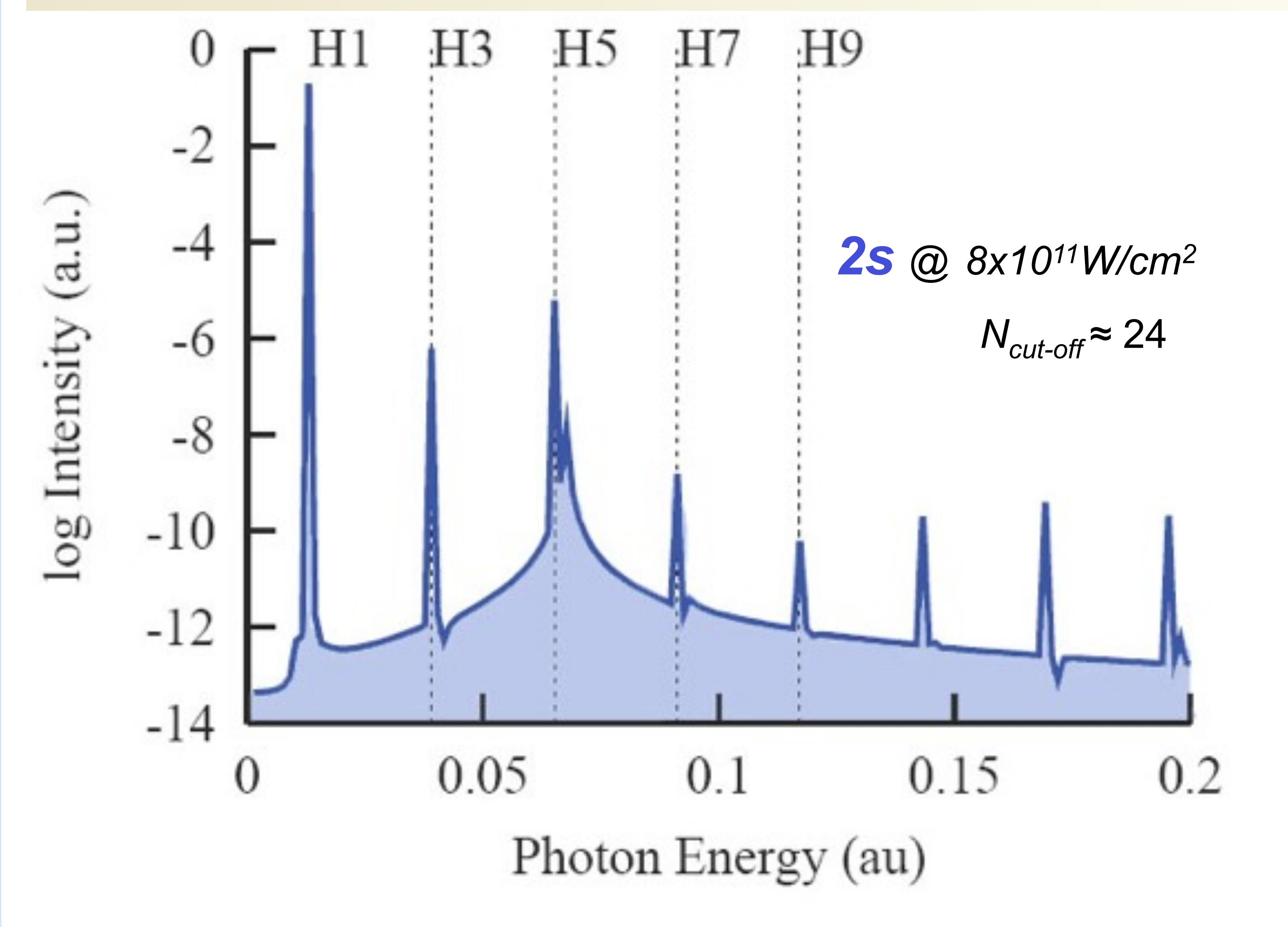

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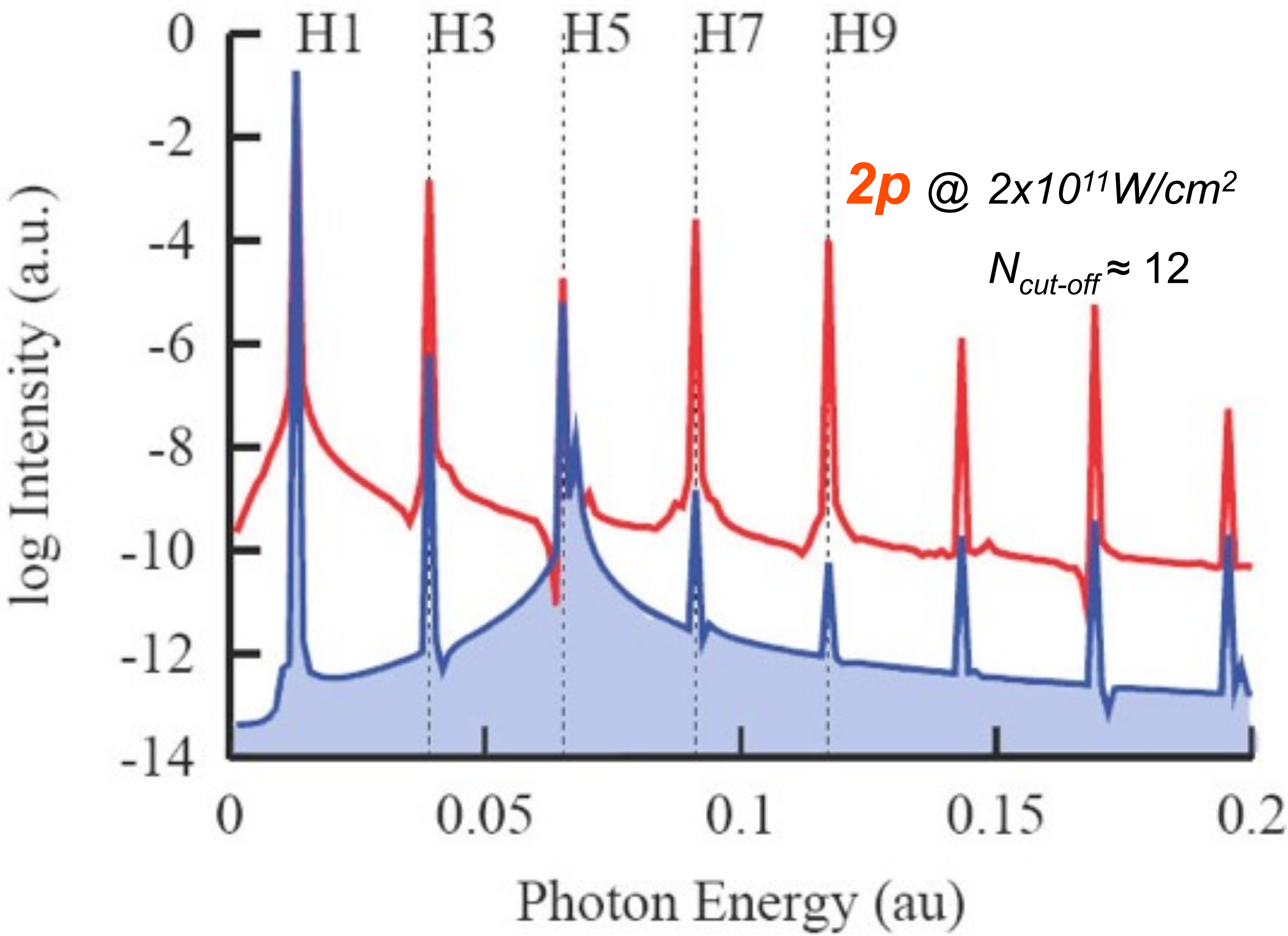

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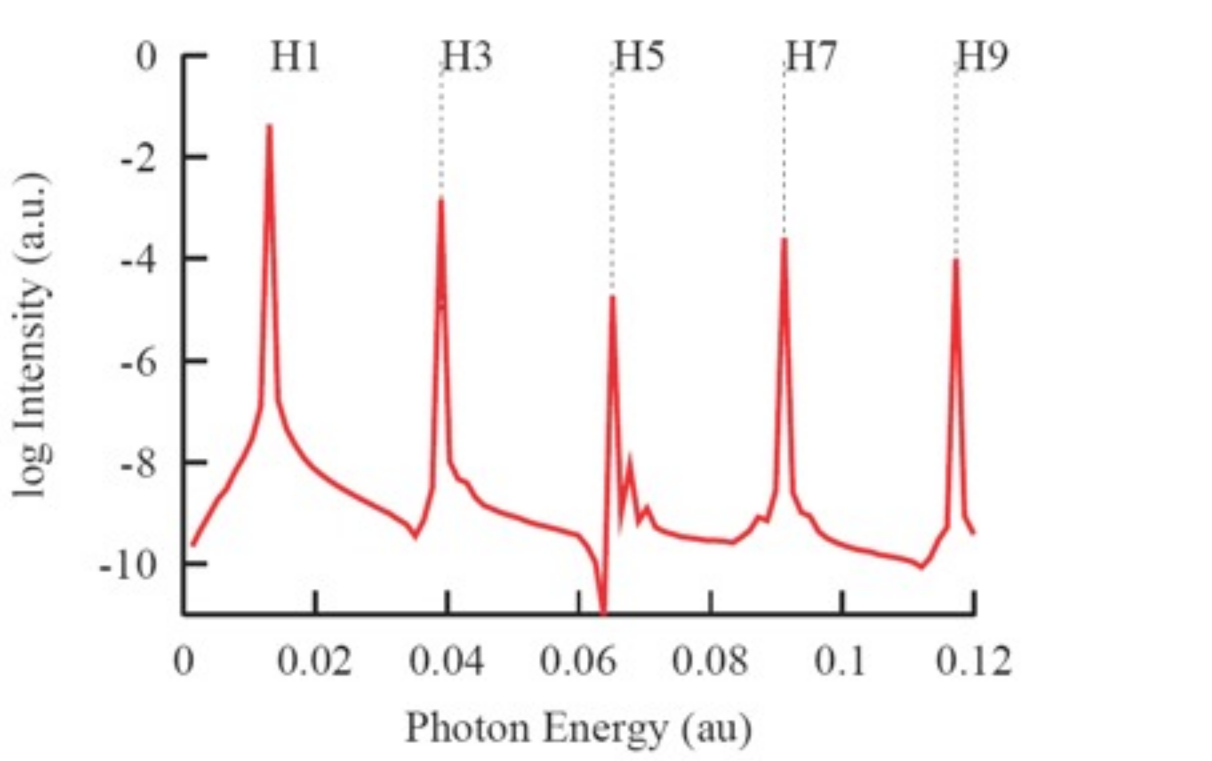

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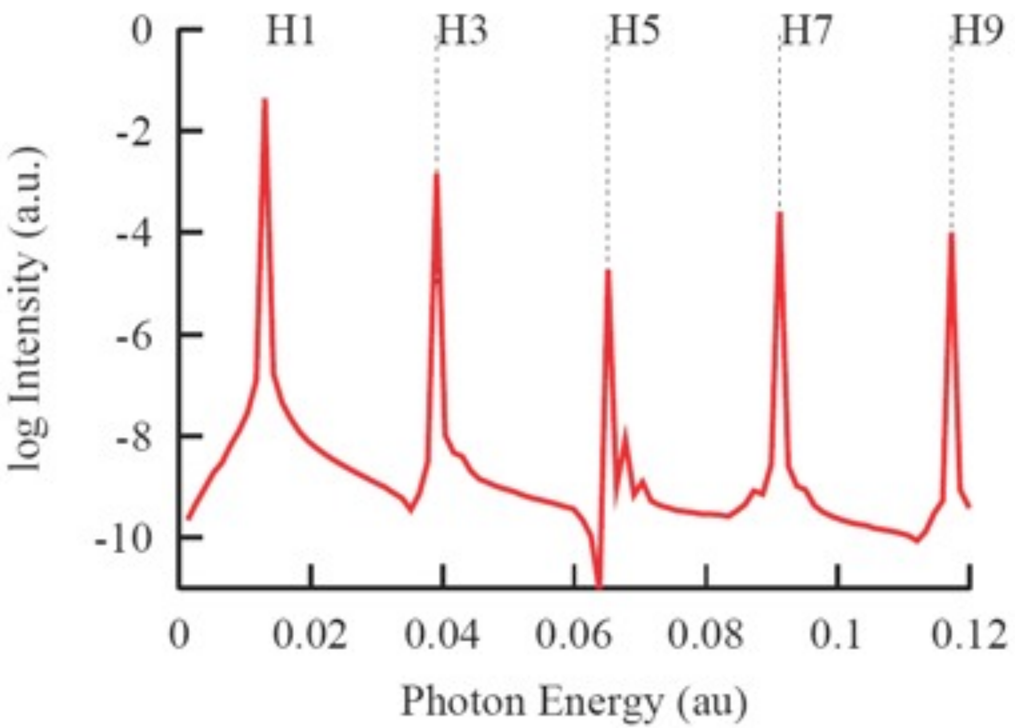

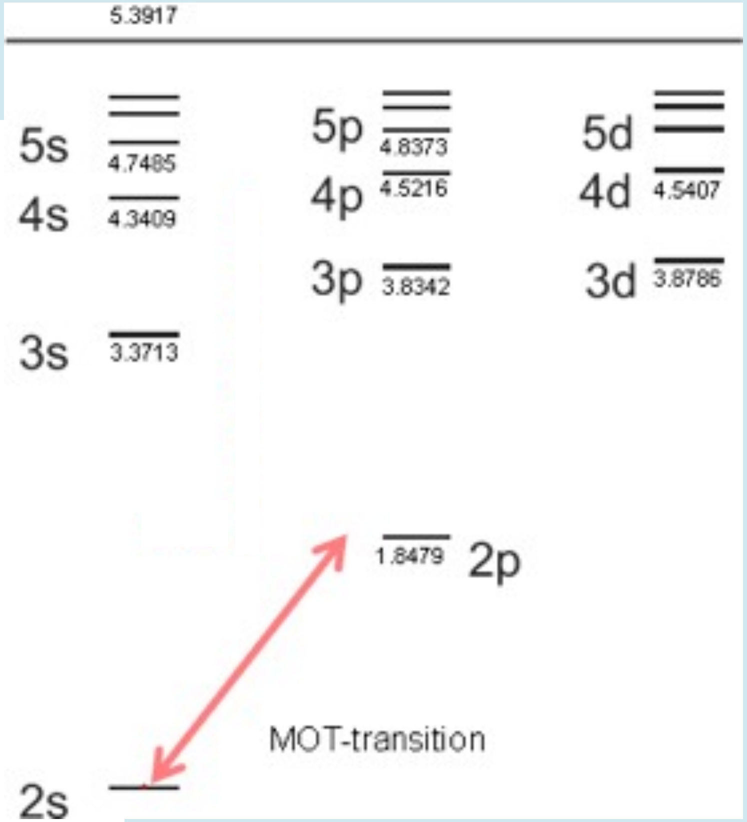

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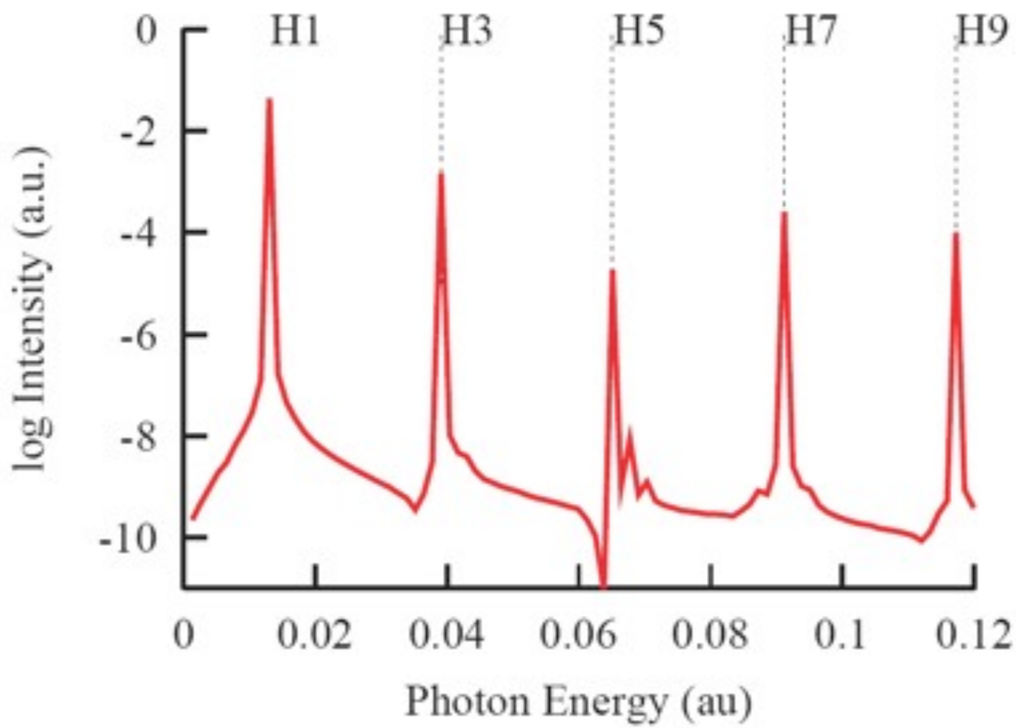

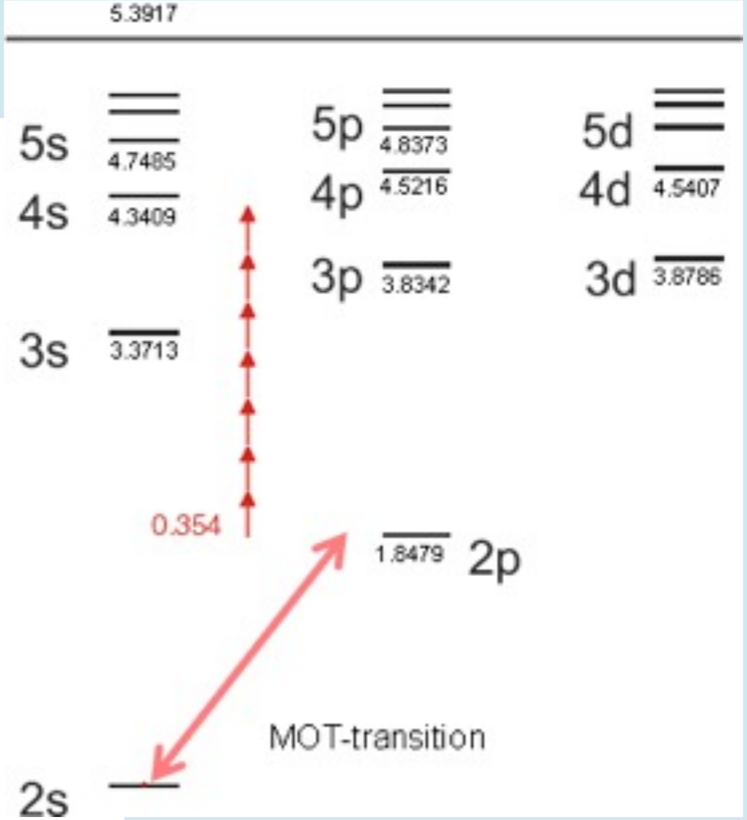

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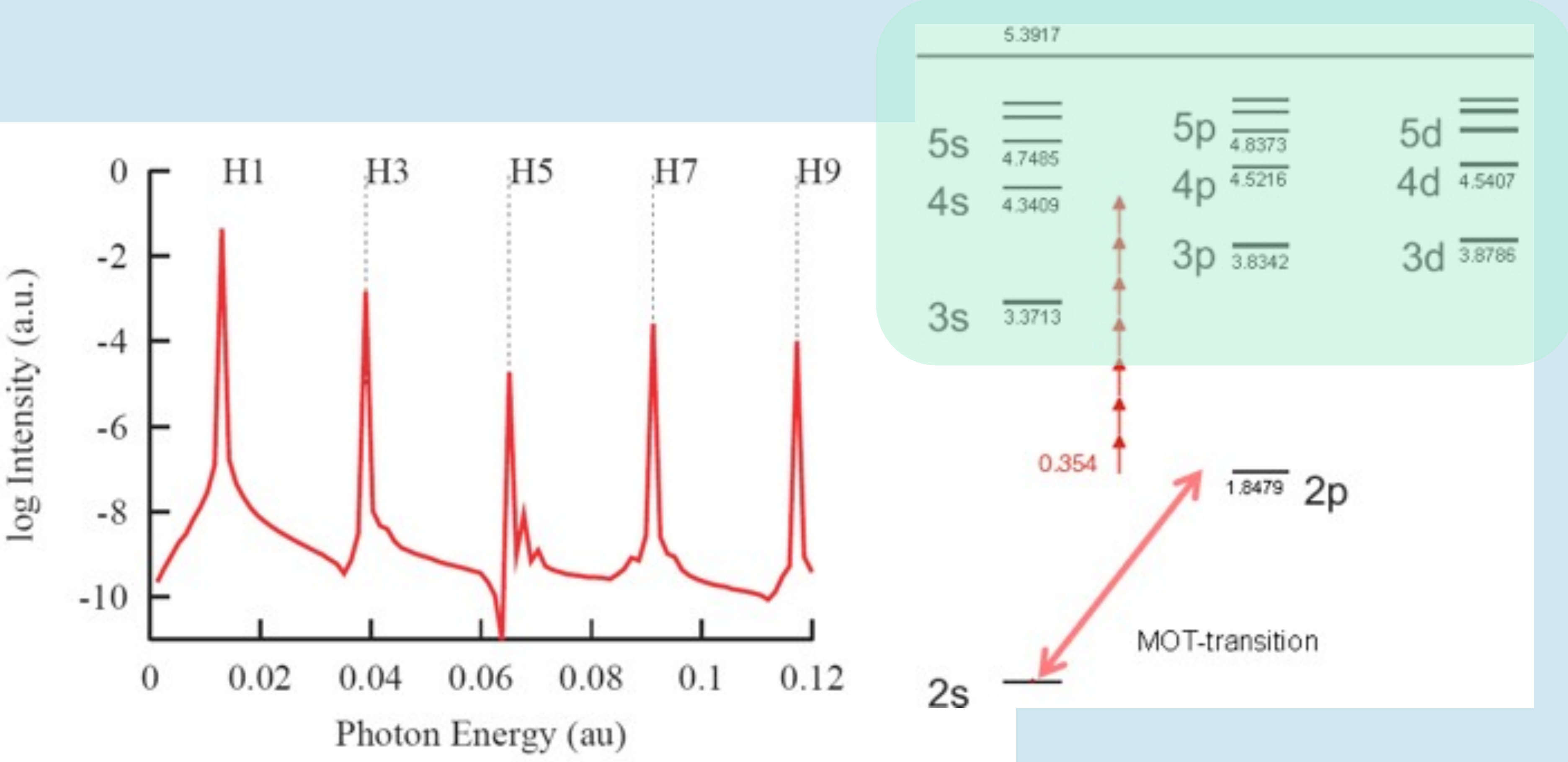

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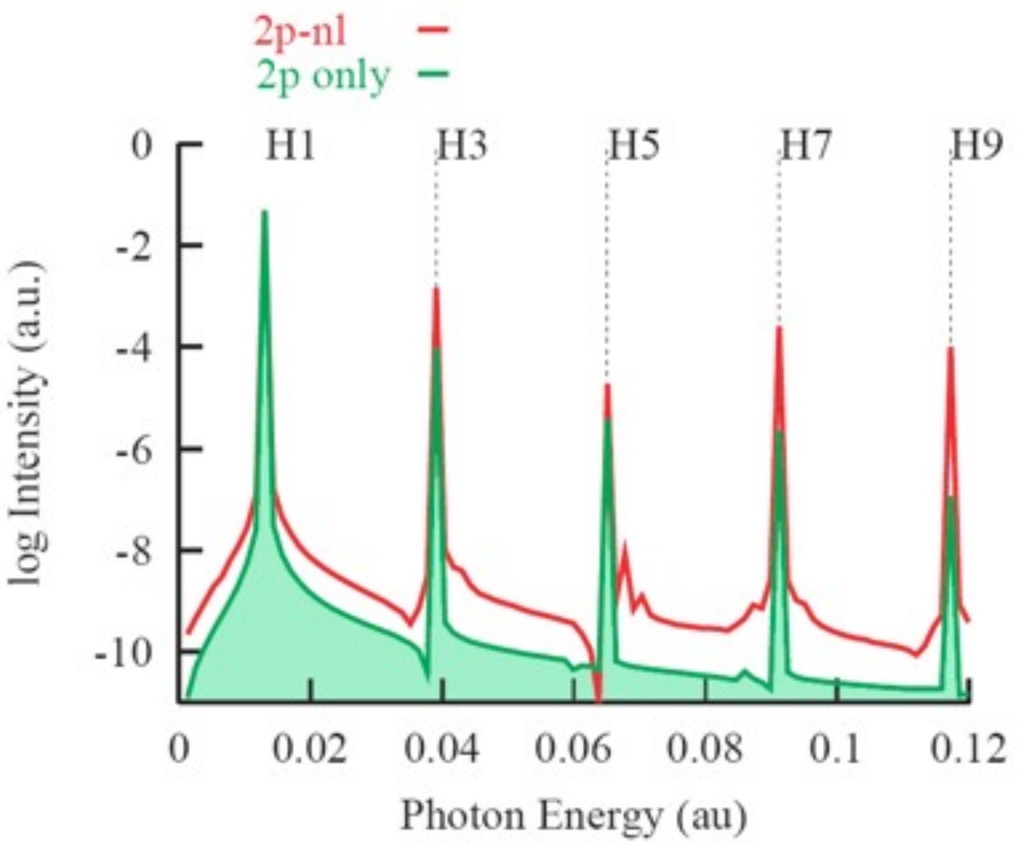

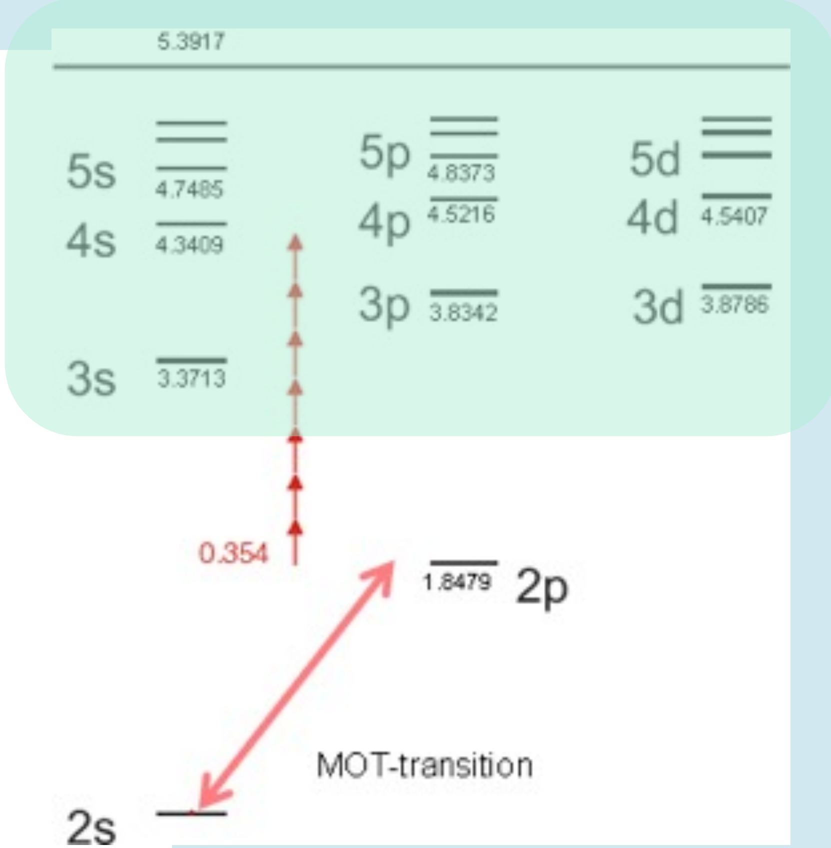

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Harmonics spectrum: effect of polarization

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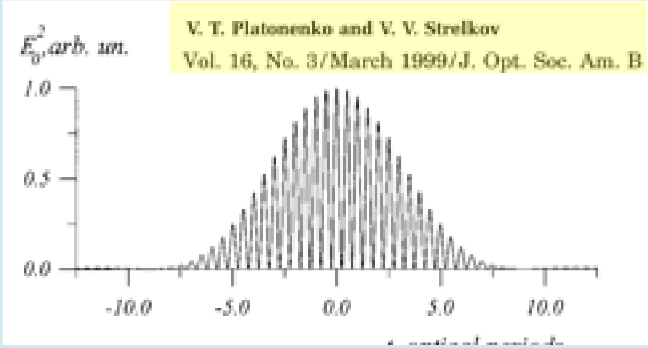

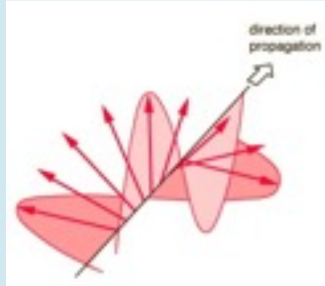

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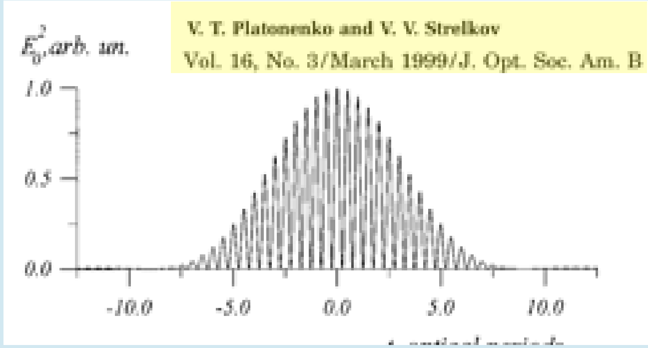

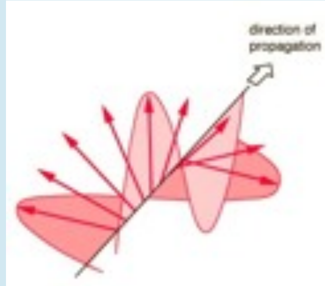

$$\hat{H}_{\rm int}(t) = E_0\Big[(x f_1(t)\cos\omega t + y f_2(t)\sin\omega t\Big]$$

$$f_1(t) = f(t) + f(t-\Delta)),\ f_2(t) = f(t) - f(t-\Delta)).$$

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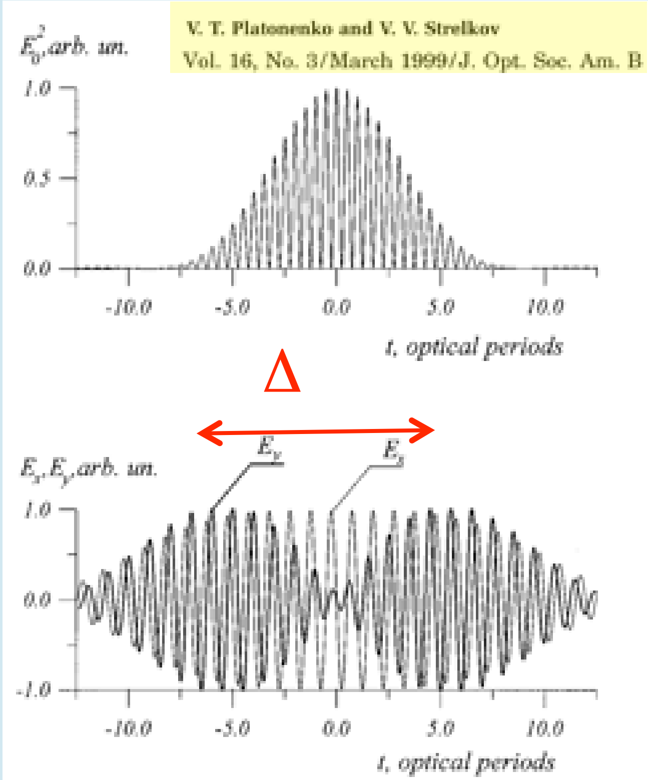

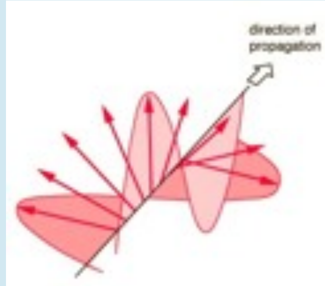

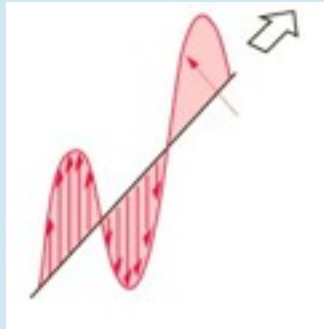

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Polarization gating: Morlet wavelet analysis

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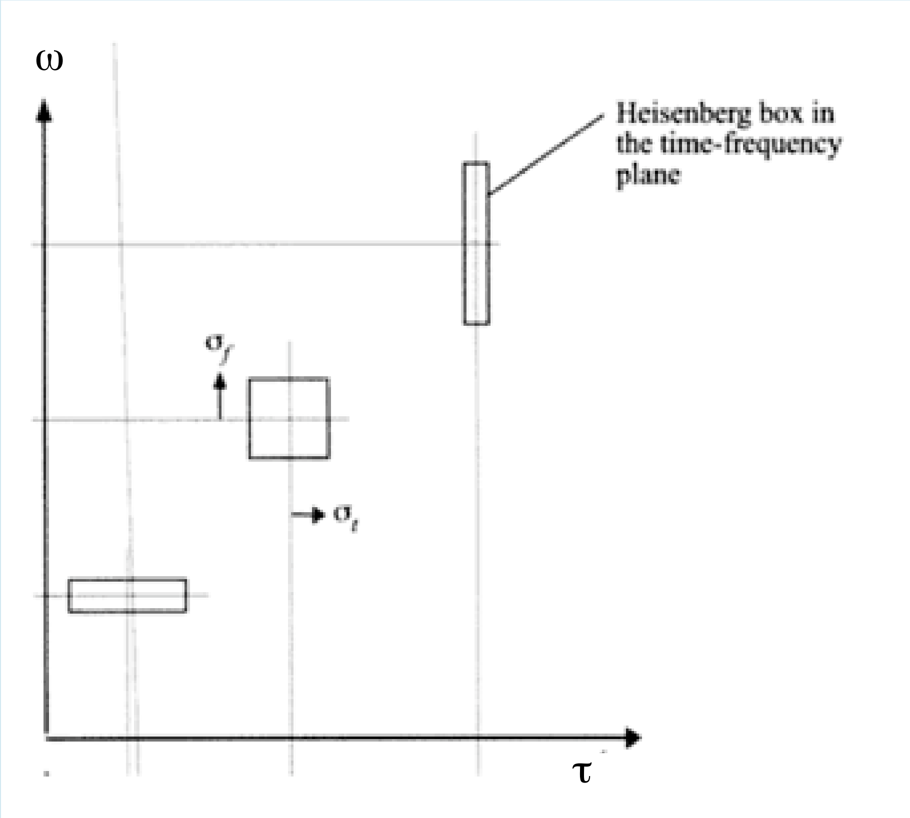

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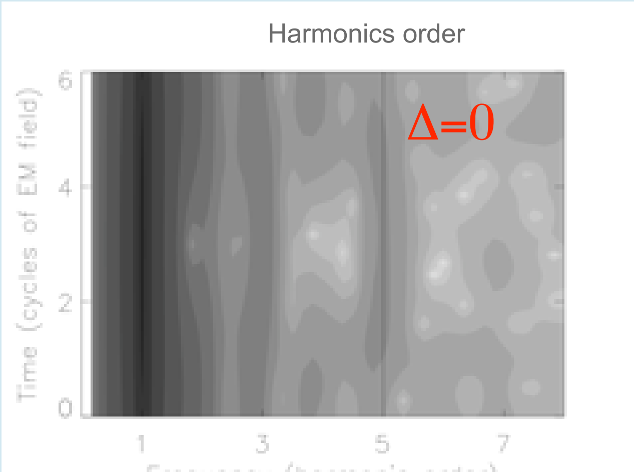

Rb $F = 0.002$ a.u. $\omega = 0.2$ eV

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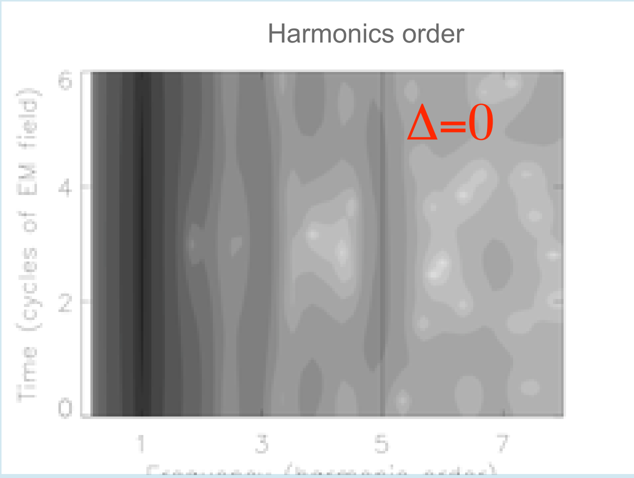

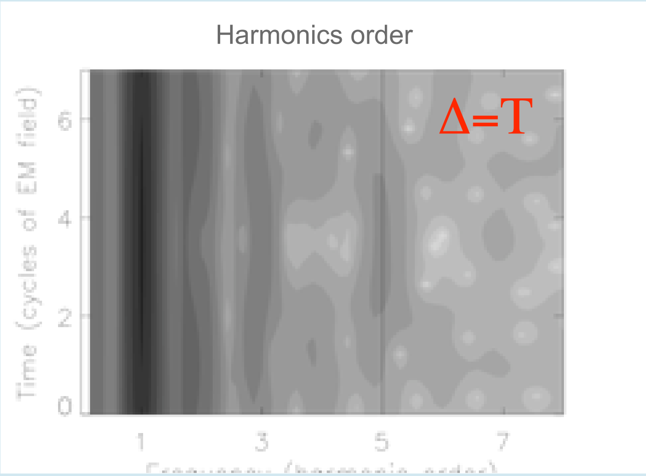

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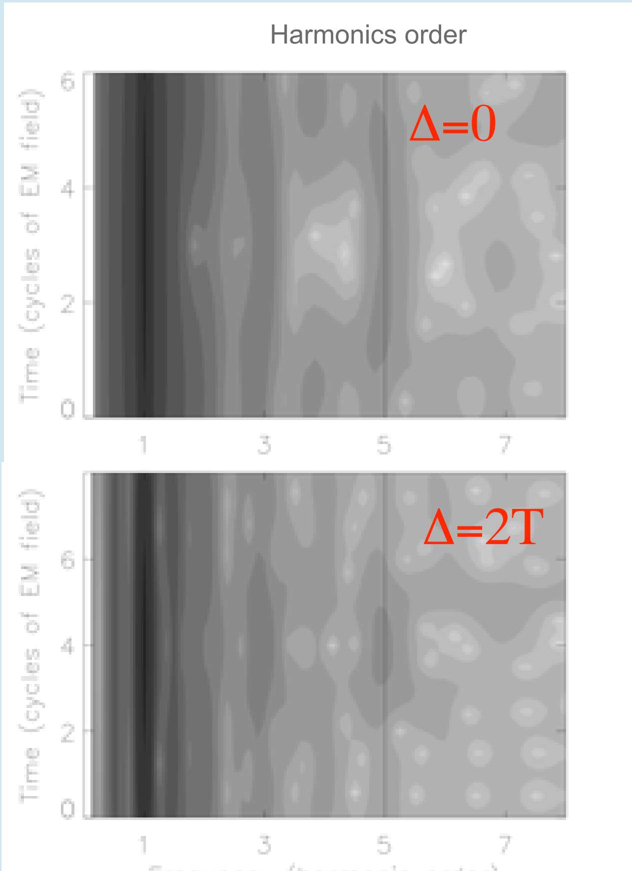

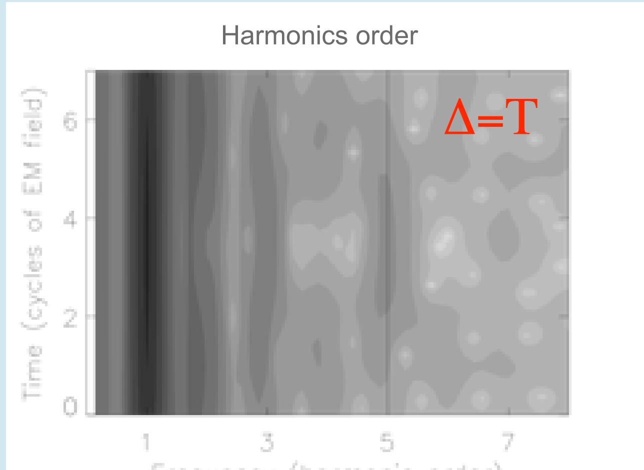

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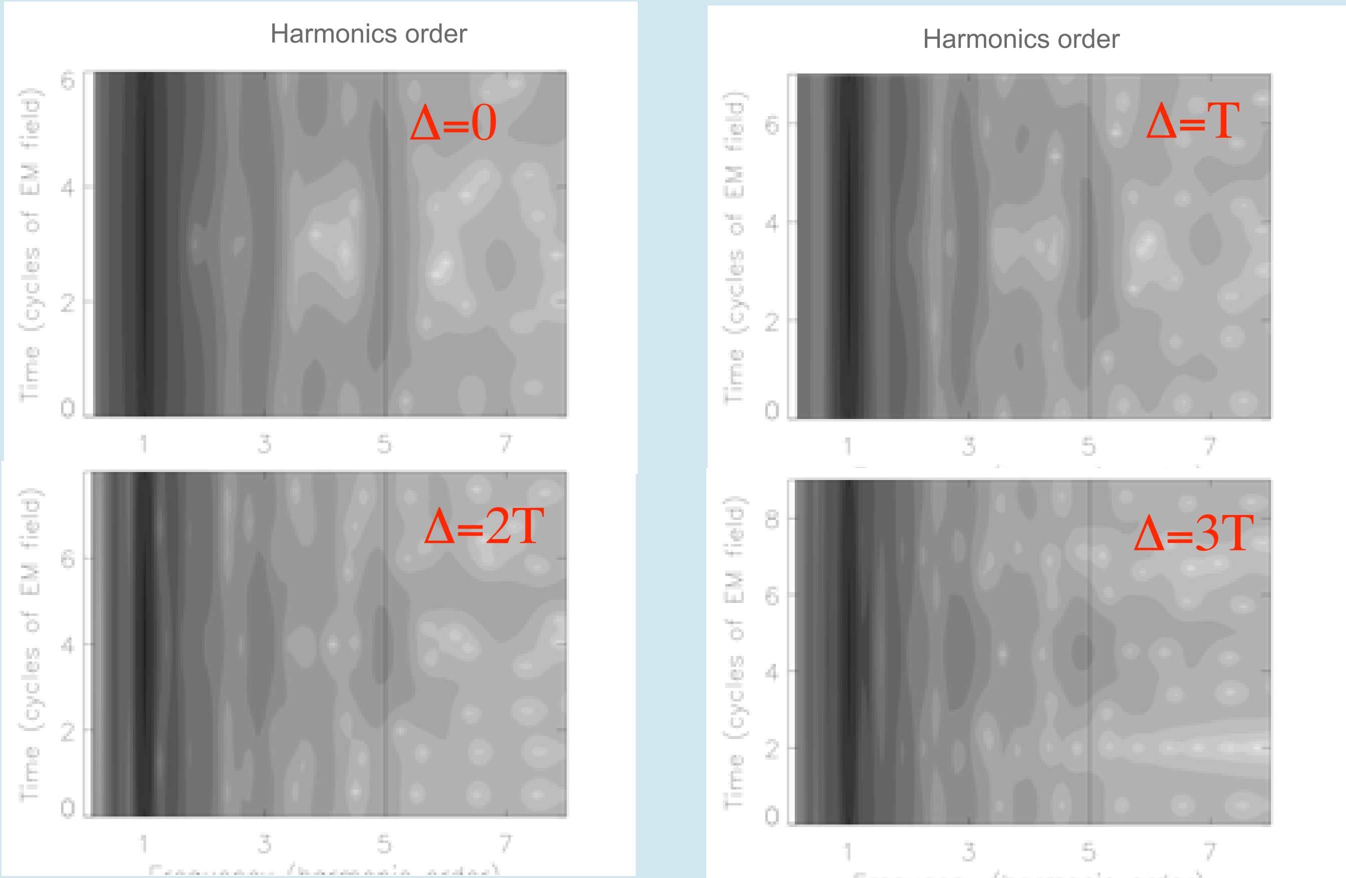

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Harmonics spectrum: wavelet-trajectory analysis

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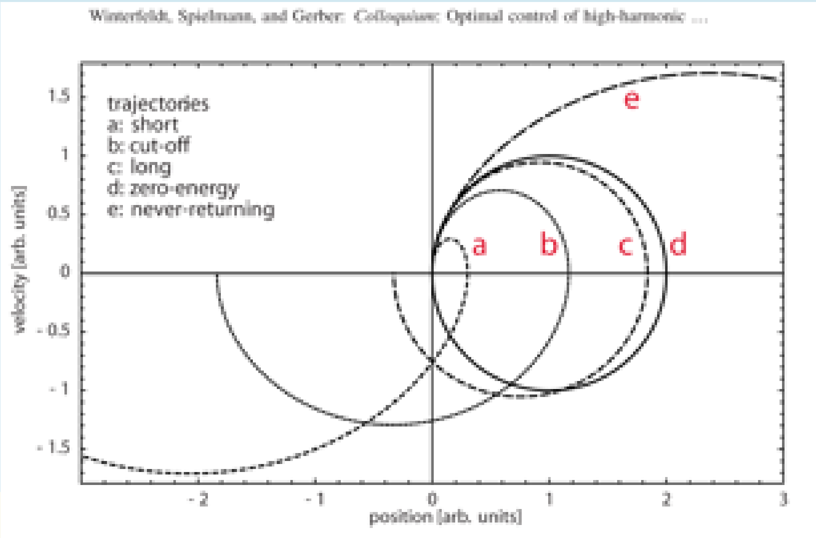

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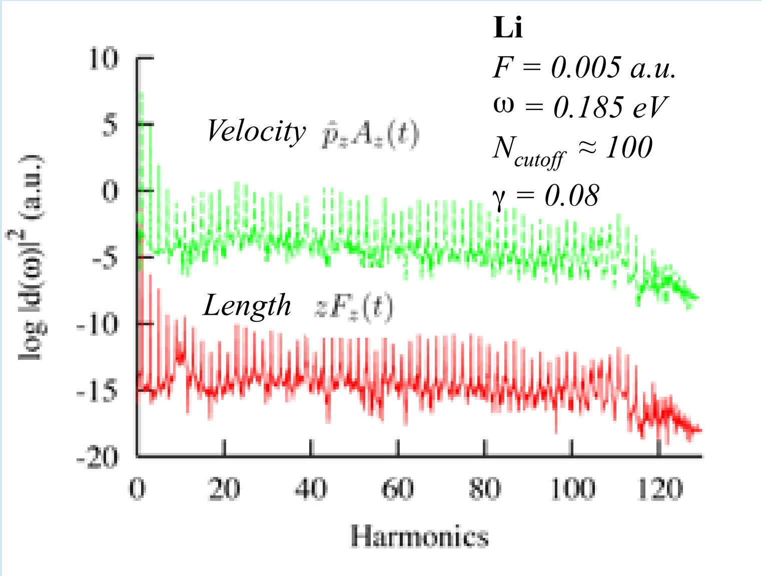

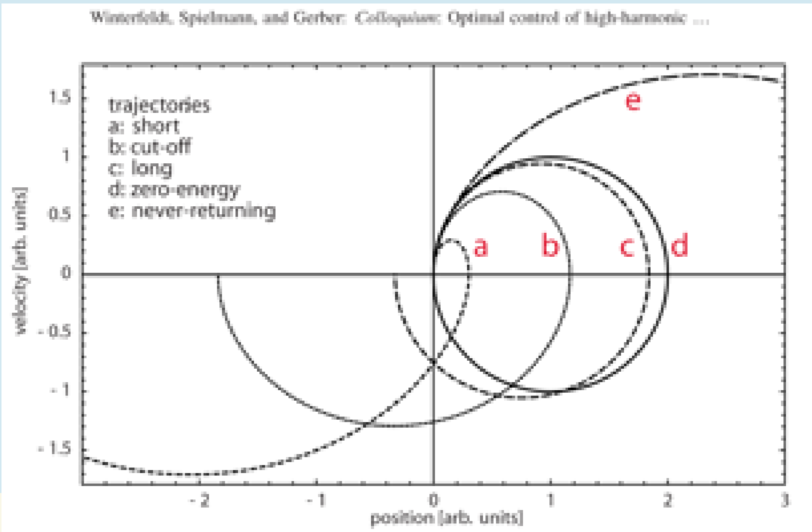

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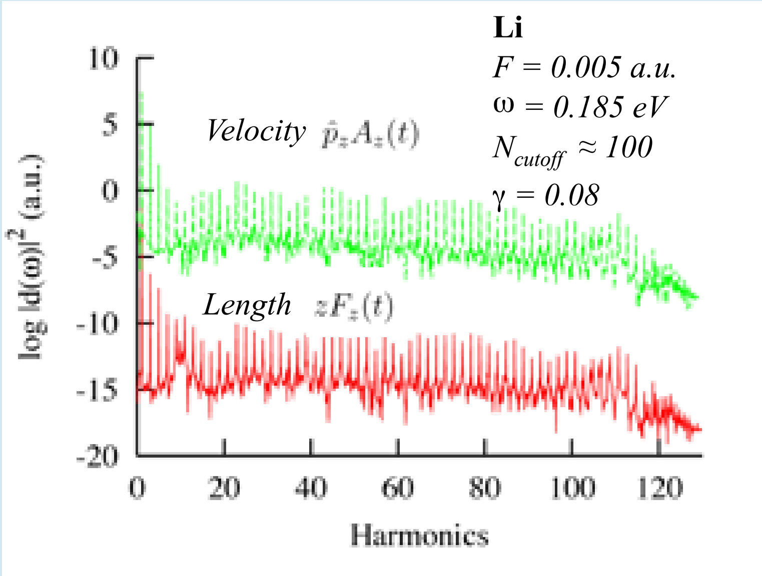

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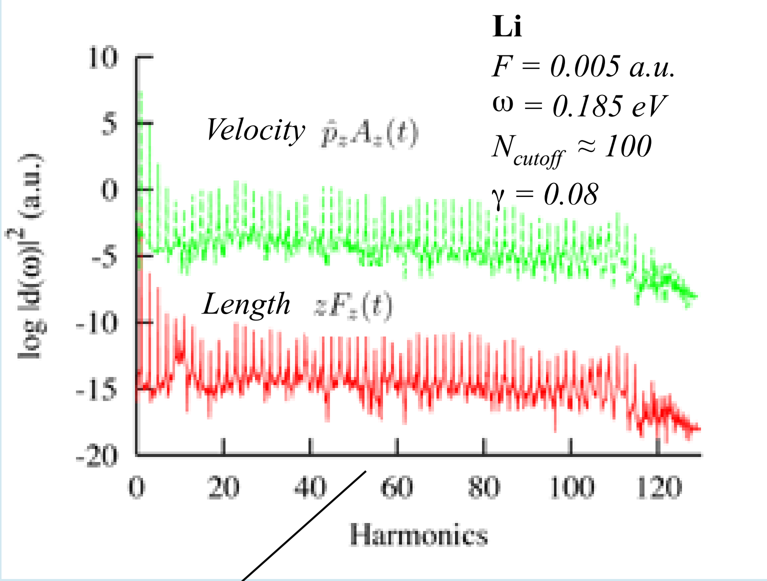

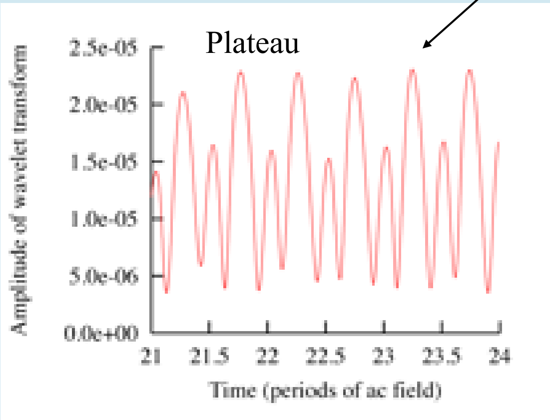

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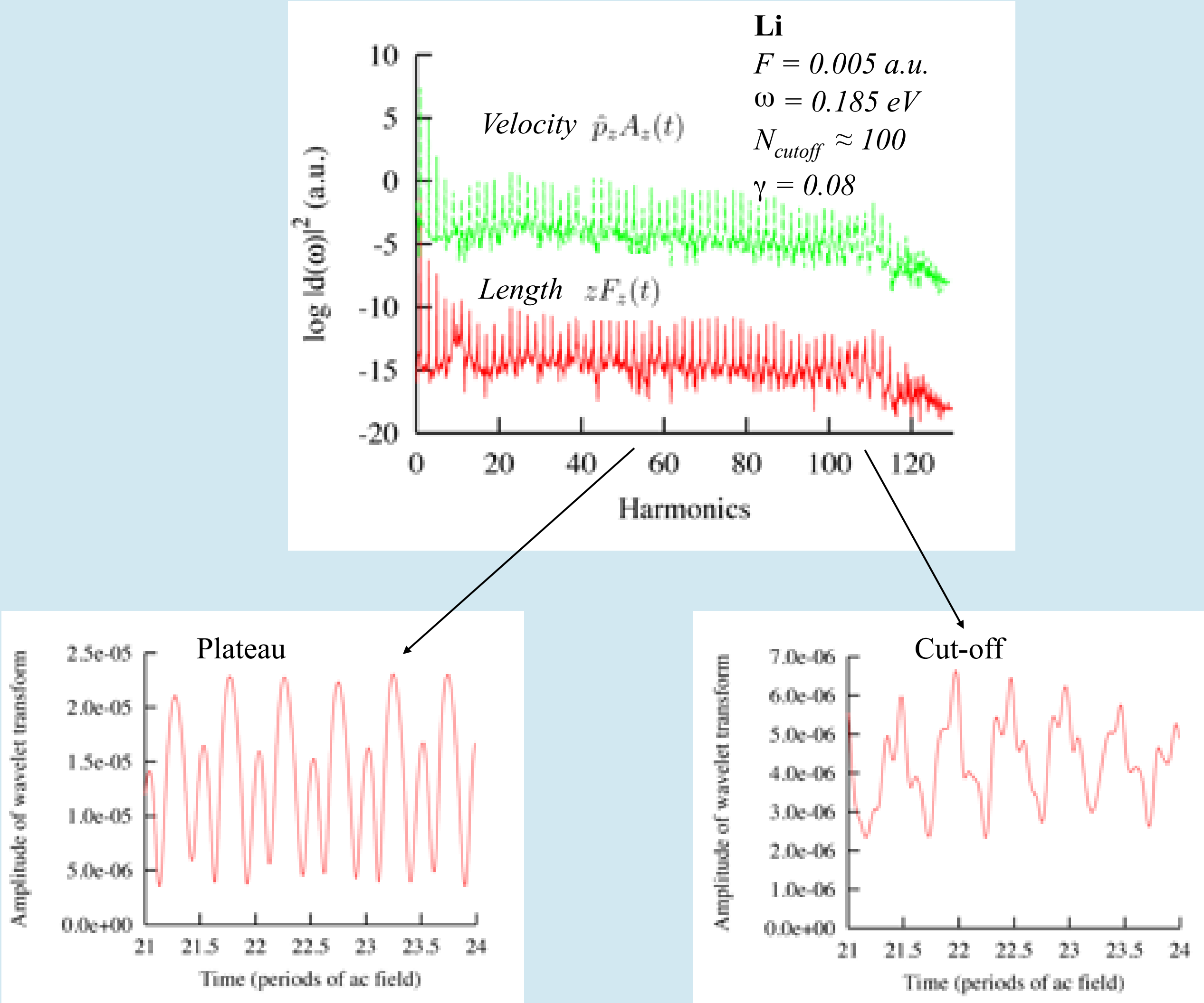

Harmonic spectrum: Effect of Chirp

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Juan J. Carrera and Shih-I Chu
PHYSICAL REVIEW A **75**, 033807 (2007)

$$\hat{V}(\mathbf{r},t) = -\mathbf{F}\cdot\mathbf{r}E(t) = -Fzf(t)\cos[\omega t + \delta(t)],$$

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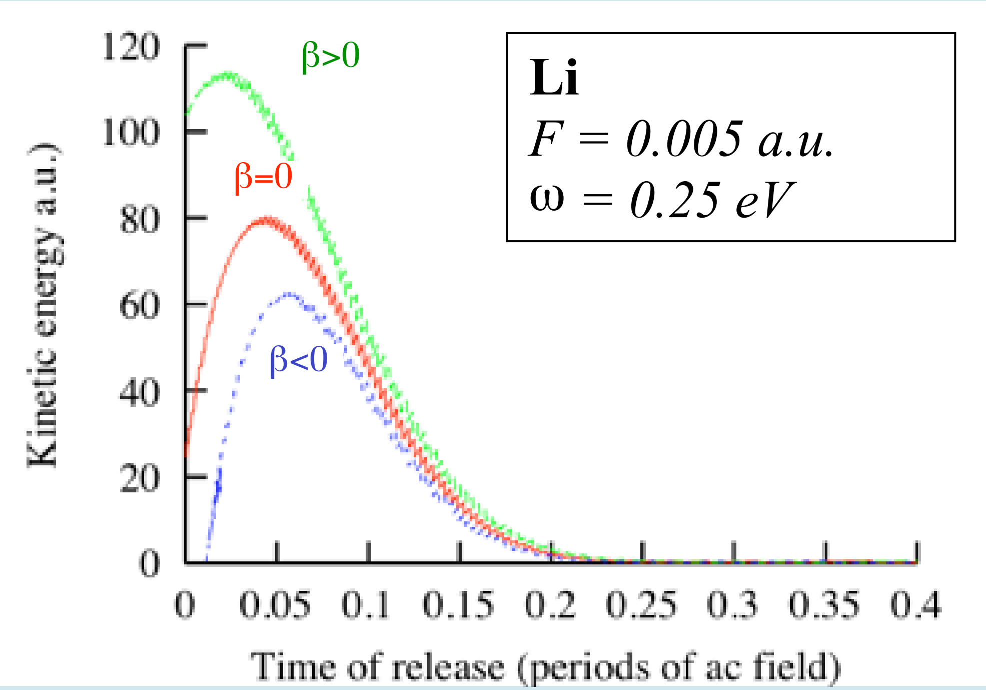

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Friday, May 15, 2009

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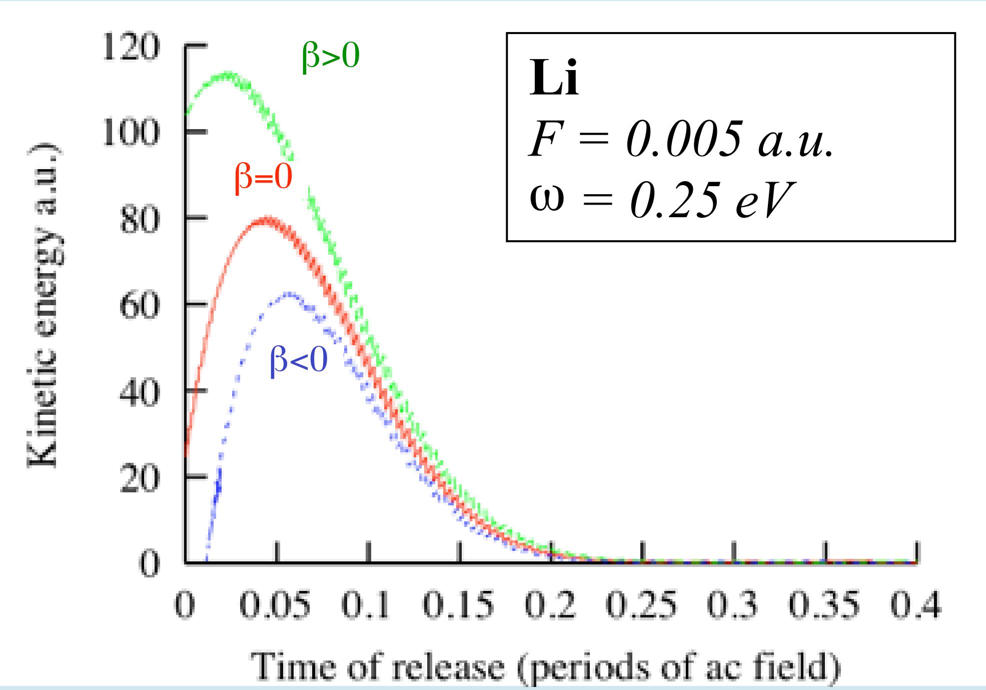

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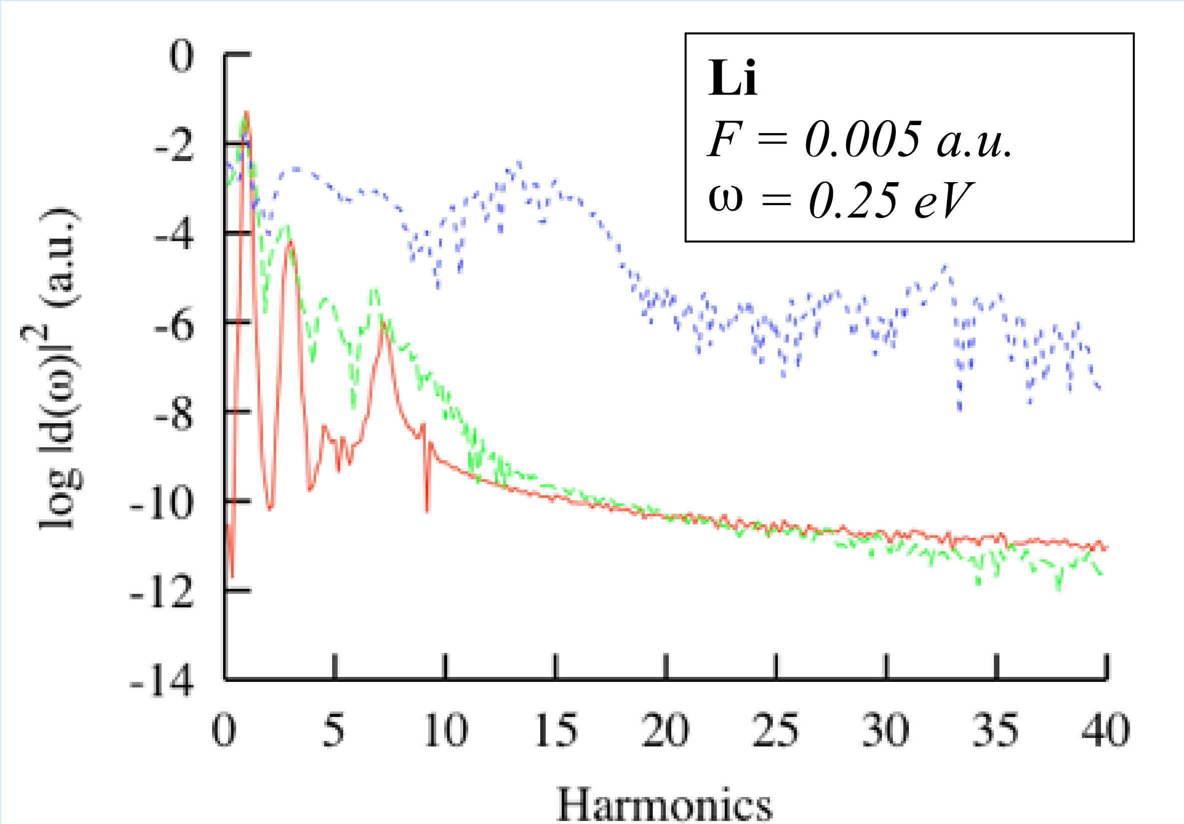
Kinetic energy at the moment of return as function of the time of release field without chirp (red), and positive and negative chirps

Harmonic spectrum: Effect of Chirp

Li
$F = 0.005\ a.u.$
$\omega = 0.25\ eV$

Harmonic spectrum: Effect of Chirp



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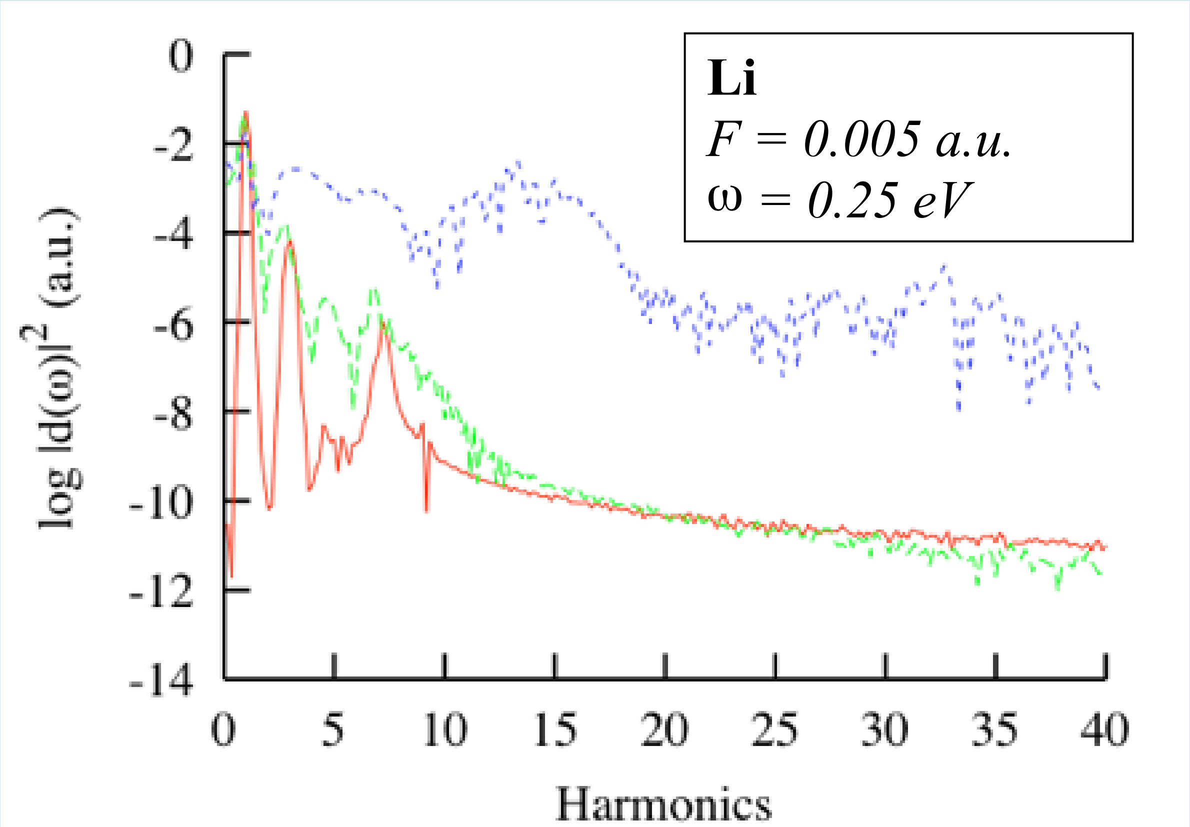

Three calculations for Li for a short pulse with increasing chirp (red, green blue progressively)

Conclusion

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  - Ionization
    - *(2γ,2e) on He*
    - *(4γ,e) on Li*
  - HHG
    - *Li $N_{cut\text{-}off} \approx 10$ optical regime*
    - *Li, Rb $N_{cut\text{-}off} \approx 100$ XUV regime*
    - *Resonant enhancement*
    - *Polarization gating*
    - *Optimal control*

- Further directions
  - Multi-photon many-electron processes on complex targets
  - More HHG control