Нелинейные процессы в ВУФ и рентгеновском диапазонах

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Revolution in VUV and X-ray physics Free electron lasers (FEL)



FLASH – Free Electron Laser in Hamburg (2005) : flux 10¹³ photons/pulse Intensity 10¹³ W/cm² Photon energy 13 eV – 100 eV Pulse duration 10-50 fs

LCLS - Linac Coherent Light Source (2009): flux 10¹² photons/pulse Photon energy 800 eV – 8000 eV Pulse duration 100-200 fs

XFEL at DESY (2013) Intensity 10¹⁶ W/cm² Photon energy 200 eV - 10 keV Pulse duration 100 fs Free electron laser



Density modulation of the electron beam (simulation) along the undulator:



FLASH – Free Electron Laser in Hamburg



Peak power 1-10 GW, pulse duration 10-50 fs

Why it is so exciting? Perspectives

EUV lithography (nano!)

Bio – imaging (bio!)

Study of deluted samples or/and processes with low cross sections

Pump-probe experiments on femtosecond or even attosecond scale, including two colour experiments

Non-linear processes in VUV and X-ray region

Problems:

a. Spectrum of the radiation





b. Time-structure of pulses. Jitter

Solutions:

... Seeded FEL

Non-linear processes in atoms

Multiphoton ionization (MPI)



 $N\hbar\omega + A \to A^+ + e_{ph}$

Prediction: M. Goeppert-Mayer, Ann. d.Phys. 9, 273, 1931 I First observation: G.S. Voronov and N.B. Delone, JETP Letters 1, 66, 1965 $w \propto \mathcal{E}^{2N} = I^N$

Simple arguments: Let suppose that photons are absorbed sequentially, independently with equal probability $w_i \propto \mathcal{E}^2 = I$ then

 $w \propto w_1 w_2 w_3 \dots w_N = w_i^N = \mathcal{E}^{2N} = I^N$

Peculiarities of VUV and X-ray range:

- 1. Dominance of transitions in continuum
- 2. Big role of inner-shell electrons

Multiple ionization of atoms

A.A. Sorokin et al. PRL 99, 213002, 2007

FLASH: I=10¹⁶ W/cm², 13.3 nm (93 eV)



To produce Xe²¹⁺ more than 5 keV or 57 photons of 93 eV are necessary

Sequential ionization – peeling of outermost electrons

 $\begin{array}{c} 1 \\ 10^{1} \\ 10^{2} \\ 10^{3} \\ 10^{4} \\ 10^{5} \\ 10^{6} \\ 10^{12} \\ 10^{13} \\ 10^{14} \\ 10^{14} \\ 10^{15} \\ 10^{16} \\$

Markis et al. PRL 102, 033002, 2009



Space averaging over FEL intensity profile!

 $w \propto I^N$

Two-photon double ionization of atoms



Example: He

Energy conservation: 2

$$2E_{ph} > E_{thr}(A^{2+})$$

Sequential ionization

$$E_{ph} > E_{thr}(A^{2+}) - E_{thr}(A^{+})$$

Direct (non-sequential) ionization

 $E_{thr}(A^{2+}) - E_{thr}(A^{+}) > E_{ph} > E_{thr}(A^{2+})/2$

In direct ionization electron energies are continuously distributed so that $E_1 + E_2 = 2E_{ph} - E_{thr}(A^{2+})$

 $38.5 \text{ eV} < \text{E}_{\text{ph}} < 54.4 \text{ eV}$ (D)

In sequential ionization electron energies are fixed:

 $E_1 = E_{ph} - E_{thr}(A^+); \quad E_2 = E_{ph} - [E_{thr}(A^{2+}) - E_{thr}(A^+)]$

Electron-electron correlations are **not necessary** for sequential double ionization

Direct two photon double ionization of He

Total angle-integrated cross section

L. Nikolopoulos and P. Lambropoulos 2007



Experiment : HHG (Hasegawa et al. 2005) FLASH (A. Sorokin et al. 2007)

J. Feist et al. PRA 77, 043420, 2008



Angular distributions of photoelectrons



 $2\gamma + He \rightarrow He^{++} + 2e$

TDCS at $E_{ph} = 42 \text{ eV}$, $E_1 = E_2 = 2.5 \text{ eV}$ Blue: Feist et al. PRA 77, 043420, 2008 Red: Hu et al. J. Phys. B 38, L35, 2005 Green: Ivanov et al. PRA 75, 033411, 2007

Strong angular correlations between electrons Strong model dependence

Up to now no experimental data !!

Sequential two-photon double ionization

5a. Angular distributions of photoelectrons

Example: Ne



Two-step approach :

1st step $\hbar \omega + Ne \rightarrow Ne^+(^2P) + e_1$ 2nd step $\hbar \omega + Ne^+(^2P) \rightarrow Ne^{++}(^{2S+1}L) + e_2$ Theory: S. Fritzsche, A. Grum-Grzhimailo, E. Gryzlova and N.M.K. J.Phys. B 41, 165601, 2008

For completely uncorrelated ionizations: $W(\vartheta) = W_0(1 + \beta P_2(\cos \vartheta))$

But the intermediate state is aligned (!) along the linear polarization direction, then

for the 2nd electron $W(\vartheta_2) = W_0(1 + \beta_2 P_2(\cos \vartheta_2) + \beta_4 P_4(\cos \vartheta_2))$

Our analysis has shown that for the 1st electron angular distribution has the same form !!

Angular distributions of photoelectrons (FLASH experiments)

M. Braune, U. Becker et al (not published yet)



Angular distributions of photoelectrons: Theory versus experiment

Experiment: M. Braune et al. (not published) Ar (38 eV), Ne (48 eV), Kr (48 eV)
M. Kurka, A. Rudenko et al. J. Phys. B, 42, 141002, 2009 Ne (44 eV)
Theory: S.Fritzsche et al. J. Phys. B, 41, 165601, 2008
A. Kheifetz J. Phys. B, 40, F313, 2007

Example: Ar





5b. Angular correlations between photoelectrons

Calculations by S. Fritzsche et al. J. Phys. B, 41, 165601, 2008 $2\gamma + Ne \rightarrow Ne^{++}(LS) + 2e$ Coincidence !



Calculations by E. Gryzlova et al. Uzhgorod University Scientific Herald, 24, 73, 2009.



Angular correlations: Experiment versus theory



Experiment: M. Kurka, A. Rudenko et al. J. Phys. B 42, 141002, 2009

Ne, $E_{ph} = 44 \text{ eV}$ $2\gamma + Ne \rightarrow Ne^{++}(^{3}P) + 2e$

Theory: S. Fritzsche et al.

Некоторые выводы

- Лазеры на свободных электронах открыли новую эру в фотофизике в области ВУФ и рентгеновского излучения.
- Начались исследования широкого круга нелинейных процессов в атомах, молекулах, кластерах...
- Открываются новые возможности в исследованиях развития атомных процессов во времени.
- В ближайшей перспективе начало исследований биологически важных молекул методом

КОНЕЦ

Спасибо за внимание !