Exploring saturation effects with ultraintense X-ray pulses

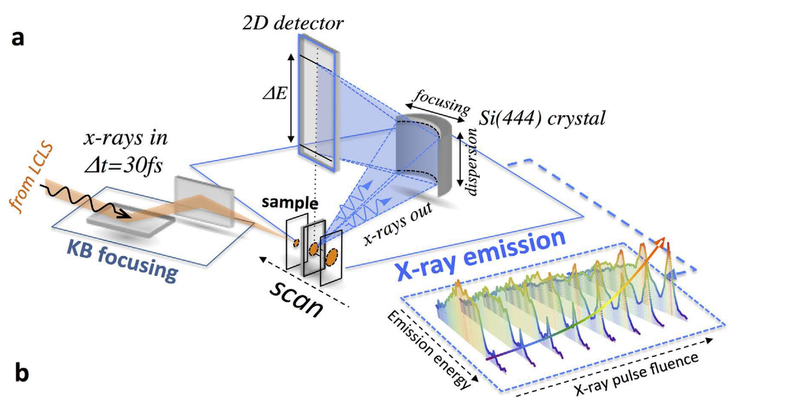
Krzysztof Tyrała[1], Klaudia Wojtaszek[1], Marek Pajek[1],Jakub Szlachetko[1]

[1]Institute of Physics, Jan Kochanowski University, Kielce, Poland.

Introduction

X-ray techniques have evolved over decades to become highly refined tools for a broad range of investigations. Importantly, these approaches rely on X-ray measurements that depend linearly on the number of incident X-ray photons. The advent of X-ray free electron lasers (XFELs) is opening the ability to reach extremely high photon numbers within ultrashort X-ray pulse durations and is leading to a paradigm shift in our ability to explore nonlinear X-ray signals. However, the enormous increase in X-ray peak power is a double-edged sword with new and exciting methods being developed but at the same time well-established techniques proving unreliable. Consequently, accurate knowledge about the threshold and saturation efects for nonlinear X-ray signals is essential. Herein we report an X-ray spectroscopic study that reveals important details on the thresholds for nonlinear X-ray interactions. By varying both the incident X-ray intensity and photon energy, we establish the regimes at which the simplest nonlinear process, two-photon X-ray absorption (TPA), can be observed. Based on time-dependent calculations in 4-level system we established intensity regimes at which TPA process should exhibit saturation effects.

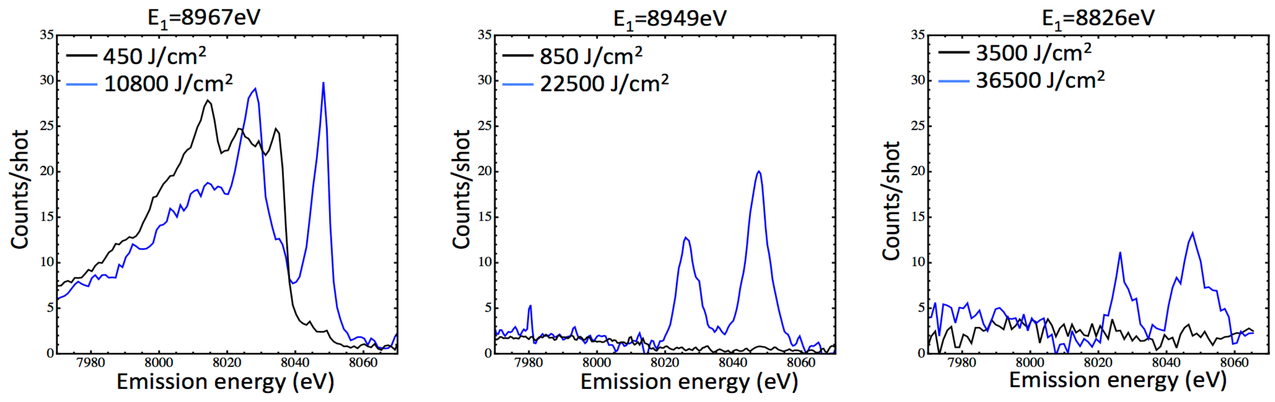
X-Experimental setup for nonlinear two-photon ray absorption spectroscopy.



a) Schematics of the experimental setup showing the Kirkpatrick-Baez (KB) mirror focusing scheme and high energy resolution X-ray emission geometry. The X-ray emission data were recorded for different X-ray pulse fluence by moving the sample along the focus direction of the KB mirrors.

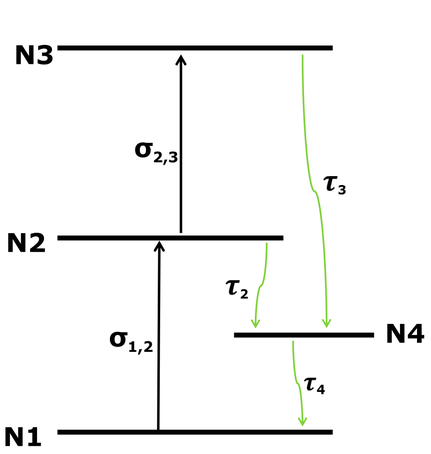
b) High energy resolution X-ray emission spectra recorded at an energy 12 eV below the K-shell ionization threshold of Cu for different incident X-ray fluences. The directions of spectral intensity changes with increasing X-ray pulse fluence are marked by black-dashed (HEROS) and red arrows (Kα emission), respectively. The spectrum difference between the highest and the lowest X-ray pulse fluence is plotted in the bottom panel.

c) Schematic representation of radiative OPA (black arrows) and TPA (red arrows) processes in the off-resonant regime (E1 < Ei). While for the OPA process the emitted X-ray energy (E2) relates directly to the incoming X-ray energy (E1), the TPA mechanism leads to an ionization event and therefore the emitted X-ray energy is constant and equal to the energy difference between the initial (Ei) and final (Ef) electronic states. The Ei and Ef correspond to absolute values of the electron binding energies, and E is the energy of the photo-excited electron. The virtual intermediate state and the initial state are characterized by lifetimes marked by τvi and τi, respectively.



X-ray emission spectra recorded for different incoming X-ray energies tuned below the K-ionization threshold at low (black) and high (blue) X-ray fluences.

Rate equations for 4-level system:



$$\frac{dN1}{dt} = -I(t) \cdot \sigma_{1,2} \cdot N1 + \frac{N4}{\tau_4}$$

$$\frac{dN2}{dt} = I(t) \cdot \left(\sigma_{1,2} \cdot N1 - \sigma_{2,3} \cdot N2\right) - \frac{N2}{\tau_2}$$

$$\frac{dN3}{dt} = -I(t) \cdot \sigma_{1,2} \cdot N1 + \frac{N4}{\tau_4}$$

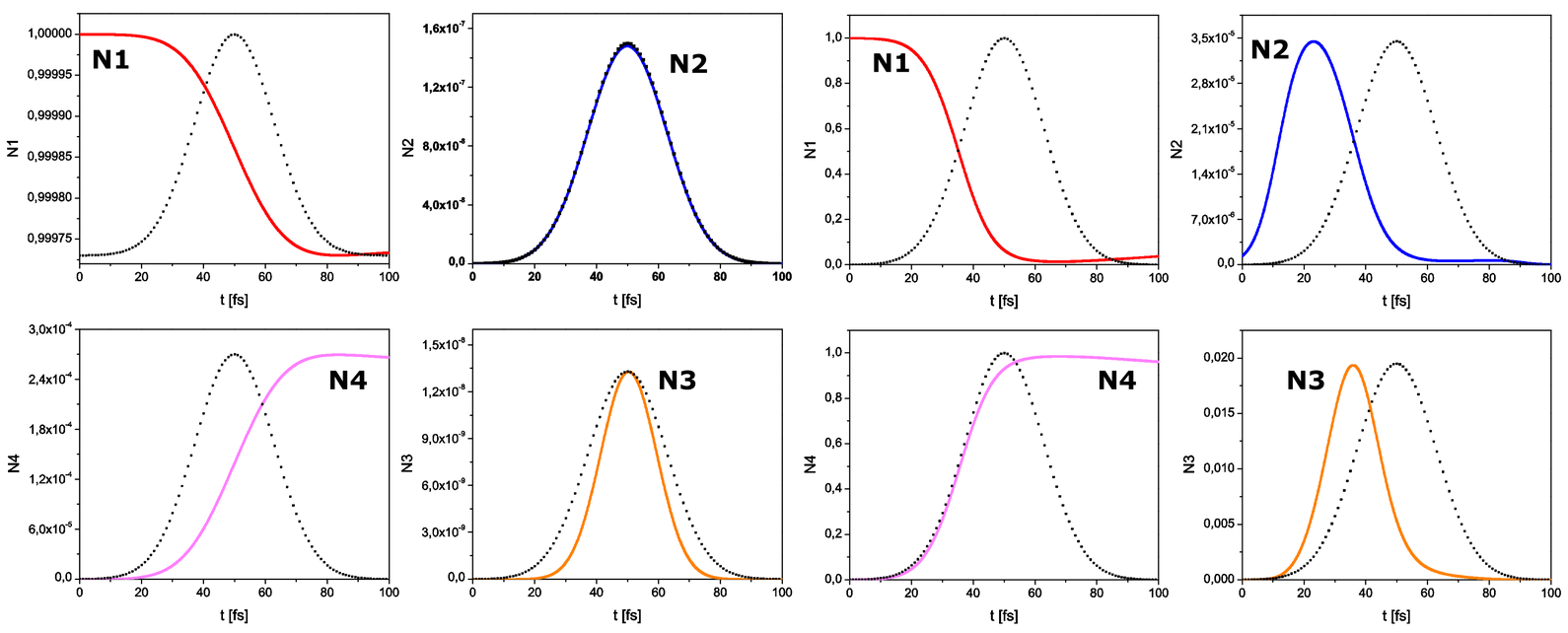
$$\frac{dN1}{dt} = \frac{N2}{\tau_2} + \frac{N3}{\tau_3} - \frac{N4}{\tau_4}$$

$$N1 + N2 + N3 + N4 = 1$$

$$\tau_4 \gg \tau_2, \tau_4$$

Where: N1 is the population of the ground state, N2 the population of the virtual intermediate state, N3 the population of continuum states of atoms (ionized states) through the second X-ray absorption and N4 represents the long living (τ_4 >>τ_2, τ_3) intermediate state. The N4 state accounts for $1s^22p^5$ electronic configuration and consecutive M->L and valence -> L transitions. τ_2, τ_3 and τ_4 for corresponding states. The incident X-ray flux is represented by I in photons/(cm2s) and $\sigma_{1,2}$, $\sigma_{2,3}$ are the cross sections in cm2 for the first and the second absorption step, respectively.

Theoretical temporal evolution for N1,N2,N3 and N4 states :



Representation of time dependent calculations of four-level system, for two different pulse intensity. States N1 are represented as a red, N2 as blue, N3 as orange and N4 as a pink line. Photon pulse (black points) with FWHM value of $30 \cdot 10^{-15}$s is shown for comparison. At low pulse intensity ($1.5 \cdot 10^{31}$ photons/cm^{2}s; $2 \cdot 10^{16}$ W/cm^2), the maximum of states N2 and N3 coincide with pulse intensity maximum. At high pulse intensity ($1.5 \cdot 10^{35}$ photons/cm^{2}s; $2 \cdot 10^{20}$ W/cm^2) value the maximum positions are significantly shifted.

Rate dependence vs. X-ray intensity:

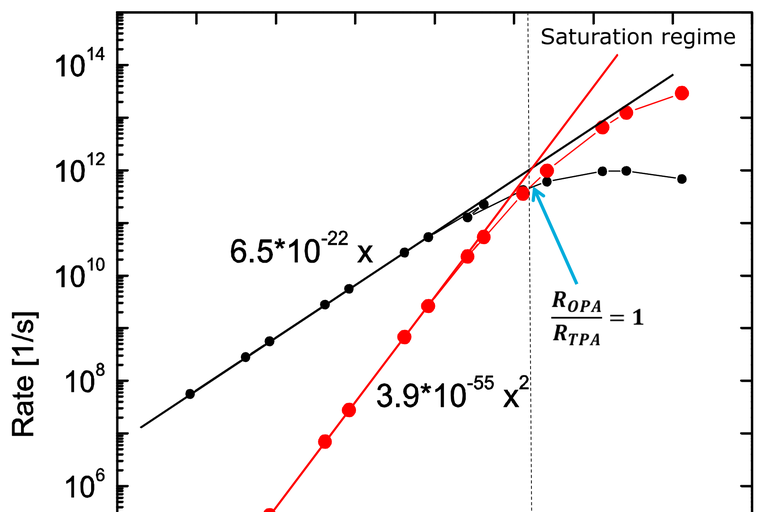


Figure above presents the dependence of rate value (Y) versus photon intensity (X). The data for OPA are plotted by black, and for TPA by red line,respectively.
The straight X and X2 lines correspond to the expected – linear and quadratic dependents for OPA and TPA processes. The crossing point for OPA and TPA rate curves, that corresponds to R_{OPA}/R_{TPA}=1, is shown by an arrow at intensity value of $2.1 \cdot 10^{33}$ photons/cm^{2}s ($3*10^{18}$ W/cm^2).

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J. Szlachetko et al. *Establishing nonlinearity thresholds with ultraintense X-ray pulses*, Scientific Reports, 13 September 2016.