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Two-photon process with X-ray free-electron laser

Kenji Tamasaku

RIKEN SPring-8 Center
E-mail: tamasaku@riken.jp

1. Introduction

The success of X-ray free-electron laser (XFEL) [1,2] is bringing breakthrough in the field of X-ray nonlinear optics. In this paper, we will discuss X-ray two-photon process: creation of double core hole (DCH) in the *K* shell [3] and direct two-photon absorption (TPA) by the *K* shell [4].

2. Sequential double core-hole creation

As the first step to study X-ray two-photon process, we focus on double-*K*-hole creation by sequential photo-ionization. Since the intermediate state is a real state, i.e., the single core-hole (SCH) state, the DCH cross section is larger than the direct two-photon absorption. Nevertheless, it is estimated to be as small as ~10^{-56} cm^4s for medium-Z atoms due to short SCH lifetimes less than 1 fs. We used 1-μm focusing system [5], and successfully observed the X-ray fluorescence, $K^h\alpha_{1,2}$, from the DCH state, which are separated from the SCH fluorescence, $K\alpha_{1,2}$.

The observation of $K^h\alpha_{1,2}$ gives a clear evidence that the intense X-rays can interact with the SCH state, in spite of the very short lifetime. The electronic property in the SCH state is considered to be quite different from the neutral state; thus, diffraction and absorption by the SCH atoms should affect the XFEL applications.

In addition, we analyze the signal ratio between $K\alpha_{1,2}$ and $K^h\alpha_{1,2}$, and estimate the pulse duration. The analysis is based on evaluation of the effect of the spiky and the pulsed temporal structure of XFEL operated in the self-amplified spontaneous emission (SASE) mode. We finally determine the pulse duration of SACLA to be 2.5 fs, in good agreement with an independent estimation of the electron beam by the accelerator.

3. Two-photon absorption

The intermediate state of TPA is a virtual state with a lifetime of ~$1/\omega$, so that the cross section is considered to be about four orders of magnitude smaller than the DCH cross section. Using more tight focusing down to ~100-nm size [6], we observed the TPA signal from germanium. We find that the pulse-energy dependence of the TPA signal deviates from quadratic and is suppressed at higher pulse energies (Fig.1). In the present experiment, the pump photon energy is tuned just above the *K*-shell TPA threshold. The *L*-shell photo-ionization makes the *K*-shell binding energy larger than double the photon energy of pump, and suppresses TPA.

We simulate the population dynamics of various electronic configurations to calculate the pulse-energy dependence of the TPA signal, and find good agreement with the experimental result, supporting our scenario.

3. Conclusions

We successfully observed X-ray two-photon processes: the sequential DCH creation and the direct two-photon absorption using XFEL. The DCH experiment shows clearly that the effect of X-ray interaction with the SCH atom need to be considered for applications of high intensity XFEL. The observation of TPA leads to X-ray TPA spectroscopy. However, the competition between TPA and one-photon ionization indicates that there may be an upper limit of the X-ray intensity to investigate the ground state property of material.

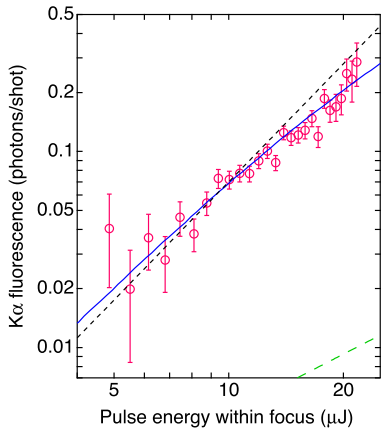

Figure 1. The pulse-energy dependence of the TPA signal (open circle), the line with slope of 2 (dashed line), and the simulation (solid line) [4].

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References

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