Determination of Absolute Cross-Sections of Nonresonant EUV-UV Two-Color Two-Photon Ionization of He

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Abstract: Single-shot photoelectron spectroscopy was performed for nonresonant EUV-UV two-color two-photon ionization of He. From data analysis on the shot-by-shot basis, the absolute cross-section was determined to be $\sigma^{(2)}(597\text{nm}, 268\text{nm}) = 4.1(6)\times10^{-52}$ cm^4 s.
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1. Introduction

Advances of free electron laser (FEL) technologies have made it possible to investigate various nonlinear phenomena in the EUV region [1]. Experiments by using optical laser synchronized with EUV-FEL provide fundamental information on how nonlinear optical processes take place by absorbing two different frequencies. In such processes, the simultaneous absorption depends not only on the field intensity of each laser pulse but also on the temporal overlapping between them. However, there is considerable instability in the synchronization due to technical limitations [2]. Such a timing jitter between the two pulses prevents from being kept the temporal overlapping optimal, and makes it difficult to evaluate the cross-section. Here, we show that single-shot analysis enables us to extract the optimal overlap in time, thereby evaluating the cross-section in more accurate manner than those derived from average values. We determine non-resonant two-color two-photon ionization of He by comparing the cross-section of Ar by single photon ionization [3]. This study provides quantitative information on the new nonlinear optical processes induced by two different frequency components which would be the basis of applications to multi-color experiments such as time-resolved studies.

2. Experiment

An EUV-FEL pulse (59.7 nm) obtained from the SCSS test accelerator facility at RIKEN Harima and an ultrashort UV laser pulse (268 nm) synchronized with FEL were focused to gaseous He and Ar in the interaction region. The mean time-delay between FEL and UV laser pulses was controlled by an optical stage with a resolution of 1 μm (Fig. 1(a)). Electrons formed by photoionization with FEL and UV laser pulses were detected shot-by-shot by a magnetic bottle type photoelectron spectrometer. The electron energy was calibrated by using atomic lines of oxygen and Auger lines of xenon for which the 3rd order harmonics of FEL at 51 nm was used.

3. Results and discussion

Figure 1(b) shows a part of photoelectron spectrum at Δt = 0 ps. Photoelectron peak at 5 eV is due to electrons formed by one-photon ionization of Ar while He peaks appear about 1 eV. From the energy conservation the peak at 0.6 eV is assigned to photoelectrons produced by non-resonant (NR) two-color two-photon ionization of He, while the peak at 1.3 eV is attributed to those by resonant (R) two-color two-photon ionization via the 2^1P state. The temporal behaviors of the NR and R peaks are plotted in Fig. 1(c). The intensity of the R component increases until Δt = 0.4 ps and stays almost constant up to Δt = 1.5 ps, which is in line with the lifetime (560 ps) of the 2^1P state. In contrast, the NR peak exhibits the maximum at Δt = 0 ps. The full width at half maximum (FWHM) of the trace was obtained to be 0.36(3) ps by carrying out a least-square fit to a Gaussian function. Since both FEL and UV laser pulse durations are about 100 fs, the temporal resolution reflects primarily to the timing jitter between the FEL and UV laser pulses. This timing jitter prevents from keeping a stable temporal overlap in the time-resolved measurements, thereby resulting in accumulation of non-resonant signals at various time delays in the averaged spectra in Fig. 1(a).

Figure 1(d) shows a distribution of the NR signals obtained from single-shot spectra at the nominal Δt = 0 ps. The broad distribution with a large contribution at the zero signal position reflects no temporal overlapping mainly

the timing jitter. To evaluate the effect of timing jitter δ, we calculate temporal overlapping $T^{(2)}(\delta)$ at Δt = 0 ps, which can be written as

$$T^{(2)}_{\mathrm{EUV,UV}}(\delta) = \int_{-\infty}^{+\infty} f_{\mathrm{EUV}}(t) f_{\mathrm{UV}}(t-\delta)dt, \quad (1)$$

where $f(t)$ is a pulse envelope normalized by the peak maximum. Figure 1(e) shows a $T^{(2)}$ distribution without the timing jitter (δ = 0). The distribution peaked at ~1, which reflects the intensity fluctuation of each FEL pulse that is simulated by using the partial coherence method [4]. On the other hand, the simulation including the timing jitter as shown in Fig. 1(f) is in good agreement with the experiment. From the comparison between Figs. 1(d) and (f), the signal intensity for the non-resonant component at Δt = 0 ps is estimated to be $S^{(2)}_{\mathrm{He+}}$ = 3.5 which is underestimated by a factor of 0.4 when the average value is used.

Since the cross-section of non-resonant two-color two-photon ionization of He in the present study can be expressed as [3],

$$\sigma^{(2)}_{\mathrm{He}}(h\nu_i, h\nu_j) = \frac{S^{(2)}_{\mathrm{He^+}}}{S^{(1)}_{\mathrm{Ar^+}}} \frac{n^0_{\mathrm{Ar}}}{n^0_{\mathrm{He}}} \frac{T_i^{(1)}}{T^{(2)}_{i,j}(\Delta t)} \frac{h\nu_j}{I_{0,j}} \sigma^{(1)}_{\mathrm{Ar}}(h\nu_i), \quad (2)$$

where $h\nu_i$ and $h\nu_j$ is photon energy of the pulse i (=EUV) and j (=UV), $S^{(2)}_{\mathrm{Ar+}}$ is the integrated intensity of the Ar peak at 5 eV, $n^0_{\mathrm{Ar}}/n^0_{\mathrm{He}}$ is the ratio of the number density of Ar and He, $T_i{}^{(1)}$ is the integral of the $f(t)$ for the EUV-FEL pulse, $I_{0,j}$ is the field intensity of the UV pulse, and $\sigma_{\mathrm{Ar}}{}^{(1)}$ is the cross-section of Ar. By using Eq.(2) with the following parameters such as $n^0_{\mathrm{Ar}}/n^0_{\mathrm{He}} = 1.2\times10^{-4}$, $T_{\mathrm{EUV}}{}^{(1)}/\, T_{\mathrm{EUV,UV}}{}^{(2)}(0) = 1.4$, $I_{0,\mathrm{UV}} = 1.4\times10^{13}$ W/cm^2, $\sigma_{\mathrm{Ar}}{}^{(1)} = 36.1$ Mb [5], the nonresonant cross-section is determined to be $\sigma^{(2)}(597\ \mathrm{nm}, 268\ \mathrm{nm}) = 4.1(6)\times10^{-52}$ cm^4 s.

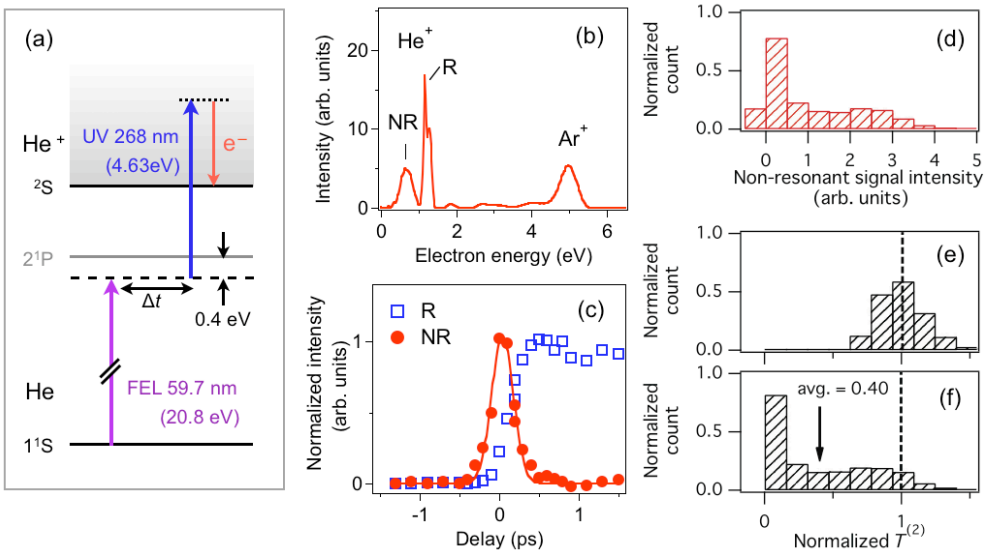

Fig. 1. (a) Schematic diagram of two-color two-photon ionization of He using EUV-FEL and UV laser pulses (b) Photoelectron spectrum at Δt = 0 ps. (c) Temporal behavior for resonant (R) and nonresonant (NR) components of the He peak. (d) Histogram of the NR signal at Δt = 0 ps. (e) Simulated histogram of $T^{(2)}(0)$ without the timing jitter. (f) Same as (e), but the timing jitter is included.

4. Summary

We have shown that single-shot analysis allows us to evaluate accurate cross-sections by comparing experiment with simulation including jitter contribution. The present study clearly shows that single-shot photoelectron spectroscopy is a powerful tool for the precise measurements of cross-section for multicolor absorption occurring in non-resonant processes.

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References

[1] e.g. M. Yabashi et al., "Compact XFEL and AMO sciences: SACLA and SCSS," J. Phys. B **46,** 164001 (2013).
[2] A. Azima et al., "Time-resolved pump-probe experiments beyond the jitter limitations at FLASH," Appl. Phys. Lett. **94,** 144102 (2009).
[3] M. Fushitani et al., "Nonresonant EUV-UV two-color two-photon ionization of He studied by single-shot photoelectron spectroscopy," Phys. Rev. A **88,** 063422 (2013).
[4] T. Pfeifer et al., "Partial-coherence method to model experimental free-electron laser pulse statistics," Opt. Lett. **35,** 3441-3443 (2010).
[5] W. Chan et al., "Absolute optical oscillator strengths for the electronic excitation of atoms at high resolution. III. The photoabsorption of argon, krypton, and xenon," Phys. Rev. A **46,** 149-171 (1992).