Photoelectron angular distributions for the two-photon ionization of helium by ultrashort extreme ultraviolet free-electron laser pulses

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Abstract

The two-photon ionization of helium atoms by ultrashort extreme-ultraviolet free-electron laser pulses, produced by the SPring-8 Compact SASE Source test accelerator, was investigated at photon energies of 20.3, 21.3, 23.0 and 24.3 eV. The angular distribution of photoelectrons generated by two-photon ionization is obtained using a velocity map imaging spectrometer. The phase-shift differences and amplitude ratios of the outgoing s and d continuum wave packets are extracted from the photoelectron angular distributions. The obtained values of the phase-shift differences are distinct from scattering phase-shift differences when the photon energy is tuned to a resonance with an excited level or Rydberg manifold. The difference stems from the co-presence of resonant and non-resonant path contributions in the two-photon ionization by femtosecond pulses. Since the relative contribution of both paths can be controlled in principle by the pulse shape, these results illustrate a new way to tailor the continuum wave packet.

(Some figures may appear in colour only in the online journal)

1. Introduction

Two-photon processes are well-known phenomena and have been extensively investigated for decades, both experimentally and theoretically. Also, these processes have been used in a variety of applications in laser optics and spectroscopy. It is also well known that the angular distribution of photoelectrons generated by multi-photon processes is directly related to the
relative amplitudes and the relative phase between different partial waves [1–6]. However, these earlier works dealt with the laser pulses in the optical range whose pulse width is very long in comparison with the modern standard of femtosecond (fs) laser technology.

The advent of near infrared (IR) fs lasers generated new opportunities to investigate the angular distributions of the resonant enhanced multi-photon ionization (REMPI) of low ionization energy samples such as alkaline metals; here, one of the main interests was the coherent control of the REMPI processes by the very intense near IR chirped fs pulses [7, 8]. For other directions of investigations for photoelectron angular distributions (PADs) explored by the fs lasers, such as the time-solved measurements, see, for example, a recent review article [9].

The advent of femtosecond intense short-wavelength lasers, i.e. extreme ultraviolet (EUV) [10, 11] and x-ray [12, 13] free-electron lasers (FELs), with fs pulse widths, has led to a renewed interest in two-photon processes in the EUV and x-ray regimes (see, e.g. [14–22]). In this paper, we address a new opportunity generated by the ultrashort EUV–FEL pulses to deviate the phase-shift difference between ionization channels from the scattering phase-shift difference, which is otherwise intrinsic to the target atom or molecule. This will eventually open a new avenue for the coherent control of the continuum wave packets by the EUV pulses.

The simplest possible two-photon process may be the single-colour two-photon single ionization of helium (He) atoms. For a theoretical study, see, for example, the work by Nikolopoulos et al [23]; van der Hart and Bingham [24] and the references therein. Kobayashi et al [25] were the first to observe this process and used it for an autocorrelation measurement of high-order harmonic pulses, and Moshammer et al [26] recently used it for an autocorrelation measurement of the EUV–FEL pulses provided by the SPring-8 Compact SASE Source (SCSS) test accelerator [11]. The absolute two-photon ionization (TPI) cross sections of He were measured using an intense high harmonic source [27] as well as the SCSS test accelerator [28]. Hishikawa et al [29] recently investigated the two- and three-photon ionization of He at the SCSS test accelerator by photoelectron spectroscopy using a magnetic bottle spectrometer. The PAD for the single-colour TPI of He, however, has not been investigated so far, except in the context of multi-colour above-threshold ionization [30, 31].

The two-photon single ionization of He produces a continuum electron wave packet which is a superposition of s (1s e S) and d (1s e d D) partial waves. For the wavelength range considered in this study (51–61 nm), the electric dipole approximation is valid, and there is no two-photon excitation of auto-ionizing resonances. The PAD provides information about the ratio of amplitudes for the s and d partial waves and their relative phase. Haber et al [32] have recently measured the PAD from near IR- and UV-laser-induced (single-photon) ionization of the 1snp excited states, which are prepared using high-order harmonics. A similar experiment has also been performed by O’Keefe et al [33] using synchrotron radiation for the preparation of excited states and a laboratory laser for the ionization. Both these experiments have confirmed that the relative phase between the s and d waves extracted from the measured PADS agrees well with the values expected from the theoretically calculated scattering phase shifts. In these studies, the results were dominated by the dynamics of a resonant intermediate state. In this work, in contrast, we investigate the situation in which resonant and non-resonant pathways coexist with similar probabilities, namely (single-colour) TPI by an fs pulse. As theoretically predicted by Ishikawa and Ueda [34], this causes changes in the relative phase between the s and d waves, which depends on the wavelength. When the central wavelength is tuned to the resonance with an excited level, for the case of fs pulses, both contributions from resonant and non-resonant ionization paths are significant, which leads to a relative phase between s and d that is distinct from the corresponding scattering phase difference. It is expected that this change in the phase difference can be revealed by means of a PAD measurement. The contribution of the non-resonant portion becomes negligible for longer pulses and for the two-step schemes as mentioned above [32, 33].

In this study, we use the velocity map imaging (VMI) technique [35, 36] to measure the PAD from the single-colour TPI of He by fs EUV–FEL pulses. The anisotropy parameters are obtained from the PAD to extract the phase differences δ and the amplitude ratios of the s and d partial waves. Our results show the presence of an extra phase shift due to comparable contributions from resonant and non-resonant paths, in agreement with our recent theoretical prediction [34] and simulation results obtained by solving the full time-dependent Schrödinger equation (TDSE).

2. Experiment

The experiments were carried out with the SCSS test accelerator in Japan. This FEL light source provided EUV pulses with a duration of ~30 fs [26] and a full-width-at-half-maximum spectral width of ~0.2 eV [37]. This experiment was designed in the following manner. The photon energies were selected to be 20.3, 21.3, 23.0 and 24.3 eV. A photon energy of 20.3 eV is well below the excitation energy of the lowest resonance 1s2p P (21,218 eV) [38] and, thus, ionization is expected to be dominated by direct, non-resonant TPI. Conversely, the 1s2p P and 1s3p P (23,087 eV) [38] states can be resonantly excited at 21.3 and 23.0 eV due to the bandwidth of the FEL pulse, and according to our recent theoretical predictions [34], we may expect that both resonant and non-resonant contributions are involved in the photoionization process. A photon energy of 24.3 eV corresponds to excitation of the 1snp P Rydberg manifold (n ~ 7). The spectral width covers several Rydberg members from 1s6p P to 1s9p P. Under this condition, we also expect that resonant and non-resonant path contributions are co-present. Doubly excited states are not accessible through two-photon transitions by the photon energy range used in this study.

The FEL beam from the SCSS test accelerator was steered by two upstream plane SiC mirrors, passed through a gas monitor detector (GMD) and then entered into the prefocusing system of the beam line. The GMD was calibrated using a
cryogenic radiometer [39]. The average pulse energy measured by the GMD during the experiments was 7–11 μJ, with a standard deviation of 2–4 μJ. The focusing system, with a focal length of 1 m, consisted of a pair of elliptical and cylindrical mirrors coated with SiC [40]. The reflectivity of each mirror was 70%. Before entering the interaction chamber, the FEL beam passed through two sets of light baffles, each consisting of three skimmers with 4.0 mm and 3.5 mm diameters, respectively. These baffles successfully removed the majority of the scattered light specularly and non-specularly reflected by the two mirrors, without reducing the photon flux. The FEL beam was then focused on a He beam at the centre of a VMI spectrometer [41]. The measured focal spot size was ∼13 μm in radius, resulting in an average intensity of typically 2–3 × 10^{13} W cm^{-2}.

Electrons produced by the TPI of helium atoms by the FEL pulses were accelerated, perpendicular to both the propagation and linear polarization axes of the FEL beam, towards a position-sensitive detector consisting of a set of microchannel plates (MCPs) followed by a phosphor screen. The positions of the detected electrons were recorded using a gated CCD camera synchronized to the arrival of the FEL pulse in the interaction chamber. A 200 ns electrical gate pulse was applied to the back of the MCPs. In our measurements, the PAD has cylindrical symmetry along the FEL polarization, and we can retrieve the three-dimensional (3D) photoelectron momentum distribution from the measured 2D projection of the momentum distribution using a mathematical procedure based on an Abel inversion, where we express the PAD in terms of a Legendre expansion. Examples of raw and inverted images are given in figure 1.

3. Results and discussion

Figure 2 displays the PADs $I(\cos \theta)$ obtained at four different photon energies, as a function of cosine of the polar angle $\theta$ relative to the polarization axis. The PADs $I(\cos \theta)$ can be described by the following expression:

$$I(\cos \theta) = \frac{I_0}{4\pi} [1 + \beta_2 P_2(\cos \theta) + \beta_4 P_4(\cos \theta)],$$

where $I_0$ is the angle-integrated intensity, and $\beta_2$ and $\beta_4$ are the anisotropy parameters associated with the second- and fourth-order Legendre polynomials $P_2(x)$ and $P_4(x)$, respectively.

Values of $\beta_2$ and $\beta_4$ obtained from the experimental PADs are listed in table 1. To investigate the processes involved in the TPI of He, we have performed numerical simulations, by solving the full-dimensional two-electron TDSE using the time-dependent close-coupling method [42–46]. We have employed chaotic pulses with a mean intensity of $2.5 \times 10^{13} W cm^{-2}$, generated...
by the partial-coherence method described in [47], for a coherence time of 8 fs and a mean pulse width of 28 fs (full-width-at-half-maximum), as recently measured by second-order autocorrelation [26], both assumed to have Gaussian profiles on average. The effect of the slight difference of the actual pulse profiles from Gaussian is small compared with that of the difference between coherent and chaotic pulses as well as that of shot-to-shot fluctuation. One can see good agreement between the experimental and simulation results in figure 2 as well as in figure 3.

The PAD results from an interference of the s and d partial waves, and can be expressed as

$$
\alpha |c_0 e^{i\delta_{c,0}} Y_{00} - c_2 e^{i\delta_{c,2}} Y_{20}|^2 = |c_0| e^{i\beta_{sc}} Y_{00} - |c_2| e^{i\beta_{sc}} Y_{20}|^2, \tag{2}
$$

where $c_l$ denotes the complex amplitude of a final state with an angular momentum $l$, $\delta_{c,l}$ denotes the scattering phase shift intrinsic to the corresponding continuum eigenfunction and $\delta_l = \arg c_l + \delta_{c,l}$ is the phase of each partial wave. If we note that $Y_{0}(\theta) = \sqrt{\frac{2l+1}{4\pi}} P_l(\cos \theta)$ and define $W = |c_0/c_2|$ and $\Delta = \delta_0 - \delta_2$, then, these are related to the anisotropy parameters as

$$
\beta_2 = \frac{10}{W^2 + 1} \left( \frac{1}{7} - \frac{W}{\sqrt{5}} \cos \Delta \right), \quad \beta_4 = \frac{18}{7(W^2 + 1)}, \tag{3}
$$

and thus $W$ and $\Delta$ can be extracted from the PAD. The experimentally obtained values of $W$ and $\Delta$ are listed in table 1. Furthermore, in figure 3, the experimental values of $W$ and $\Delta$ are compared with the values extracted from TDSE simulations as described in [48], as a function of photon energy $h\omega$. The agreement between the experimental and theoretical values is reasonable for both $W$ (panel (a)) and $\Delta$ (panel (b)). For comparison, the theoretical values of the scattering phase-shift difference $\Delta_{sc} \equiv \delta_{sc,0} - \delta_{sc,2}$ [49] are also plotted by the solid line.

Within the framework of the second-order time-dependent perturbation theory [34], $c_l$ can be defined in such a way that its real and imaginary parts correspond to the resonant and non-resonant paths, respectively. If the pulse is non-resonant, then $c_0$ and $c_2$ are pure imaginary, resulting in $\Delta = 0$. In this study, the measurement at $h\omega = 20.3$ eV corresponds to this situation; indeed, we find that $\Delta \approx 0$ in this case, as shown in figure 3.

Let us now turn to the situation where the pulse is resonant with an excited state or the Rydberg manifold. If a resonant TPI path is dominant, $\Delta$ is again close to $\Delta_{sc}$, since $c_0$ and $c_2$ are both real. On the other hand, if the contributions from both the resonant (via a single or several resonant levels) and non-resonant paths (via all the intermediate levels) are present, then $\Delta \neq \Delta_{sc}$ in general [34] and an extra phase-shift difference $\Delta_{ex} \equiv \Delta - \Delta_{sc} = \arg c_0/c_2$ occurs. In this study, the pulses with $h\omega = 21.3$, 23.0 and 24.3 eV induce resonant TPI via $1s2p^1P$, $1s3p^1P$, and a Rydberg manifold ($1snp^1P$ with $n = 6 – 9$), respectively. We can see in figure 3 that the relative phase $\Delta$ deviates from the scattering phase-shift difference $\Delta_{sc}$ for these three photon energies; the difference $\Delta_{ex}$ increases gradually with increasing photon energy and becomes very significant at 24.3 eV. At $h\omega = 21.3$ and 23.0 eV, the simulation values of $\Delta_{ex}$ are slightly smaller than for a lower intensity, indicating the departure from the perturbative limit due to the high intensity. The observed deviation $\Delta_{ex}$ clearly demonstrates the co-presence of the resonant and non-resonant path contributions in these experiments, as recently predicted in [34]. This situation presents a contrast to the case of the photoionization from excited $p$ states [32, 33], where the non-resonant path is absent and, as a result, $\Delta = \Delta_{sc}$. Although the co-presence of the resonant and non-resonant path contributions has been

\begin{table}[h]
\centering
\caption{Experimentally obtained anisotropy parameters $\beta_2$ and $\beta_4$, and the extracted values of $W$ and $\Delta$.}
\begin{tabular}{lllll}
\hline
$h\omega$ (eV) & $\beta_2$ & $\beta_4$ & $W$ & $\Delta$ \\
\hline
20.3 & 1.14 $\pm$ 0.07 & 1.96 $\pm$ 0.03 & 0.561 $\pm$ 0.016 & 1.60 $\pm$ 0.05 \\
21.3 & 0.268 $\pm$ 0.019 & 0.384 $\pm$ 0.063 & 2.39 $\pm$ 0.23 & 1.61 $\pm$ 0.04 \\
23.0 & 0.948 $\pm$ 0.010 & 1.32 $\pm$ 0.15 & 0.977 $\pm$ 0.116 & 1.67 $\pm$ 0.04 \\
24.3 & 2.11 $\pm$ 0.10 & 0.841 $\pm$ 0.006 & 1.43 $\pm$ 0.01 & 2.47 $\pm$ 0.07 \\
\hline
\end{tabular}
\end{table}

Figure 3. (a) Amplitude ratio $W$ and (b) phase-shift difference (relative phase) $\Delta$ extracted from experimental PADs (red diamonds) and calculated from wavefunctions obtained by TDSE simulations (black bullets). Theoretical scattering phase-shift difference $\Delta_{sc}$ [49] is represented by a blue solid line in panel (b).
implicitly used in the coherent control of resonance-enhanced multi-photon processes (see, e.g. [7, 8, 51]), an explicit consideration of intermediate levels other than the resonant one is not necessary in most cases. In this study, on the other hand, the contribution from non-resonant intermediate levels is essential to account for $\Delta_{\alpha}$ [34], which explains why $\Delta_{\alpha}$ is larger for a higher photon energy, i.e. for smaller level spacing.

It may be worth pointing out the similarity between the TPI via a Rydberg manifold and the two-photon above-threshold ionization. In the case of the 24.3 eV excitation, the intense ultrashort EUV pulses used in this study coherently excite several Rydberg states $1snp_n^1P$ with $n = 6–9$. When the spectral width of the pulse contains a sufficient number of Rydberg levels$^{10}$, the Rydberg manifold behaves similar to the continuum near the threshold and both the relative phase $\Delta$ [34] and the TPI yield [50] would smoothly vary when measured by increasing the photon energy across the ionization threshold. It should be noted that the extra phase-shift difference due to free–free transitions plays a significant role in the recently observed time delays in photoemission by attosecond EUV pulses [52–56].

4. Conclusion

We have measured the PAD from the TPI of He by fs EUV FEL pulses provided by the SCSS test accelerator in Japan using a VMI spectrometer. From the anisotropy parameters of the PAD, we extracted phase-shift differences $\Delta$ and amplitude ratios $W$ of the s and d partial waves at four different photon energies ($\hbar\omega = 20.3, 21.3, 23.0$ and 24.3 eV). As a result, we have demonstrated that the co-presence of the resonant and non-resonant path contributions in the TPI by fs pulses causes an additional phase shift in the photoelectron wave packet. The relative contributions from the resonant and non-resonant paths can in principle be controlled by chirping the EUV pulses, which may pave a way to tailor continuum wave packets. Such an experiment will become feasible in the near future at FEL facilities where the pulses can be controlled in the range of a few to a few tens of fs.

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