Two-Photon Rabi Oscillations of Excited He Atoms in Ultrafast Strong Laser Field Ionization

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Abstract: Intensity dependence of polarized He (2^1P) atoms in intense NIR laser fields are investigated by single-shot photoelectron spectroscopy, revealing two-photon Rabi oscillations between the initial 1s2p and 1s*n*f (*n* = 5,6) Rydberg states.

OCIS codes: (020.0020) Atomic and molecular physics; (300.6420) Spectroscopy, nonlinear

1. Introduction

Ionization is one of the most important elementary processes of atoms in intense laser fields. For the laser field intensity of ~10^{13} W/cm^2, the ionization process has been understood by multiphoton absorption. The ionization rate becomes significantly enhanced when several Rydberg states are energetically shifted into resonance by interactions with the laser field (Freeman resonance) [1,2]. Atoms prepared in a polarized state allow us to identify ionization pathways associated with the specific magnetic sublevels, thereby giving more insight into the ionization processes. In the present study, we investigate ionization dynamics of polarized He in the 2^1P state in intense NIR laser fields (~800 nm) by single-shot photoelectron spectroscopy

2. Experiment

Horizontally polarized free-electron-laser (FEL) pulses at 58.4 nm obtained from the SCSS Test Accelerator at RIKEN HARIMA institute were used for the preparation of polarized He atoms in the 2^1P state. The FEL pulses were introduced into the interaction region by using a pair of elliptical and cylindrical mirrors and the beam size (2ω) in the focal spot was estimated to be 40 μm (ω: beam waist). Linearly polarized ultrashort NIR laser pulses (795 nm, < 300 μJ, ~50 fs) from a regenerative amplifier Ti:Sapphire laser system were focused to the polarized He atoms in a long focusing geometry. The resultant beam size (2ω = 160 μm) was sufficiently large compared to that of FEL, thus making it possible to ionize the polarized He atoms placed in the central part of the NIR laser field (Fig. 1(a)). In order to monitor the 2^1P-state population, which varies shot-by-shot due to fluctuation of FEL pulses, ultrashort UV laser pulses (250 nm) were employed 10 ps in advance with respect to the NIR laser pulse. Photoelectron yield by the single-photon ionization from the 2^1P state was used to normalize the population. The time-sequence of FEL, UV and NIR laser pulses is summarized in Fig. 1(b).

Photoelectrons produced in the interaction region were detected by using a magnetic bottle type photoelectron spectrometer. The typical detection size of the spectrometer is 1×1×1 mm^3 in front of the tip of the cone-shape permanent magnet. This confined detection size against the confocal beam parameter (~ 20 mm) of the NIR laser pulse allows us to observe multiphoton processes by the NIR laser pulse with an almost single filed intensity, thereby minimizing the effect of the spatially-varied intensity distribution (Fig. 1(a)). The energy resolution of the spectrometer was estimated to be $E/\Delta E$ = 20 for photoelectron energies E < 100 eV under present experimental conditions.

3. Results and discussion

Figure 1(c) shows a photoelectron spectrum of polarized He (2^1P) atoms in intense NIR laser fields (3.7 TW/cm^2) in the perpendicular configuration. Photoelectron peaks seen at ~1 and ~2.6 eV can be assigned to electrons formed by the three-photon ionization and its above threshold ionization, respectively, while a weak peak observed at 1.58 eV is due to those produced by single-photon ionization by UV laser pulses from the 2^1P state. Fine structure peaks observed at 0.97 and 1.14 eV show almost no energy shift by the increase in the NIR laser field intensity, indicating that the ionization proceeds in resonant with intermediate states. From the energy conservation, these intermediate states are assigned to the 1s5f and 1s6f Rydberg states whose energy levels are located about 3.1 eV(= $2h\nu_{NIR}$) from the initial 2^1P state.

Figure 1(d) shows a peak height of the 1s5f (blue circles) and 1s6f (red circles) peaks as a function of NIR laser field intensity I_{NIR} in the range of 0 - 7.5 TW/cm^2 in the perpendicular configuration. The 1s6f peak increases with modulations as I_{NIR} increases while the 1s5f peak exhibits less clear oscillations. For the 1s6f peak, the peak height reaches the maxima at ~ 1.5 and ~ 3.5 TW/cm^2 as well as the minima at ~ 2.2 and ~ 5.5 TW/cm^2, showing the increasing oscillation period.

To understand the experimental behavior, simulation based on time-dependent Schrödinger equation for He (2^{1}P) atoms in the presence of NIR laser fields is carried out. The simulated I_{NIR} dependence is shown for the 1s5f (blue curve) and 1s6f (red curve) peaks in Fig. 1(d), which is in a good agreement with the experiment. It turns out from the simulation that there are coherent population transfers (Rabi oscillation) between the 1s*n*f (n = 5,6) Rydberg and 2^{1}P states by the NIR laser field through two-photon process, which accordingly leads to modulations of the photoelectron yield. The different behavior between 1s5f and 1s6f peaks is attributed to the different detuning from resonance in two-photon level, which modulates the two-photon Rabi oscillation frequency. The present study indicates that ionization from high Rydberg states can be suppressed by multiphoton coherent interactions with lower-lying states.

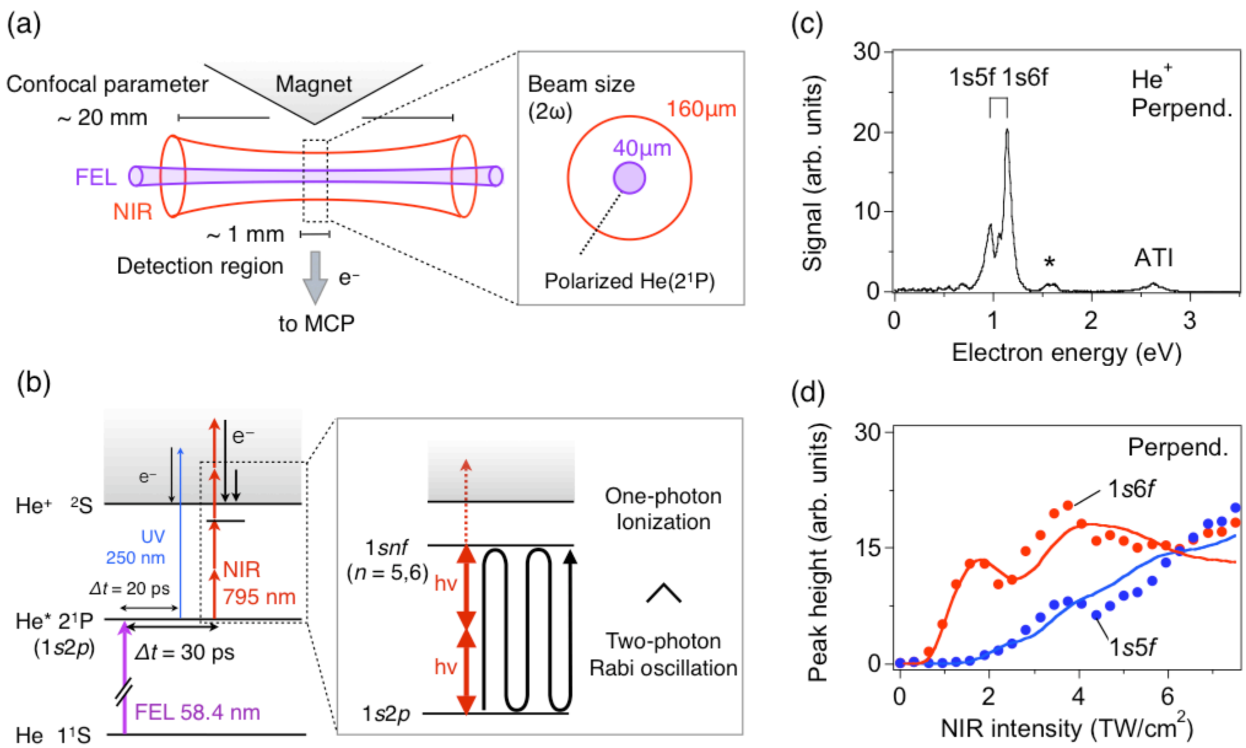

Fig. 1. (a) Schematic diagram of the spatial arrangement of FEL and NIR laser pulses in the interaction region. (b) Relevant energy levels of He atoms and the time sequence of FEL, UV and NIR laser pulses. (inset) Schematic diagram of the two-photon Rabi oscillation of polarised He (2^{1}P) atoms in intense NIR laser fields. (c) Photoelectron spectrum of polarized He (2^{1}P) atoms by NIR laser pulses with an intensity of 3.7 TW/cm^2. Peaks around 1 and 2.6 eV are due to electrons produced by three-photon ionization and its above threshold ionization (ATI) while the peak marked by the asterisk is attributed to electrons formed via one-photon ionization by UV laser pulse, which is used for monitoring the population of the 2^{1}P state. (d) Peak height of the 1s6f (red) and 1s5f (blue) peaks as a function of NIR laser field intensity in the range of 0-7.5 TW/cm^2 in the perpendicular configuration for experiment (circles) and simulation (curves).

4. Summary

We have investigated coherent resonant multiphoton processes in strong field ionization of polarized He (2^{1}P) atoms. The spatially aligned excited-state distribution makes it possible to selectively probe the coherent process by using the linear polarized light with a single intensity achieved in a long-focusing geometry.

We are grateful to the SCSS Test Accelerator Operation Group at RIKEN for their continuous support.

References

[1] R.P. Freeman et al., "Above-threshold ionization with subpicosecond laser pulses," Phys. Rev. Lett. **59,** 1092-1095 (1987).
[2] H. Helm and M. J. Dyer, "Resonant and nonresonant multiphoton ionization of helium," Phys. Rev. A **49,** 2726 -2733 (1994).