Coherent control of XUV absorption and photoemission by the simultaneous irradiation of ultrashort XUV and laser pulses

Kenichi Ishikawa and Katsumi Midorikawa
Laser Technology Laboratory, RIKEN (The Institute of Physical and Chemical Research), Hirosawa 2-1, Wako-shi, Saitama 351-0198, Japan
ishiken@postman.riken.go.jp

Abstract: We numerically show that the absorption of an XUV pulse by a hydrogen atom is transiently reduced by a femtosecond laser pulse and that the enhanced photoemission depends on the timing of the two pulses.

OCIS codes: (020.6580) Stark effect; (190.4160) Multiharmonic generation

1 Introduction

The recent progress in obtaining light pulses with a shorter duration and wavelength has enabled us to generate laser pulses with a duration of a few fs, and the high harmonic generation (HHG) can now produce intense coherent extreme ultraviolet (XUV) and soft x-ray pulses, whose duration may be even shorter.

A laser field increases the ionization threshold and shifts the electronic levels of atoms through the ponderomotive and ac-Stark effect, respectively [1]. The ponderomotive shift is directly observed in the above-threshold ionization electron energy spectra [2]. These shifts also manifest themselves as changes in the XUV and soft x-ray absorption. For the case of relatively long, low-intensity laser pulses, the ac-Stark shift was observed, e.g., for Xe Rydberg levels [3], and the ponderomotive-shift-induced hindering of x-ray absorption was discussed by Kálmán [4]. There exists, however, few literature [5] on the change in XUV absorption for the case of femtosecond, intense laser pulses, which is expected to be qualitatively different from the low-intensity case, since such laser pulses not only shift the levels and the ionization threshold but also may induce various nonlinear effects such as HHG, laser-assisted excitation and ionization.

In the present study, we numerically investigate the response of a hydrogen atom under the simultaneous irradiation of ultrashort intense laser and resonant XUV pulses. Our results show that the XUV absorption can be controlled in a very short timescale by a laser pulse. We also find that the photoemission including HHG is enhanced by the XUV pulse and that its yield depends much on the overlap of the two pulses.

2 Model

To study the interaction of a hydrogen atom initially in the ground state with an XUV and a Ti:sapphire laser ($\hbar\omega_L = 1.55\,\mathrm{eV}$) pulses, we numerically solve the time-dependent Schrödinger equation (TDSE):

$$i\frac{\partial\Phi(r,t)}{\partial t} = \left[-\frac{1}{2}\nabla^2 + V_{\mathrm{at}} + H_L(t) + H_X(t)\right]\Phi(r,t), \tag{1}$$

where V_{at} is the atomic potential, which is Coulombic for a hydrogen atom, and the atom-field interaction Hamiltonians for the laser and the XUV pulses are,

$$H_L(t) = -zF_L(t)\sin(\omega_L t) \quad \text{and} \quad H_X(t) = -zF_X(t)\sin(\omega_X t), \tag{2}$$

respectively, with F_L and F_X being the envelope of the pulses (the atomic units are used throughout). Here we have assumed that both the laser and the XUV fields are linearly polarized in the z-direction. Equation (1) is integrated using the alternating direction implicit (Peaceman-Rachford) method [6]. We can evaluate the number of absorbed XUV photons by calculating the decrease of the $1s$ population, since under the conditions considered in the present study the laser pulse alone hardly excites nor ionize a ground-state hydrogen atom, and the multi-photon absorption of XUV pulses is negligible.

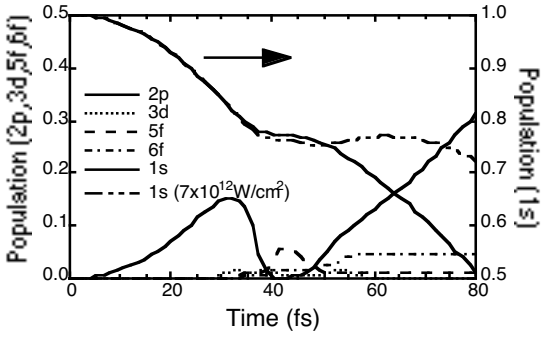

Fig. 1: Evolution of the population of several bound levels when a laser pulse is applied to a hydrogen atom during a resonant XUV pulse irradiation.

3 Control of the XUV absorption by a laser pulse

Figure 1 shows how the population of several levels evolves when a hydrogen atom is exposed to an XUV field and a shorter laser pulse is injected during the XUV irradiation. The XUV pulse shape is trapezoidal, with linear turn-on and -off, each with a duration of one laser period ($T_L = 2.6\,\mathrm{fs}$), the total duration is $32T_L$, and the peak intensity is $3 \times 10^{10}\,\mathrm{W/cm^2}$. The XUV photon energy is $\hbar\omega_X = 10.2\,\mathrm{eV}$, resonant with the $1s - 2p$ transition. The laser pulse shape is gaussian centered at $t = 16T_L$ with a duration (full width at half maximum) of $4T_L$ and a peak intensity of $10^{13}\,\mathrm{W/cm^2}$. When the laser field is sufficiently weak, the atom behaves as a two-level system and undergoes Rabi oscillation: during the first ($t < 25\,\mathrm{fs}$) and the final ($t > 60\,\mathrm{fs}$) stage of the interaction, the XUV absorption leads to the decrease of the $1s$ population and the increase of the $2p$ population, while the population of the other levels remain unchanged. In the intermediate stage, the depopulation of the $1s$ level slows down with the increase of the laser intensity, and its population is nearly constant for a few fs around $t = 42\,\mathrm{fs}$. This indicates that the XUV absorption is effectively suppressed for a very short time by the laser-induced shift and broadening of the $2p$ level. We can see that unlike the ground level, the $2p$ population is not frozen even during the laser pulse irradiation: it first decreases nearly down to zero and then increases again. We can also see that the laser pulse induces transitions between different excited levels. Thus, in this stage the atom cannot be viewed simply as a detuned two-level system; its response is highly nonlinear, involving many different levels and the continuum.

We have found that the atomic response is approximately linear with respect to XUV intensity, while it exhibits a complicated dependence on laser intensity, reflecting again a highly nonlinear nature of the interaction. In Fig. 1 we have also plotted the $1s$ population for the laser peak intensity of $7 \times 10^{12}\,\mathrm{W/cm^2}$. Also in this case, the atom undergoes Rabi oscillation after the laser field has been virtually turned off. In contrast to the previous case, however, the $1s$ population first increases (the XUV light is amplified). Thus, by carefully choosing the parameters of the ultrashort laser pulse, we can control the XUV absorption and manipulate the atomic state within the interval of a few fs.

We have also considered the situation where the XUV photon energy is slightly above the ionization potential and found that also in this case the XUV absorption can be reduced by a laser-induced ponderomotive upshift of the ionization potential, though the effect is smaller than in Fig. 1.

4 Photoemission yield

In the present section we discuss how the simultaneous application of a resonant XUV and an intense laser pulses affects the harmonic generation (more precisely, photoemission at $\omega = \omega_X + n\omega_L$, where n is an odd integer). In Fig. 2 we show the photoemission spectra for different values of XUV photon energy $\hbar\omega_X$, one (10.2 eV) of which is resonant with the $1s - 2p$ transition, along with the results for the cases where only laser is applied. Both pulses consist of a linear ramp for 5 laser cycles followed by 15 additional cycles at a constant intensity indicated in the figure. We can see that photoemission is greatly enhanced when the

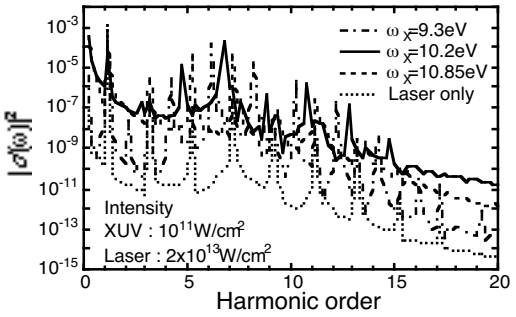

Fig. 2: Photoemission spectra under the simultaneous laser and XUV irradiation for different values of XUV photon energy $\hbar\omega_X$.

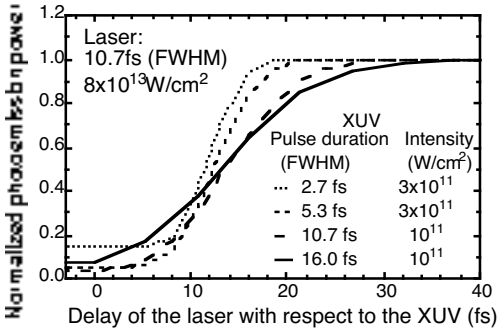

Fig. 3: Photoemission power (normalized to unity at $t_d = 40\,\mathrm{fs}$) at $\omega > 7.5\omega_L$ vs. delay t_d of the laser pulse with respect to the XUV pulse.

XUV pulse is applied. It should be noted that the yield does not depend much on $\hbar\omega_X$ except the shift of the peak positions, indicating that the XUV pulse needs not be resonant with an electronic transition for this enhancement, since the resonant XUV absorption is suppressed by the laser field. Such an enhancement may find an application as an efficient, tunable XUV and soft x-ray source.

Let us now imagine that the laser pulse is delayed with respect to a resonant XUV pulse ($\hbar\omega_X = 10.2\,\mathrm{eV}$). The photoemission in such a case is expected to be further enhanced since the $2p$ level is effectively populated by the XUV pulse. Figure 3 shows the power of the photoemission at $\omega > 7.5\omega_L$ as a function of the delay. Both pulse shapes are gaussian with a duration and intensity indicated in the figure. As expected, the photoemission yield is much higher when the XUV pulse precedes the laser than in the case of no delay. In the intermediate case the yield varies fast, depending on the overlap of the two pulses. From Fig. 3 we can see that the extent of this range is approximately proportional to the XUV pulse duration. This holds true even when the XUV pulse is much shorter than the laser pulse, and may provide a method to measure pulse durations in the XUV and soft x-ray domain with fs resolution.

5 Conclusions

We have studied the interaction of a hydrogen atom with the simultaneous irradiation of ultrashort laser and resonant XUV pulses by solving TDSE. The XUV absorption can be suppressed in a fs timescale by a laser pulse. The laser field not only decouples the atom from the XUV pulse but also induces the transition between excited levels and ionization. The photoemission is enhanced when an additional XUV pulse is applied, and dramatically changes when we vary the delay of the laser pulse with respect to the XUV pulse by the amount corresponding to the XUV pulse duration.

References

1. A. D. Bandrauk, *Molecules in laser fields* (Dekker, 1993).
2. R. R. Freeman *et al.*, "Above-threshold ionization with subpicosecond laser pulses," Phys. Rev. Lett. **59**, 1092 (1987).
3. T. R. O'Brian *et al.*, "Verification of the ponderomotive approximation for the ac Stark shift in Xe Rydberg levels," Phys. Rev. A **49**, R649 (1994).
4. P. Kálmán, "Hindering effect of an intense laser field in near-absorption-edge laser-assisted x-ray absorption," Phys. Rev. A **39**, 3200 (1989).
5. L. Chen, "The possibility of x-ray pulse shortening through laser induced ac Stark shift in atoms," J. Appl. Phys. **73**, 2629 (1992).
6. K. C. Kulander, K. J. Schafer, and J. L. Krause, "Time-dependent studies of multiphoton processes" in *Atoms in intense laser fields*, M. Gavrila, ed. (Academic, New York, 1992) pp. 247–300.