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Coherent control of extreme uv absorption and photoemission by the simultaneous irradiation of ultrashort extreme uv 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
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We present a theoretical study on the response of a hydrogen atom to the simultaneous irradiation of ultrashort extreme ultraviolet (xuv) and laser pulses. The calculation is based on the time-dependent Schrödinger equation. The xuv photon energy is chosen to induce the $1s$-$2p$ transition. Our results indicate that we can transiently reduce the xuv absorption and change the atomic state within a few femtoseconds by injecting a femtosecond laser pulse. The laser-induced photoemission is affected by the addition of an xuv pulse and its yield depends much on the temporal overlap of the two pulses.

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The recent progress in obtaining coherent light pulses with a shorter duration and wavelength has enabled us to generate laser pulses with a duration of a few femtoseconds [1], and the high-harmonic-generation (HHG) technique can now produce quite intense coherent extreme ultraviolet (xuv) and soft x-ray pulses [2]. In the HHG medium, the fundamental laser beam and generated xuv pulse coexist. It is expected that they strongly influence each other, especially, when the xuv pulse lies at or near an absorption line of the medium. However, studies of the effects of their interplay on, e.g., xuv absorption and harmonic yield have been scarce. When an atom is placed in a laser field, the electronic levels are shifted through the ac-Stark effect [3–12], and the ionization threshold is increased by the ponderomotive effect [13–15]. These effects are not only observed as a peak shift and a fine structure in photoelectron energy spectra [3–7,13–15], but should also manifest themselves as a change in xuv and soft x-ray absorption. Although the suppression of xuv and x-ray absorption by the laser-induced ac-Stark [16] and ponderomotive effect [17] was discussed for relatively long, low-intensity laser pulses, there exists no time-dependent study valid for ultrashort, intense pulses, which may be qualitatively different from the low-intensity case. It is also expected that the interplay of the xuv and laser pulses affects the photoemission from the atom including the HHG. Although its impact on the photoemission yield is a great concern to the HHG experiments, it has not been explicitly studied so far, while high-order frequency mixing using two laser pulses was investigated [18]. In this Rapid Communication, we investigate the interaction of a hydrogen atom with the simultaneously applied ultrashort intense laser and xuv pulses by numerically integrating the time-dependent Schrödinger equation. Our results show that the atom exhibits a surprisingly rich response. The resonant xuv absorption can be suppressed during a very short time of laser irradiation. The latter also induces transitions between excited levels and ionization, which results in a jump of the atomic state. The laser-induced photoemission from the atom is significantly affected by the addition of the xuv pulse compared with the case where only the laser field is applied, and its yield depends on the delay between the two pulses. The xuv pulse considered in the present study is not necessarily a harmonic of the laser pulse but includes a wide range of coherent xuv radiation such as x-ray laser and synchrotron radiation.

To study the interaction of a hydrogen atom initially in the ground state with xuv and Ti:sapphire laser ($\hbar\omega_L = 1.55$ eV) pulses, we solve the TDSE,

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

where V_{at} is the atomic potential, which is Coulombic for a hydrogen atom, and the atom-field interaction Hamiltonians for the laser, H_L, and the xuv pulse, H_X, are

$$H_{L,X}(t) = -zF_{L,X}(t)\sin(\omega_{L,X}t), \quad (2)$$

with $F_{L,X}$ being the envelope of each pulse. Here we have assumed that both fields are linearly polarized in the z direction. The time scale considered in the present study, at most 120 fs, is much shorter than the lifetime of excited levels of H (e.g., that of the $2p$ state is 1.6 ns [19]). and the dephasing time due to collisions for the case of atomic gases, which is typically longer than several tens of picoseconds. Thus, we can safely assume that the interaction of the hydrogen atom with xuv and laser pulses is coherent and is described by Eq. (1). Equation (1) is numerically integrated using the alternating-direction implicit (Peaceman-Rachford) method [20].

We first consider the laser-induced change in xuv absorption. We can evaluate the number of absorbed xuv photons by calculating the decrease of the $1s$ population, since under the conditions of the present study the laser alone hardly excites nor ionizes the ground-state hydrogen atom and the multiphoton xuv absorption is negligible. Figure 1 shows the dependence of the xuv absorption spectrum on laser peak intensity I_L. The shape of each pulse is Gaussian centered at $t = 21.3$ fs with a duration $\tau_{X,L}$ (full width at half maximum) of 10.7 fs, and the xuv peak intensity I_X is 3×10^{10} W/cm^2. In the laser-free case (solid line), the spectrum has a peak

*Electronic address: ishiken@postman.riken.go.jp

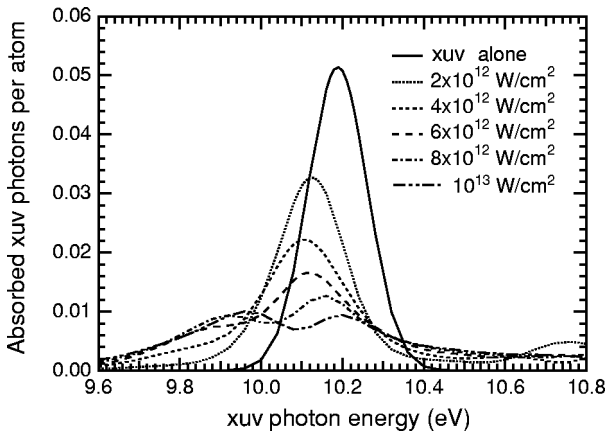

FIG. 1. xuv absorption spectra of a hydrogen atom in the presence of a laser field ($\hbar\omega_L=1.55$ eV) with different values of peak intensity. Both xuv and laser pulses have a Gaussian pulse shape with a duration of 10.7 fs, and the xuv peak intensity is 3×10^{10} W/cm^2.

corresponding to the $1s$-$2p$ transition (10.2 eV) and a width determined by the pulse duration through the uncertainty relation. In the presence of the laser pulse, the spectrum is shifted as well as broadened due to the ac-Stark effect.

Figure 2 presents how the population of several bound levels of a hydrogen atom evolves when an ultrashort laser pulse is injected during the xuv irradiation. The xuv pulse has a constant intensity $I_X=3\times10^{10}$ W/cm^2 with a linear turn-on whose duration is one laser period. The xuv photon energy $\hbar\omega_X$ is 10.2 eV, which lies near the lower limit of the xuv range. The laser pulse shape is Gaussian centered at 42.7 fs with $\tau_L=10.7$ fs and $I_L=10^{13}$ W/cm^2. When the laser field is sufficiently weak, the atom behaves as a two-level system and undergoes Rabi oscillations: during the first ($t<25$ fs) and the final ($t>60$ 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 populations of the other levels remain unchanged. In the intermediate stage (25 fs$<t<$60 fs), the depopulation of the $1s$ level slows down with the increase of the laser intensity, and its population is nearly constant for a few femtoseconds around $t=42.7$ fs, indicating that the xuv absorption is almost completely suppressed. Another feature of Fig. 2 is that the $2p$ population is not frozen even during the laser-pulse irradiation, and that the laser pulse induces transitions between excited levels. Thus, in this stage the atom cannot simply be viewed as a detuned two-level system; its response is highly nonlinear, involving many levels and the continuum.

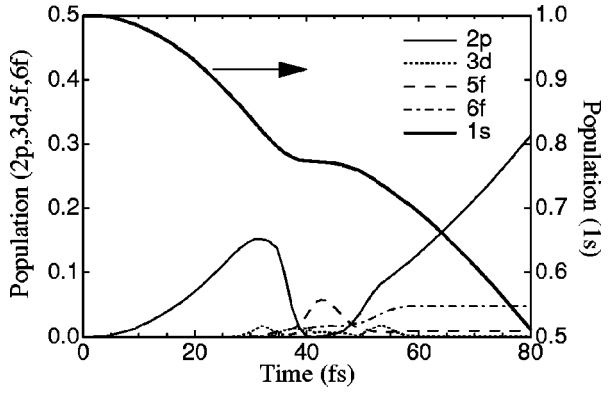

FIG. 2. Evolution of the population of several levels of H when a laser pulse ($\tau_L=10.7$ fs, $I_L=10^{13}$ W/cm^2) centered at $t=42.7$ fs is injected during a resonant xuv pulse ($\hbar\omega_X=10.2$ eV, $I_X=3\times10^{10}$ W/cm^2) irradiation.

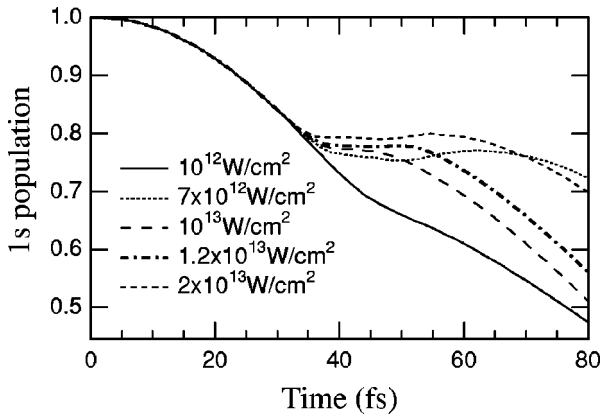

FIG. 3. Evolution of the $1s$ population of H when a laser pulse ($\tau_L=10.7$ fs) with different values of peak intensity is injected during a resonant xuv pulse ($\hbar\omega_X=10.2$ eV, $I_X=3\times10^{10}$ W/cm^2) irradiation.

In Fig. 3 we plot the evolution of the $1s$ population for different values of the laser peak intensity I_L. Each pulse has the same shape as in Fig. 2 and $I_X=3\times10^{10}$ W/cm^2. We can see that the atomic response exhibits a complicated dependence on I_L, reflecting a highly nonlinear nature of the interaction. It should be noted that for certain values of laser peak intensity, after the laser pulse irradiation the $1s$ population first increases (the xuv pulse is amplified) in contrast to the case in Fig. 2. As the laser intensity gets higher, the reduction of xuv absorption begins earlier and becomes stronger. The behavior of a two-level system interacting with a coherent xuv pulse can be described by the optical Bloch equations [21]. In this model we begin with expressing the wave function $\Phi(\mathbf{r},t)$ as a linear combination $\Phi(\mathbf{r},t)=c_1(t)\Phi_1(\mathbf{r})+c_2(t)\Phi_2(\mathbf{r})$ of the eigenfunctions $\Phi_{1,2}$ of the two levels, and then introduce a vector $\mathbf{R}$ with three components $(R_1,R_2,R_3)=(\rho_{21}+\rho_{12},i\rho_{21}-i\rho_{12},\rho_{22}-\rho_{11})$, where $\rho_{21}=\rho_{12}^*=c_2c_1^*\exp(i\omega_X t)$ and $\rho_{ii}=|c_i|^2$ $(i=1,2)$. Then the time evolution of $\mathbf{R}$ is given by [21]

$$\dot{R}_1=-\Delta R_2,\quad \dot{R}_2=\Delta R_1+pR_3,\quad \dot{R}_3=-pR_2, \qquad (3)$$

where $\Delta=\omega_{21}-\omega_X$ with ω_{21} being the transition energy and p denotes the properly normalized transition-dipole moment. Equation (3) has a general solution $R_3=A\sin(\Omega t+\phi)+C$, where $\Omega=\sqrt{\Delta^2+p^2}$ is the Rabi frequency, and A,ϕ, and C are real constants. As can be seen from Fig. 3, one of the roles of the laser pulse is to change the values of the constants (A,ϕ,C), and this jump depends on I_L.

In Fig. 4 we show an example of the temporal evolution of the $1s$ and $2p$ population in case the xuv photon energy $\hbar\omega_X=10.3$ eV is slightly detuned from the $1s$-$2p$ transi-

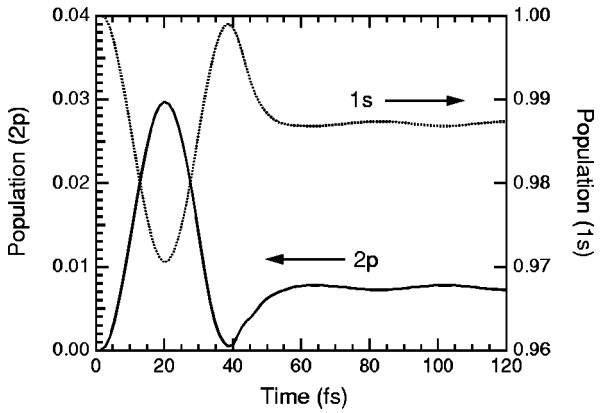

FIG. 4. Evolution of the $1s$ (right axis) and $2p$ (left axis) population of H when a laser pulse ($\tau_L=10.7$ fs, $I_L=2\times10^{12}$ W/cm^2) is injected during a slightly detuned xuv pulse ($\hbar\omega_X=10.3$ eV, $I_X=3\times10^{10}$ W/cm^2) irradiation.

tion. Each pulse is of the same shape as in Fig. 2, with $I_X=3\times10^{10}$ W/cm^2 and $I_L=2\times10^{12}$ W/cm^2. It is striking that the population of $1s$ and $2p$ remains nearly constant even after the laser pulse has been turned off. It follows from Eq. (3) that the population difference R_3 is constant if **R** initially satisfies

$$R_2=0 \quad \text{and} \quad \Delta R_1+pR_3=0. \tag{4}$$

The atomic state at $t>60$ fs in Fig. 4 corresponds to this situation. It should be noted that Eq. (4) would never be satisfied without the laser field since the atom is initially in the ground state ($R_1=R_2=0, R_3=-1$). It is the laser-pulse irradiation that enables the jump of the Bloch vector **R** into the state satisfying Eq. (4). This observation, along with the results shown in Fig. 3, suggests the possibility to manipulate the atomic state within the interval of a few femtoseconds by carefully choosing the parameters of the ultrashort laser pulse.

We briefly mention the case where $\hbar\omega_X$ is slightly larger than the ionization potential. In this case one would expect that the laser-induced ponderomotive upshift of the ionization potential leads to the reduction of ionization and xuv absorption [17]. We have, however, found that the reduction is much smaller than in the case of resonant xuv pulses. This is because the xuv pulse is absorbed through laser-assisted ionization [22] as well as resonantly by high-lying levels.

The HHG from atomic gases irradiated by an intense femtosecond laser is one of the most promising ways to produce coherent xuv pulses. Let us now discuss how the application of an additional xuv pulse affects the HHG, or more generally, the photoemission from a hydrogen atom. In Fig. 5 we show photoemission spectra for different values of $\hbar\omega_X$ along with the result for the case where only the laser is applied. Both pulses consist of a linear ramp for five laser cycles followed by 15 additional cycles at a constant intensity $I_X=3\times10^{10}$ W/cm^2 and $I_L=2\times10^{13}$ W/cm^2. The spectrum is obtained from the Fourier transform of the dipole acceleration during the last five cycles. We can see from this figure that, in addition to the usual harmonic peaks, the photoemission spectra have peaks at a photon energy $\hbar\omega=\hbar\omega_X+n\hbar\omega_L$ with n being an even integer, and that the latter peaks are higher than the harmonic peaks. In the case where the xuv pulse is the seventh harmonic of the laser ($\hbar\omega_X=10.85$ eV), the HHG itself is enhanced. Another feature that can be seen in Fig. 5 is that the photoemission yield does not depend much on $\hbar\omega_X$ except the shift of the peak positions. This is because the ac-Stark effect not only shifts but also broadens the $2p$ level as we have seen in Fig. 1. As a result, the atomic response to the xuv pulse is strong in a relatively wide spectral range. This indicates that the xuv pulse is not required to be resonant with an electronic transition for the photoemission enhancement. These observations imply that the emission in Fig. 5 can be viewed as a high-order frequency mixing [18] enhanced by the resonance induced by the ac-Stark broadening. We expect a similar effect also in the case where the xuv or soft x-ray pulse is nearly resonant with an inner-shell electronic transition of more complicated atoms such as rare gases. We can also look

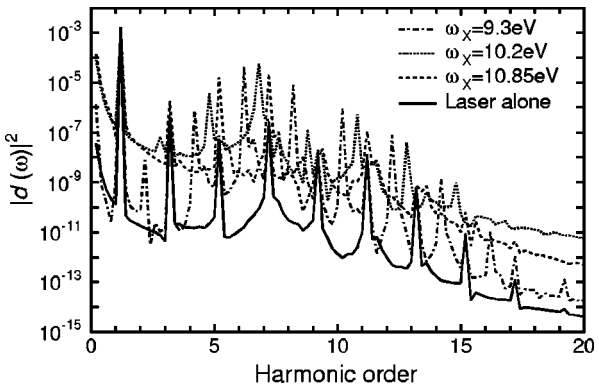

FIG. 5. Photoemission spectra from H under the simultaneous laser ($I_L=2\times10^{13}$ W/cm^2) and xuv ($I_X=3\times10^{10}$ W/cm^2) irradiation for different values of xuv photon energy $\hbar\omega_X$. The pulse shapes are specified in the text.

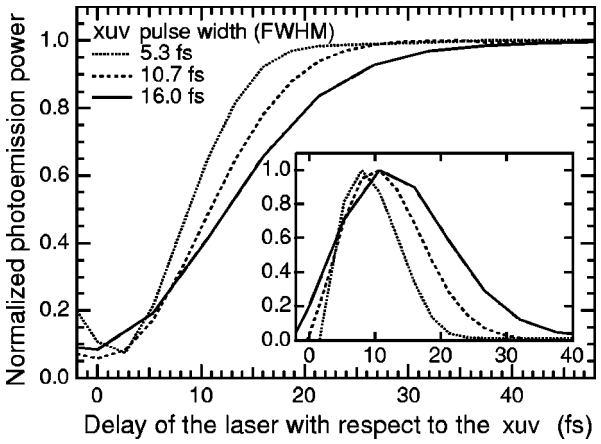

FIG. 6. Total power of photoemission from H at $\hbar\omega>7.5\hbar\omega_L$ (normalized to unity at $\Delta t=50$ fs) vs delay Δt of the laser pulse ($\tau_L=10.7$ fs, $I_L=2\times10^{13}$ W/cm^2) with respect to the xuv pulse ($I_X=3\times10^{10}$ W/cm^2). Inset: the derivative of the total photoemission power with respect to Δt (normalized to unity at the peak of each curve).

at Fig. 5 from another point of view as follows. Let us imagine that the xuv pulse is generated by an x-ray laser, which is, in general, intense but not tunable. Our results suggest that by irradiating an atomic gas simultaneously with this xuv pulse and a laser pulse, we can obtain coherent xuv pulses whose wavelength is different from that of the original pulse. This may find an application as an efficient, tunable xuv and soft x-ray source.

So far we have considered the situation where both pulses are applied with no delay. When the xuv pulse is exactly resonant with an electronic transition, its interaction with the atom is much stronger in the absence of the laser field than in its presence. It is, therefore, expected that the photoemission yield is even higher if the resonant xuv pulse ($\hbar\omega_X = 10.2$ eV) is applied *before* the laser pulse, preparing a coherent superposition of 1*s* and 2*p* states. This situation is similar to that in Ref. [23]. In fact, as can be seen from Fig. 6, which shows the total power of photoemission at $\hbar\omega > 7.5\hbar\omega_L$ as a function of the interval Δt between the two Gaussian pulses, the photoemission yield is much higher when the xuv pulse ends before the turn-on of the laser pulse than when both are applied simultaneously ($\Delta t = 0$). In the intermediate case, where the two pulses partially overlap, the yield varies fast, depending on the overlap. In the inset of Fig. 6 we show the derivative of the photoemission power with respect to Δt. The width of the curves is approximately proportional to τ_X, and this may provide a method to measure pulse durations in the xuv and soft x-ray domain with femtosecond resolution. It is worth noting that the approximate linear relation holds even when the xuv pulse is shorter than the laser pulse, since the photoemission is lowered as soon as the leading edge of the laser pulse overlaps the xuv pulse.

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