Effect of partial temporal coherence of XUV pulses in IR-laser-assisted photoionization

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The effect of partial temporal coherence of free-electron laser (FEL) pulses in IR-laser-assisted short-pulse photoionization of atoms is theoretically analyzed. In a typical two-color photoionization experiment, when an atom is ionized by extreme ultraviolet photons in the presence of a strong IR laser field, several sidebands are produced in the electron spectrum at both sides of the photoline. The stochastic nature of the FEL radiation leads to a broadening of the sidebands. When the coherence time is short, shorter than the inverse frequency of the IR field, the sidebands cannot be observed. However, in angle-resolved experiments a certain structure in the spectrum can be seen as a remnant of the sideband gross structure.

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I. INTRODUCTION

Free-electron lasers (FELs) operating in the extreme ultraviolet (XUV) and x-ray region boosted tremendously the physics of the interaction of short-wavelength radiation with matter. Extremely high brilliance and ultrashort pulses of FELs make it possible to explore new frontiers in atomic and molecular physics, material science, and molecular biology. Especially promising is the emerging possibility to study the dynamics of ultrafast electronic processes using pump-probe excitation schemes whereby two ultrashort pulses with controlled temporal delays impinge on a sample. The time resolution of this scheme is determined by the precise knowledge of the duration of the pulses as well as their synchronization. Recently many efforts have been devoted to the investigation of the temporal properties of the FEL radiation, such as pulse duration [1,2], temporal profile [3], pulse arrival time [4–7], and temporal coherence [8–11].

The interpretation of experimental results obtained with FEL radiation and the extraction of fundamental properties have encountered certain general difficulties. Almost all existing FELs are based on the self-amplified spontaneous emission (SASE) principle [12]. This means that the radiation emitted by electron bunches is chaotic and characterized by irreproducible pulses. Their intensity, and temporal and spectral shapes vary significantly from shot to shot. Each individual pulse may be considered as a random number of intensity spikes (coherent regions) of random amplitudes [3,13]. Such a structure can strongly influence the investigation of any photoinduced processes [14] and it should be taken into account for the interpretation of the experimental results [15,16]. Whenever in principle it is possible to experimentally determine the parameters of the pulses on a shot-to-shot basis and later on to sort the experimental data according to the measured parameters, such an approach is extremely difficult, and realized only in rare cases [17]. In the majority of experiments, the measured quantity is the average over an ensemble of pulses.

One of the methods used in studying the temporal properties of the FEL radiation and the fundamental interaction of this radiation with target atoms is the laser-assisted photoionization of atoms or two-color short-pulse experiments [18]. In these experiments an atom is ionized by the XUV pulse in the presence of the synchronized strong infrared (IR) laser field. Contrary to the FEL, the optical laser pulses are well reproducible and their parameters are rather stable. Therefore the IR laser field can serve as a reference for studies of the FEL parameters. The emitted photoelectron interacting with the optical field can absorb or emit additional photons. Thus in the photoelectron spectrum the sidebands are formed at both sides of the photoline, separated by an energy interval equal to the optical laser quantum [19,20]. The existence of the sidebands is a pure interference effect, therefore in photoionization experiments the coherence properties of the exciting FEL photons can noticeably influence the results of the measurements. Thus the study of laser-assisted photoionization can serve as a supplementary method of controlling the coherence properties of the FEL pulses.

In this paper we investigate how the temporal coherence of the XUV pulse affects the properties of the sidebands. To this end, we use a simple model of the two-color atomic photoionization developed earlier [21]. The stochastic properties of the FEL radiation are described using the statistical approach (see, e.g., Ref. [14]).

II. THEORY AND CALCULATIONS

We consider photoionization of an atom by an XUV femtosecond pulse in the presence of a moderately strong (10^{12}–10^{13} W/cm^2) IR laser field. We suppose that photoionization occurs far from threshold so that the photoelectron has sufficiently large energy (several tens of eV). In this case, using the strong-field approximation (SFA) [22], one can present the amplitude of photoionization in the following form [21] (atomic units are used throughout unless otherwise indicated):

$$\mathcal{A}_{\vec{k}} = -i \int_{-\infty}^{\infty} dt\, \tilde{\mathcal{E}}_X(t) \langle \Psi_f \psi_{\vec{k}}(t) \mid \hat{D} \mid \Psi_0 \rangle e^{i(E_b - \omega_X)t}, \quad (1)$$

where $\tilde{\mathcal{E}}_X(t)$ is the envelope of the XUV pulse, ω_X is its mean frequency, E_b is the binding energy (positive) of the electron,

Ψ_0 and Ψ_f are the initial atomic and final ionic wave functions, respectively, and $\hat{D}$ is the dipole operator. In the following we ignore the influence of the IR field on the bound atomic and ionic states, which is a sufficiently good approximation for the moderate IR fields considered here. The wave function $\psi_{\vec{k}}(t)$ in Eq. (1) describes the "dressed" photoelectron in the IR laser field, which is characterized by the final (asymptotic) momentum $\vec{k}$. Within the SFA, the wave function of the photoelectron is represented by the nonrelativistic Volkov wave function [23]

$$\psi_{\vec{k}} = \exp\{i[\vec{k} - \vec{A}_L(t)]\vec{r} - i\Phi(\vec{k},t)\}. \tag{2}$$

Here

$$\Phi(\vec{k},t) = \frac{1}{2}\int_t^\infty dt'[\vec{k} - \vec{A}_L(t')]^2, \tag{3}$$

where $\vec{A}_L(t)$ is the vector potential of the optical laser field, which is defined as $\vec{A}_L(t) = \int_t^\infty dt'\vec{\mathcal{E}}_L(t')$ [here $\vec{\mathcal{E}}_L(t)$ is the IR laser electric field vector]. To simplify the expressions, hereafter we consider ionization from an s subshell. We assume that both XUV and IR pulses are linearly polarized along the z axis. Reducing the matrix element in Eq. (1) to a single-electron dipole amplitude, $d_{sp}[\vec{k} - \vec{A}_L(t)]$, and collecting Eqs. (1)–(3), one can obtain the following expression:

$$\mathcal{A}_{\vec{k}} = -i\int_{-\infty}^{\infty} dt\tilde{\mathcal{E}}_X(t)d_{sp}[\vec{k} - \vec{A}_L(t)]Y_{1,0}[\theta_0(t),0] \times e^{i\Phi(\vec{k},t)}e^{i(E_b-\omega_X)t}, \tag{4}$$

where $Y_{1,0}(\theta,\phi)$ is a spherical harmonic. The dipole matrix element depends on the quantity $\vec{k} - \vec{A}_L(t)$, which is the electron momentum at the moment of its emission from the atom. Since the dipole matrix element is only weakly dependent on the energy in the considered energy interval, we assume it to be a constant and ignore it in the calculations. Due to the axial symmetry of the process with respect to the z axis, only the $m = 0$ component contributes. The angle θ_0 defines the direction of electron emission from the atom before propagation in the optical laser field. This angle is connected with the detection angle θ after propagation in the IR field by the relation

$$\theta_0(t) = \arccos\{[k_z - A_L(t)]/k_0(t)\}, \tag{5}$$

where $k_z = k\cos\theta$ and $k_0^2(t) = [\vec{k} - \vec{A}_L(t)]^2$. For practical applications, it is convenient to rewrite the amplitude (4) as

$$\mathcal{A}_{\vec{k}} = -i\int_{-\infty}^{\infty} dt\tilde{\mathcal{E}}_X(t)Z(t;k,\theta), \tag{6}$$

with

$$Z(t;k,\theta) = d_{sp}\exp\left[i\left(\frac{k^2}{2} - \omega_X + E_b\right)t\right] \times e^{iX(t,k_z)}Y_{1,0}[\theta_0(t),0], \tag{7}$$

where

$$X(t,k_z) = \frac{1}{2}\int_{-\infty}^{t} dt'\left\{[k_z - A(t')]^2 - k_z^2\right\}. \tag{8}$$

The presentation of the phase $X(t,k_z)$ in such a form is convenient for computations since it is zero when the IR field is absent. When the IR field is operative the phase is not large.

In order to account for partial temporal coherence of the XUV pulses, we use a conventional statistical approach. We have to compute $|\mathcal{A}_{\vec{k}}|^2$ and average it over an *ensemble* of FEL pulses. Then the average differential cross section for photoelectron emission is

$$\begin{aligned}\langle\sigma(\vec{k})\rangle &\equiv \langle|\mathcal{A}_{\vec{k}}|^2\rangle \\ &= \int_{-\infty}^{\infty} dt\int_{-\infty}^{\infty} dt'\,\langle\tilde{\mathcal{E}}_X(t)\tilde{\mathcal{E}}_X^*(t')\rangle Z(t;k,\theta)Z^*(t';k,\theta) \\ &= \int_{-\infty}^{\infty} dt\int_{-\infty}^{\infty} dt'\,g_1(t,t')\tilde{\mathcal{E}}_X^0(t)\tilde{\mathcal{E}}_X^{0*}(t') \\ &\quad\times Z(t;k,\theta)Z^*(t';k,\theta),\end{aligned} \tag{9}$$

where we have introduced the ensemble-averaged envelope of the XUV pulse, $\tilde{\mathcal{E}}_X^0(t) = \sqrt{\langle|\tilde{\mathcal{E}}_X(t)|^2\rangle}$, and the first-order temporal correlation function

$$g_1(t,t') = \frac{\langle\tilde{\mathcal{E}}_X(t)\tilde{\mathcal{E}}_X^*(t')\rangle}{[\langle|\tilde{\mathcal{E}}_X(t)|^2\rangle\langle|\tilde{\mathcal{E}}_X(t')|^2\rangle]^{1/2}}. \tag{10}$$

It is obvious that only the first-order correlation function enters the description since only one-photon absorption of the XUV frequency is considered. In the case of a stationary stochastic process, the temporal correlation function depends only on the time difference, $g_1(t,t') = g_1(t - t')$. We use this simple assumption for the light coherent properties, although strictly speaking it is valid when the XUV pulse is much longer than the coherence time. For a more general consideration it would be necessary to know the correlation function as a function of two variables. Making a Fourier transformation, one can present the ensemble-averaged double-differential cross section (DDCS) as

$$\langle\sigma(\vec{k})\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega\tilde{g}_1(\omega)|Q(\omega;k,\theta)|^2, \tag{11}$$

where

$$\tilde{g}_1(\omega) = \int_{-\infty}^{\infty}\exp(-i\omega\tau)g_1(\tau)d\tau \tag{12}$$

and

$$Q(\omega;k,\theta) = -i\int_{-\infty}^{\infty}\exp(-i\omega\tau)Z(\tau;k,\theta)\tilde{\mathcal{E}}_X^0(\tau)d\tau. \tag{13}$$

Formulas (11)–(13) have a simple physical interpretation. Equation (13) shows that the quantity $Q(\omega;k,\theta)$ is the amplitude of the photoionization by the averaged FEL pulse, $\tilde{\mathcal{E}}_X^0(\tau)$, but with the shifted frequency $\omega_X + \omega$ [compare with Eq. (6)]. Meanwhile, Eq. (11) tells that the ensemble-averaged DDCS can be considered as a weighted incoherent average of the DDCSs calculated for the pulse $\tilde{\mathcal{E}}_X^0(\tau)$ with the frequency shifted by ω. The weighting factor is given by the Fourier transform of the correlation function. One may say that the time dependence of the average pulse envelope determines the coherent process, while the decoherence is introduced through the stochastic shift of the carrier frequency. Of course, it is valid only if the FEL radiation is considered as a stationary stochastic process. A less precise picture can be obtained if one interprets Eq. (13) as an amplitude for the photoionization by

the averaged pulse with the final energy of the photoelectron shifted by ω. This interpretation is reasonable when the electron energy is large, as assumed in our approach initially. With this interpretation, one can treat the final ensemble-averaged DDCS as a result of the incoherent pileup of the curves of the coherent DDCSs for an averaged pulse with a stochastically shifted electron energy. The contribution of each curve is weighted again by the Fourier transform of the correlation function. One can expect that as a result of such averaging each line of the spectrum becomes broader.

The conventional form for the temporal correlation function is Gaussian,

$$g_1(\tau) = \exp\left(-\frac{\tau^2}{\tau_0^2}\right), \tag{14}$$

and its Fourier transform is

$$\tilde{g}_1(\omega) = \frac{\tau_0}{2\sqrt{\pi}} \exp\left(-\frac{\tau_0^2\omega^2}{4}\right). \tag{15}$$

One usually characterizes the temporal coherence properties of the FEL beam by the coherence time, defined by the relation [12,24]

$$\tau_c = \int_{-\infty}^{\infty} d\tau g_1(\tau) d\tau = \sqrt{\pi}\tau_0. \tag{16}$$

The parameter τ_c varies from 0.2 to 9 fs for different FELs [13] and different photon energies [11].

Using Eqs (11)–(16), one can calculate the cross sections of the laser-assisted photoionization for the XUV pulses with partial temporal coherence. As a particular example, we consider the two-color short-pulse photoionization of He. We assume that the ensemble-averaged XUV pulse has a Gaussian profile [3,13] with a duration of 5 fs [full width at half maximum (FWHM) of the electric field]. The electron kinetic energy without an IR field is set equal to 220 eV. We suppose that the IR laser has an 800-nm wavelength, 3.5×10^{12} W/cm^2 intensity, and a pulse duration of 30 fs. For definiteness we assume that the IR and XUV pulses completely overlap and the maxima of the two envelopes coincide. The coherence time τ_c has been varied.

III. RESULTS AND DISCUSSION

Figure 1 shows the calculated differential cross section $\langle\sigma(\vec{k})\rangle$ for two cases: (a) for the electron emission along the polarization vector of both pulses $\theta = 0^\circ$ and (b) for the emission at the angle $\theta = 60^\circ$. The thin black curve shows the spectrum calculated for a fully coherent XUV pulse. Up to ten sidebands are clearly seen at both sides of the main line ($E_0 = 220$ eV). Note that the intensity of the sidebands strongly varies, revealing the gross structure [21,25]. This gross structure is explained by the interference of electrons emitted at different moments of time within one optical cycle. A similar structure has been discussed in above-threshold ionization in Refs. [26,27]. Following an approximate approach used in Refs. [21,25], one can estimate the overall width ΔE of the spectral area where the sidebands are observed, $\Delta E \approx 2A_{L0}\sqrt{2E_0}\cos\theta$, where A_{L0} is the amplitude of the IR vector potential. In our example this gives about ~38 eV for $\theta = 0^\circ$ and ~19 eV for $\theta = 60^\circ$, in

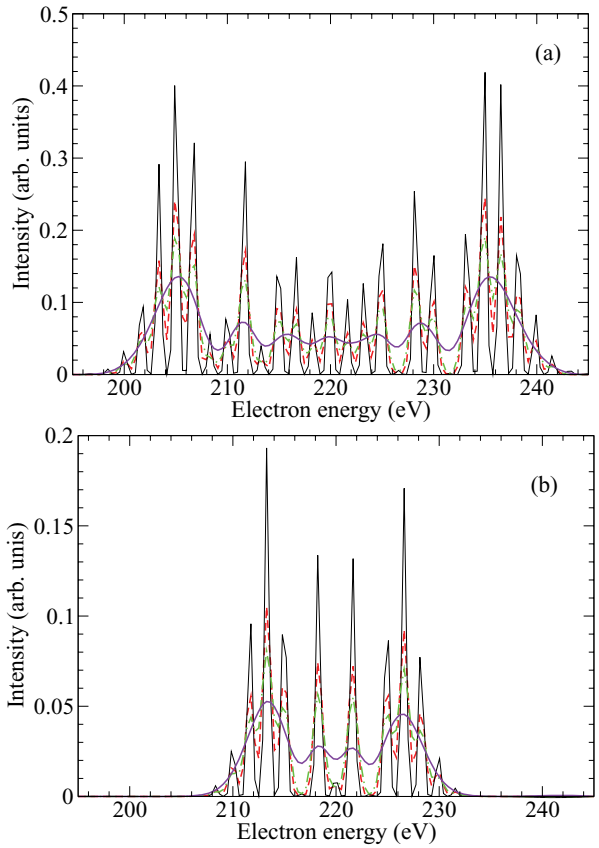

FIG. 1. (Color online) The calculated electron spectra for different values of coherence time. The observation angle is (a) $\theta = 0^\circ$ and (b) $\theta = 60^\circ$. Thin solid (black) line: Fully coherent pulse; dashed (red) line: $\tau_c = 4.8$ fs; dotted-dashed (green) line: $\tau_c = 3.4$ fs; thick solid (violet) line: $\tau_c = 1.4$ fs. See other parameters in text.

agreement with the results, shown in Fig. 1. The further series of curves in Fig. 1 show the spectra at decreasing coherence time. One clearly sees that the sidebands become broader, the minima between them smear out, and finally at $\tau_c = 60$ a.u (1.4 fs) the individual sidebands cannot be observed at all. However, the variations in the spectrum associated with the gross structure are still clearly visible. Note that the FEL pulse with the coherence time τ_c can be considered as a set of spikes of duration τ_c randomly distributed within the pulse. For $\tau_c = 1.4$ fs the spike duration is shorter than the period of the IR laser field (2.6 fs). As it was shown in Ref. [21], in this case the photoelectron spectrum is strongly asymmetrical with respect to the unperturbed photoline, showing a clear effect of streaking (shift of the main maximum to higher or lower energies depending on the delay between the IR and XUV pulses) (see Fig. 3 of the above reference [21]). Ensemble averaging, i.e., averaging over randomly distributed spikes, which leads to the formation of $\tilde{\mathcal{E}}_X^0$, is in a sense equivalent to the averaging over their delay time. It results in an almost symmetrical spectrum (see Fig. 1) with the large maxima on both sides of the spectrum, corresponding to maximal streaking of the individual spikes.

Figure 2 shows the spectra integrated over all emission angles. Such spectra can be observed in experiments with a magnetic bottle-type spectrometer [28]. As it was shown

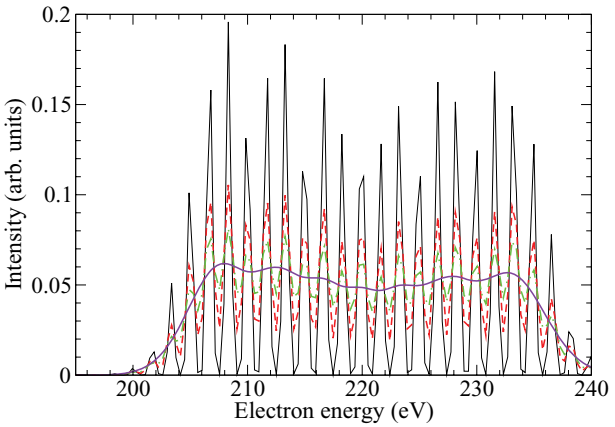

FIG. 2. (Color online) The calculated electron spectra integrated over emission angles for different values of coherence time. Notations are the same as in Fig. 1.

in Ref. [25], in this case the gross structure is much less pronounced. The sidebands, clearly seen for the fully coherent pulse, gradually fade out when the coherence time decreases. At a coherence time $\tau_c = 60$ a.u. (1.4 fs) the sidebands cannot be observed and the spectrum become almost uniform with an overall width of $\Delta E \approx 2A_{L0}\sqrt{2E_0}$.

According to Eqs. (11)–(13) the DDCS averaged over an ensemble of pulses can be considered as a convolution of the DDCS for the purely coherent averaged pulse and the Fourier transform of the correlation function. If we assume that the average pulse envelope is Gaussian, it is easy to show that the observed width of each spectral line is a sum of the width Γ_p determined by the XUV pulse bandwidth and the width $\Gamma_{\rm coh} \approx \sqrt{4\pi}/\tau_c$ associated with the partial coherence. In a real experiment the additional width $\Gamma_{\rm det}$ appears due to the finite energy resolution of the detector, so that

$$\Gamma_{\rm obs}^2 \approx \Gamma_p^2 + 4\pi/\tau_c^2 + \Gamma_{\rm det}^2. \qquad (17)$$

It is clear that if $\Gamma_p^2 + 4\pi/\tau_c^2 \geqslant \omega_L^2$ (here ω_L is the optical laser photon energy, in our case $\omega_L = 1.55$ eV) the sidebands cannot be resolved even with an ideal detector. When the FEL pulse consists of short spikes (small coherence time), one can neglect Γ_p and obtain that for $\sqrt{4\pi}/\tau_c \geqslant \omega_L$ the sidebands cannot be resolved. In the case considered, this estimate gives the condition for the sideband observation $\tau_c \geqslant 1.5$ fs, in good agreement with our computations.

At first glance, the present result contradicts the fact that the sidebands have been first observed in the experiments with practically incoherent light of the plasma source [29]. However, that experiment was performed with Auger electrons. As we show elsewhere, in this case the influence of coherence of the ionizing light plays a minor role. On the contrary, the coherence is important for the observation of the sidebands for photoelectrons. In fact, the first observation of the sidebands for photoelectrons [19] has been made with high-order harmonic radiation as a source of the XUV light which has rather high coherence.

IV. CONCLUSIONS

In conclusion, we have shown that in the two-color ionization by an IR-laser-assisted FEL pulse, the partial coherence of the FEL pulse can be taken into account by using the temporal correlation function of the XUV pulse electric field. Assuming the stochastic properties of the FEL pulse to be stationary, we have shown that the final DDCS can be obtained by averaging the DDCS, determined by the average pulse, over the frequency (or final electron energy) jitter with the weight given by the Fourier transform of the temporal correlation function. If the coherence time is small, less than the inverse frequency of the optical laser field, the sidebands cannot be observed. However, in experiments with angle-resolved photoelectron detection, on both sides of the photoline an additional spectral structure is predicted to appear, which is the remnant of the gross structure of the sidebands. The photoline in the angle-integrated spectrum acquires a width that is equal to the spread of the sidebands in the laser field.

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