I. INTRODUCTION

The rapidly developing techniques of the attosecond metrology based on the concept of attosecond streaking [1–3] made it possible to perform time-resolved measurements of atomic photoionization. This offers the possibility of gaining insights into electron dynamics and fine details of the photoionization process occurring on the attosecond timescale.

These techniques have already revealed unexpected and not yet fully understood phenomena, which are currently being discussed in the literature. In Ref. [4] it was found, for example, that photoionization events from different subshells of a Ne atom proceed apparently with a certain relative time delay, which could not be accounted for by theoretical calculations [4,5].

The theoretical framework allowing us to describe time delay in scattering or weak-field photoionization processes and that was used, for example, in Refs. [4,5] has been laid out in Ref. [6]. More details of this theory will be given in the next section. We may mention here that it connects the observed time delay with characteristics of the atomic potential, which are conveniently encapsulated in the energy derivatives of the scattering phase shifts. In this picture the value of the time delay for the photoionization process driven by the weak electromagnetic (EM) field is, as we shall see, just one half of that for the scattering process. The reason for this is clear. The photoionized electron that escapes the atom has, roughly speaking, to traverse only half the distance that the scattered electron traverses. If the time delay for the photoionization process is determined by the properties of the atomic potential only, then, for the purpose of the calculation of this quantity, photoionization may be considered as “half” a scattering process.

This result is obtained for the photoionization in a weak EM field. For brevity, we shall call this part of the observed time delay, which is determined by atomic potential only, the Coulombic part.

The time delay might change if we consider photoionization in a stronger EM field. It is the purpose of the present work to study how the observed time delay depends on the intensity of the driving laser pulse. As we shall see, for the time delay in a stronger EM field (even of moderately high field strength), we may obtain results that differ quite noticeably from the weak-field Coulombic part.

II. TIME DELAY THEORY

We shall briefly describe below the theoretical approach applicable for calculating time delay for the process of photoionization in a strong EM field. This approach is based on the time delay theory developed in [6,7].

We consider a hydrogen atom initially in the ground state driven by a laser pulse, which, for definiteness, we shall call the EM field, and that was used, for example, in Refs. [4,5] has been laid out in Ref. [6]. More details of this theory will be given in the next section. We may mention here that it connects the observed time delay with characteristics of the atomic potential, which are conveniently encapsulated in the energy derivatives of the scattering phase shifts. In this picture the value of the time delay for the photoionization process driven by the weak electromagnetic (EM) field is, as we shall see, just one half of that for the scattering process. The reason for this is clear. The photoionized electron that escapes the atom has, roughly speaking, to traverse only half the distance that the scattered electron traverses. If the time delay for the photoionization process is determined by the properties of the atomic potential only, then, for the purpose of the calculation of this quantity, photoionization may be considered as “half” a scattering process.

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Theoretical procedure will allow us to obtain the time delay for a strong EM field, and the results we obtain are given below. To avoid unnecessary complexity needed to describe photoionization in atomic systems with more than one electron, we shall choose atomic hydrogen as the target atom in our calculations. Atomic units are used throughout the paper.

ψ(t) = \int a_k \phi_k^\rightarrow e^{-iE_k t} dk,

where \phi_k^\rightarrow are the set of scattering hydrogen states (with ingoing boundary conditions), a_k = \langle \phi_k^\rightarrow | \Psi(t) \rangle e^{iE_k t}, and E_k = k^2/2.

It is easy to see that a_k defined in Eq. (2) are time independent for the field-free evolution of hydrogen atom for t > 4T, when the laser pulse is gone.

If we are interested in describing electron motion in a particular direction with a given asymptotic momentum q (as we shall do below), we can introduce yet another wave function: \Psi_q(t) = \hat{P}_q \Psi(t), projecting wave function (2) on a narrower set \Omega of the hydrogen states \phi^\rightarrow. This can be
done simply by restricting the integration domain in Eq. (2) to some neighborhood of \( q \). We can, for example, define the integration domain as \( |k - q| < \rho \) with some \( \rho \), which will thus determine the wave-packet spread in the momentum space. Such a wave-packet state describes an electron reaching the far-off detector with a given asymptotic momentum \( q \). Another possibility to extract information from the wave function \( \Psi(t) \) defined by Eq. (2) about wave packets traveling in a particular direction (which we will actually use) is based on the Gabor transform of this function, which will be discussed below.

It is not difficult to obtain an asymptotic equation for the function \( r(t) \) trajectory followed by the crest of a wave packet described by wave function \( \Psi_2(t) \). Following standard prescriptions of the saddle-point method, using large-\( r \) asymptotic expressions for the scattering states \( \phi_k \propto e^{i k r + i/2(r,k)} \), and writing \( a_k = |a_k|e^{iv(k)} \) for the wave-packet coefficients, one can easily obtain for large \( t \)

\[
\begin{align*}
\mathbf{r}(t) &\sim q(t) - \frac{d\gamma(q(t), v)}{d\mathbf{v}}|_{v=q} - \frac{d\mathbf{v}(q(t))}{dq},
\end{align*}
\]

where \( \gamma(q,v) = \frac{1}{2} \ln(uv + uv) \). The term containing the derivative of \( \gamma(u,v) \) gives the well-known Coulomb corrections to the trajectory. This term grows logarithmically with \( t \) for large \( t \).

If phase \( v(q) \) depends only on \( q \), then we can put in the last equation \( \frac{d\gamma(q)}{dq} = q \frac{d\gamma(E)}{dE} \), where \( E = q^2/2 \). Eq. (3) can then be rewritten

\[
\begin{align*}
\mathbf{r}(t) &\sim q(t) \left[ t - \frac{d\gamma(E)}{dE} \right] - \frac{d\gamma(q(t), v)}{d\mathbf{v}}|_{v=q}.
\end{align*}
\]

The term with the \( d\gamma(E)/dE \) derivative can be interpreted as the time delay, the quantity that will be of interest to us below.

As we have seen, time delay is determined by the derivative of the phase of the coefficient \( a_k \), taken at the point \( k = q \), corresponding to the asymptotic momentum of the electron. It is easy to write an analytical expression for \( a_k \) in the case of the weak EM field, when perturbation theory (PT) is applicable. Using the well-known PT formula, one can write in the first order of the perturbation theory for \( t > 4T \) (i.e., after the end of the pulse, when coefficients \( a_k \) become time independent)

\[
\begin{align*}
a_k &= -i \int_{-4T}^{4T} \langle \phi_k^* \hat{H}_{\text{int}}(\tau) |\Psi(0)\rangle e^{i(E_0 - E_0)\tau} d\tau,
\end{align*}
\]

where \( \Psi(0) \) and \( E_0 \) are the wave function and energy of the initial (ground) state of hydrogen atom, respectively, and \( \hat{H}_{\text{int}} \) is an interacting Hamiltonian describing atom-EM field interaction.

Using partial wave expansion for the ingoing scattering state [8],

\[
\phi_k = \sum_{l,m} i^l e^{-i\delta(k,l)} R_{kl}(r) Y_{lm}(r/k),
\]

the well-known dipole selection rules [(only \( p \)-wave in the above expansion contributes to the matrix element in Eq. (5)]}, one can easily see that in the first order of the perturbation theory phases \( v(k) \) of the coefficients \( a_k \) in Eq. (5) can be represented as \( v(k) = \delta(k,1) + \text{const} \). We obtain thus the well-known result that in the PT regime the time delay [which is given by the derivative of the phase \( v(k) \)] is given by the derivative of the scattering phase shift in the final \( p \) state of the hydrogen atom and is one half of the time delay value for the scattering process [4,6,7].

For an electron energy of 0.15 a.u., which corresponds to a frequency of 0.65 a.u. of the laser pulse we consider, this will give us a perturbative value of 136 as for the time delay.

In the first order of perturbation theory no effects of the EM field on the time delay are present. It is for this reason that we called the time delay thus obtained Coulombic. This quantity reflects only the properties of the Coulomb potential in which an electron moves after it escapes from the atom. To see the effect of the EM field on the time delay, we must solve the TDSE nonperturbatively.

III. QUANTUM-MECHANICAL CALCULATION OF TIME DELAY

We solve the TDSE for the hydrogen atom in the presence of the external EM field given by Eq. (1)

\[
\begin{align*}
\frac{i}{\hbar} \frac{\partial \Psi}{\partial t} &= [\hat{H}_{\text{atom}} + \hat{H}_{\text{int}}(t)]\Psi,
\end{align*}
\]

where \( \hat{H}_{\text{atom}} \) is the Hamiltonian of the field-free atom and operator \( \hat{H}_{\text{int}}(t) \) describes the interaction of the atom and the EM field. We use the velocity form of this operator:

\[
\hat{H}_{\text{int}}(t) = \mathbf{A}(t) \cdot \mathbf{p}.
\]

The vector potential in Eq. (8) is defined as \( \mathbf{A}(t) = -\int_{-T}^{T} \mathbf{F}(\tau) d\tau \). The quadratic \( \mathbf{A}^2(t) \) term in the interaction Hamiltonian (8) is omitted. This term can be removed through a gauge transformation [9], which is equivalent to multiplying the wave function by an unimportant phase factor.

The computational procedure we employ to solve the TDSE follows closely the procedure used in our work [10] for the calculation of the above-threshold ionization spectra of hydrogen. We shall give, therefore, only a brief description of the procedure.

To treat spatial variables, the TDSE is discretized on a grid with a step size \( \delta r = 0.05 \) a.u. in a box of size \( R_{\text{max}} = 1000 \) a.u.

The wave function is represented as

\[
\Psi(r,t) = \sum_l f_l(r,t) Y_l(\theta),
\]

where the summation in Eq. (9) is restricted to \( l = 0 - L_{\text{max}} \). The particular value of the parameter \( L_{\text{max}} \) is determined by the convergence properties of Eq. (9). The value we used in our calculations reported below was \( L_{\text{max}} = 15 \). We have performed several checks to ensure that this value of \( L_{\text{max}} \) (as well as the value of 1000 a.u. for the box size) is more than sufficient to solve accurately the TDSE for the EM fields of the peak strength not exceeding 0.3 a.u. which we consider below.

To propagate the wave function (9) in time, we use the matrix iteration method (MIM) developed in [11]. It has been shown [10,12] that this technique can be used to accurately solve the TDSE in strong EM fields.

We performed calculations for values \( E_0 = 0.0534, 0.1, 0.2, 0.3 \) a.u. of the peak strength of the EM

\[
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\]
field in Eq. (1). Using solution $\Psi(t)$ of the TDSE that we obtain, we can compute projection $\tilde{\Psi}(t)$ defined in Eq. (2).

In Fig. 1 we show results for the electron spectra and the quantity $r(t) = \langle \tilde{\Psi}(t)|r|\tilde{\Psi}(t) \rangle$, representing the expectation value of the radial coordinate in the projected state $\tilde{\Psi}(t)$ as a function of time.

We would have obtained essentially the same result for $r(t)$ had we used the projected states $\Psi_0(t)$ introduced above for the calculation of the expectation values. We recall that these wave-packet states describe electron motion with a fixed asymptotic electron momentum $q$. The reason that we would obtain almost exactly the same dependence of the electron-nucleus distance in time for all directions (except possibly the directions perpendicular to the polarization axis) is that even for $E_0 = 0.3$ a.u. ionization predominantly proceeds into the $l = 1$ channel. That leaves effectively only the term with $l = 1$ in the partial-wave expansion (6), except for the ionization in the direction perpendicular to the polarization axis, where the $p$-wave contribution identically vanishes. It is easy to see that the expectation values of operator $r$ computed using states $\tilde{\Psi}(t)$ and $\Psi_0(t)$ differ only if $\Omega$ is a region centered around a vector perpendicular to the $z$ axis.

As one can see from the right panel of Fig. 1, trajectories $r(t)$ for $E_0 = 0.1$ and $E_0 = 0.3$ a.u. are quite similar, apart from the fact that group velocities are slightly different (which is a consequence of the fact that the maxima of the wave packets are at slightly different energies, as shown in the left panel of Fig. 1). We could, in fact, try to obtain qualitative information about the time delays by analyzing these trajectories, as was done in [4] and in our paper [5]. It is rather difficult to obtain accurate values for time delay by performing such trajectory analysis since time delay is only the third term in the asymptotic expansion (3) for the electron trajectory. A more direct way to determine time delay is to compute the phases of the coefficients in the Eq. (2), which can easily be done once the solution of the TDSE has been obtained. As we mentioned above, even for EM fields as large as $E_0 = 0.3$ a.u., ionization predominantly proceeds into the $l = 1$ channel. It is easy to observe from the partial-wave expansion in Eq. (6) that, in this case, phases of the coefficients $a_k$ in Eq. (2) are functions of $k$ only, and we can use, therefore, expression (4) to compute time delay.

The results of such calculations of time delays for various directions of the asymptotic electron momentum $q$ are shown in Fig. 2 for the various values of the peak strengths of the EM field. We use a coordinate system with the $z$ axis directed along the polarization vector of the laser pulse, where $\theta$ is an angle between the vector of asymptotic electron momentum $q$ and the positive direction of the $z$ axis. Since we consider the case of linear polarization of the laser pulse, the physical picture we obtain is invariant under rotations around the $z$ axis. In particular, time delay is a function of angle $\theta$ only.

All curves in Fig. 2 exhibit rapid variation in the vicinity of the direction perpendicular to the polarization axis. This is a consequence of the fact that in the direction perpendicular to the polarization axis, the $p$-wave contribution to the time delay is identically zero, and partial waves with $l \neq 1$ in Eq. (6) become important.

As one can see from Fig. 2, this region of rapid variation of time delays as functions of $\theta$ is rather narrow. For the $\theta$ values lying outside this narrow region around $\pi/2$, time delays are nearly constant. This is a consequence of the fact that we have mentioned above, that for the field parameters we consider, photoionization into the $l = 1$ channel is dominating, even for a fairly strong EM field of 0.3 a.u., as long as the asymptotic electron momentum is not perpendicular to the $z$ axis. It is easy to see that if we compute coefficients $a_k$, retaining only the $p$ wave in the partial-wave expansion given by Eq. (6), then phases of $a_k$ will not depend on $\theta$.

More interesting is the fact that the EM field has a considerable effect on the time delays, making them much smaller than the perturbative values. This effect is clearly visible even for rather small values of the EM fields. As one can see from Fig. 2, results for time delays for $E_0 = 0.0534$ a.u. and $E_0 = 0.1$ a.u. are close to each other and to the perturbative value of 136 as we gave above, although, even for $E = 0.1$ a.u., which by all standards belongs to the PT domain for hydrogen, the effect of the EM field on the time delay is clearly discernible. With increasing field strength the effect of the EM field on the time delay quickly becomes more important than the effect of the Coulomb field given by the perturbation theory, modifying the Coulombic value of the time delay (using the terminology we introduced above) quite considerably.

To understand why time delay is affected so strongly by the EM field, we have to take a more detailed look at the photoionization event in which an electron reaches the detector...
with a given asymptotic momentum $\mathbf{q}$. As we discussed above, one way to study such a process would be to follow the evolution in time of a wave packet described by the wave function $\Psi_2(t)$, obtained if we restrict the integration domain in momentum space to some neighborhood of $\mathbf{q}$ in Eq. (2).

Another possibility, which is perhaps more physically transparent, will be used below. It is based on the Gabor transform of the wave function.

**IV. GABOR TRANSFORM**

Following Goldberger and Watson [8], we consider a set of the states $\Phi_{R\mathbf{q}}(\mathbf{r}) = (2\pi)^{-\frac{3}{2}} e^{i\mathbf{q} \cdot \mathbf{r}} G(\mathbf{r} - \mathbf{R})$, where $G(\mathbf{u})$ is any function such that $\int |G(\mathbf{u})|^2 \, d\mathbf{u} = 1$. It is easy to see that completeness relation $\int \Phi_{R\mathbf{q}}(\mathbf{r}_1)\Phi_{R\mathbf{q}}(\mathbf{r}_2) \, d\mathbf{R}/d\mathbf{q} = \delta(\mathbf{r}_1 - \mathbf{r}_2)$ holds, and therefore, $\Phi_{R\mathbf{q}}$ form a complete set. A square integrable $\Psi(\mathbf{r})$ can thus be represented as

$$\Psi(\mathbf{r}) = \int T(\mathbf{R}, \mathbf{q}) \Phi_{R\mathbf{q}}(\mathbf{r}) \, dR/d\mathbf{q}.$$  \hspace{1cm} (10)

with $T(\mathbf{R}, \mathbf{q}) = \int \Phi_{R\mathbf{q}}(\mathbf{r}) \Psi(\mathbf{r}) \, d\mathbf{r}$. In particular, if we choose a Gaussian function for $G(\mathbf{u})$ in these formulas, we obtain the so-called Gabor transform. We shall employ this choice below, using $G(\mathbf{u}) = Ne^{-a^2u^2}$, with $a = 0.05$ and normalization coefficient $N = (2\pi^2/\pi)^{3/2}$.

The Gabor transform and closely related wavelet transforms [13–15] are useful if one wishes to obtain information about the behavior of a function in conjugate domains, for example, in time and frequency domains for the process of harmonics emission [15]. We shall be interested below in applying the Gabor transform to study the wave function $\Psi(t)$ given by (2) in the coordinate and momentum domains. At a given moment of time $t$, Gabor transform (10) represents $\Psi(t)$ as a superposition of wave packets for which expectation values of coordinate and momentum are $\mathbf{R}$ and $\mathbf{q}$, respectively. If we fix $\mathbf{q}$, then Gabor transform coefficients $T(\mathbf{R}, \mathbf{q})$ provide information about the spatial distribution of the wave packets traveling with a given group velocity. In particular, the value of $\mathbf{R}$ for which $|T(\mathbf{R}, \mathbf{q})|$ has a maximum gives us the position of the center of the wave packet traveling with group velocity $\mathbf{q}$ in the coordinate space.

Parameter $a$ in the Gaussian sets the balance between spatial and momentum space resolution, which we can hope to achieve using the Gabor transform representation (10). Increasing this parameter, we increase the spatial resolution at the expense of the momentum space resolution and vice versa. For the particular value of the parameter $a$ that we use, a wave packet with the envelope described by the Gaussian $G(\mathbf{r} - \mathbf{R})$ will have a spread of approximately 20 a.u. in coordinate space and 0.05 a.u. in momentum space. We shall be interested below in the qualitative picture of the motion of the wave-packet crest. For the purposes of this qualitative analysis, such spatial resolution will be sufficient. It is more important for us in this problem to have higher resolution in the momentum space. For the electron energies of the order of 0.15 a.u. and the corresponding momenta $q_0 \approx 0.5$ a.u. that we consider, the Coulomb scattering phases, which determine time delays in the weak-field limit, vary quickly with energy. To get the correct picture of the wave-packet motion, we should therefore use wave packets with spread $\Delta q \ll q_0$. Our choice of the parameter $a$ guarantees that this condition is satisfied.

To illustrate this picture, we show in Fig. 3 the results for the absolute values of the Gabor transform coefficients for the peak strength of EM field $E_0 = 0.1$ a.u. for several moments of time after the end of the EM pulse. We employ the same coordinate frame we used above, presenting results for the time delays in Fig. 2, with the $z$ axis along the polarization vector and the $x$ axis lying in the perpendicular direction.

The Gabor transform coefficients shown in Fig. 3 have been computed for values of the momentum that the electron has at the moments of time $t = 4T, 8T, 12T$, and $16T$. These values were computed as expectation values of momentum operator using the wave function (2). The direction of the vector $\mathbf{q}$ was chosen to make an angle $\pi/4$ with the $z$ axis.

Tracking the motion of the maximum of $|T(\mathbf{R}, \mathbf{q})|$ in the $(x, z)$ plane, we can describe the motion of the wave packets in coordinate space in time. In particular, the time dependence of the distance of this maximum from the nucleus will provide us with the same information as the trajectory $r(t)$ shown in Fig. 1.

To see why time delay is affected considerably by even a moderately strong EM field, we shall also need results of the Gabor transform for stronger EM fields. In Fig. 4 we give the distribution of the absolute values of the Gabor transform coefficients for $E_0 = 0.3$ a.u. Results are shown for the moment of time $t = 4T$, corresponding to the end of the pulse.

We show in Fig. 4 the distribution of the absolute values of $|T(\mathbf{R}, \mathbf{q})|$ for two directions of the velocity vector, making angles $\pi/4$ (right panel) and $3\pi/4$ (left panel) with the polarization axis. Using these results, we can explain the rapid decrease of the time delays with EM field strength that we found above, as shown in the nex section.

**V. TIME DELAY IN STRONG EMFIELDS**

To get a simple qualitative picture, let us assume first that after the end of the EM pulse (i.e., for $t > t_0 = 4T$), the center of the electron wave packet moves along a straight line: $r(t) = q(t - t_0) + r_0$, where $r_0$ is the position vector at $t = t_0$.

We can rewrite this equation as $r(t) = (t - t_0 + q \cdot r_0/q_0^2) + r_0^\perp$, where $r_0^\perp$ is the component of $r_0$ perpendicular to the vector $\mathbf{q}$. We see that if $r_0^\perp$ can, for some reason, be neglected, the previous equation shows that time delay in this simple model is given by $t_0 - q \cdot r_0/q_0^2$. This expression shows that for a given fixed positive $t_0 = 4T$, the larger the value of $\mathbf{q} \cdot r_0$ is, the smaller the value of the time delay is for a given trajectory.

It is not difficult to see that this is precisely what Figs. 3 and 4 tell us. In both cases, as one can observe from Figs. 3 and 4, $|r_0^\perp|$ is small compared to the component of $r_0$ parallel to the vector $\mathbf{q}$. This is a consequence of the fact that ionization, as we have seen, proceeds predominantly into the $l = 1$ channel, hence resulting in the small value for the impact parameter of the classical electron trajectory. As one can see from Figs. 3 and 4, the value of $r_0$ for $E_0 = 0.3$ a.u. corresponding to the maximum of $|T(\mathbf{R}, \mathbf{q})|$ for $t = t_0 = 4T$ is considerably larger than $r_0$ for $E_0 = 0.1$ a.u. The simplified trajectory model
FIG. 3. (Color online) Distribution of absolute values of Gabor transform coefficients $|T(R, q)|$ as function of $R$ for a momentum vector $q$ making an angle $\pi/4$ with the direction of the polarization vector. For the peak intensity of EM field $E_0 = 0.1$ a.u., $|T(R, q)|$ are shown for the moments of time corresponding to (top left) the end of the pulse, (top right) 4 cycles after the end of the pulse, (bottom left) 8 cycles after the end of the pulse, and (bottom right) 12 cycles after the end of the pulse.

The simple model presented above neglected completely any influence of the Coulomb potential on the electron motion for $t > t_0$. It is not difficult to get rid of this simplification. As one can see from Fig. 5, the values of $r_0$ are quite large (26 a.u. in the weak-field limit and rapidly increasing with EM field). We can therefore use in Eq. (2) large-$r$ asymptotic expressions for the hydrogen scattering states, which will lead us to Eqs. (3) and (4). These asymptotic expressions for the wave-packet trajectory differ from the straight-line trajectories we used above only in presence of the Coulomb terms due to the derivatives of the function $\gamma(u, v)$. These terms vary very weakly (logarithmically) with $r_0$ and, consequently, with $E_0$. As long as we are interested in the relative difference of time delays for different EM field strengths, the effect of these terms can therefore be neglected.
VI. CONCLUSION

We have studied the dependence of the time delay on the EM field strength for the process of photoionization from the ground state of a hydrogen atom. In the weak EM field limit, the time delay for this process is determined only by the properties of the atomic potential, which is expressed by the well-known formulas connecting time delay and scattering phase shift.

The situation changes as the EM field strength increases. As we saw, even for relatively small field strengths, which are still in the domain of the perturbation theory for the hydrogen atom, the time delay undergoes quite a considerable change. The reason why the EM field has this rather pronounced effect on time delay can be understood by using the simple model that we discussed above. The value of $r_0$ that we determined above as the expectation value of the radial coordinate at the moment $t = t_0$ can be thought of as an initial condition for the subsequent motion of the center of the electron wave packet. This value grows rather quickly with the EM field strength, resulting in a fast decrease of the time delay.

This effect of the EM field on the time delay could be made even more dramatic had we considered higher photon frequencies. In the weak-field limit, the time delay, determined by the derivative of the scattering phase shift with respect to energy, will be very small (this derivative decreases very quickly with energy). We would then be able to observe considerable change in the time delay for much smaller EM field strengths.

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