

Essential Conditions for Dynamic Interference

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We develop general quantitative criteria for dynamic interference, a manifestation of a double-slit interference in time which should be realizable with brilliant state-of-the-art high-frequency laser sources. Our analysis reveals that the observation of dynamic interference hinges upon maximizing the difference between the dynamic polarization of the initial bound and the final continuum states of the electron during the light pulse while keeping depletion of the initial state small. These two properties, Stark shift and depletion, can be determined from electronic structure calculations avoiding expensive propagation in time. Confirmed by numerical results, we predict that this is impossible for the hydrogen ground state but feasible for excited states; this has been exemplified for the case of the hydrogen $2p$ state.

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Interference is a basic concept ruling optical as well as quantum mechanical wave phenomena, most prominently realized through variations of the double-slit scenario, by means of photoelectron spectroscopy with short-wavelength radiation even on an atomic level [1–4]. The advent of intense laser pulses with a finite pulse length has contributed a new natural double-slit scenario in the time domain: A wave packet that is launched for some dynamic reason at a certain time of the raising part of the pulse, in principle, encounters the same laser envelope amplitude at a certain time during the falling part of the pulse, constituting a double slit in time. If the source for the wave packet has not changed between the two “slits,” interference of both wave packets with maximal possible contrast results, depending on the time interval between the slits. This scenario was experimentally seen [5] and theoretically described [6] early on for bound-state population transfer with low-frequency pulses. It was touched upon in the context of stabilization study with the high-frequency Floquet theory for above-threshold ionization [7,8] to finally become topical under the name dynamic interference in the soft x-ray domain for femtosecond pulses [9,10]. Indeed, the breathtaking development of intense light sources towards attosecond pulse lengths [11] and x-ray frequencies [12] has tremendously broadened the parameter range available for light-matter interaction and, consequently, for the fundamental phenomenon of dynamic interference.

In order to trigger experiments and gain an understanding of the general phenomenon of dynamic interference, in the following, we will work out the parameter windows where dynamic interference is prominent on very different scales of time and energy. Formulating the relevant properties of the laser pulse and the target electron leads us to the appropriate theoretical framework for dynamic interference. Making use of the minimal analytical model described before [10], we will show that only its version

in the (reduced) velocity gauge can be safely used. As it turns out, the same is true for numerical implementations of dynamic interference, although for different reasons. Surprisingly and in contrast to previous claims, we also find that ionization of hydrogen from its ground state does not exhibit dynamic interference, whereas ionization from an excited state does indeed result in dynamic interference.

The soft x-ray regime we will be mainly concerned with here (electron excess energies below 100 eV) is challenging from a theoretical point of view, since single-photon ionization in the vacuum ultraviolet regime *cannot* be taken as the indication for a standard perturbative light-matter coupling: First, there may be substantial depletion of the ionized state during the pulse, and second, multiphoton processes can be involved as indicated by appreciable dynamic Stark shifts (also referred to as ac Stark shifts) of energies. However, due to the weak transitions in the continuum, multiphoton interaction does not lead to substantial multiphoton ionization, in contrast to infrared or optical pulses [13].

For this intermediate regime of light-matter coupling, which is neither fully perturbative nor does it lead to multiphoton ionization, we will identify the two dimensionless parameters δ and γ accounting for the dynamic Stark shift and depletion of the initial state, respectively. The appearance of dynamic interference depends on a suitable ratio of these two parameters.

We start from the standard minimal-coupling Hamiltonian written in the velocity gauge:

$$\hat{H}^{\text{vel}} = \frac{1}{2} [\hat{\mathbf{p}} + \mathcal{A}(t)]^2 + V(\hat{\mathbf{r}}), \quad (1a)$$

where V is some external potential and

$$\mathcal{A}(t) = \mathcal{A}_0 g(t) \cos(\omega t) \quad (1b)$$

is the vector potential of the laser pulse with a Gaussian envelope $g(t) = \exp(-t^2/T^2)$. We use atomic units throughout the text unless noted otherwise. We represent solutions $|\psi(t)\rangle$ of the time-dependent Schrödinger equation for (1) with an expansion into field-free (bound and continuum) states φ_α :

$$|\psi(t)\rangle = e^{-(i/2) \int^t dt' \mathcal{A}^2(t')} \sum_{\alpha} |\varphi_{\alpha}\rangle e^{-iE_{\alpha}t} a_{\alpha}(t), \quad (2)$$

where the exponential prefactor transforms away $\frac{1}{2} \mathcal{A}^2(t)$ appearing in (1a). We will call this new gauge the *reduced velocity gauge*. The index α comprises all quantum numbers defining the eigenstate, which in the case of continuum states are the energy E and the symmetry κ . The standard 1st-order time-dependent perturbation theory predicts the amplitude (of continuum states at energy E)

$$a_{E,\kappa} = -i \mathbf{p}_{E,\kappa} \int dt \mathcal{A}(t) e^{i[E-E_{\text{in}}]t} a_{\text{in}}(t) \quad (3)$$

for ionization to a final energy E from initial energy E_{in} . The dipole matrix elements $\mathbf{p}_{E,\kappa} \equiv \langle \varphi_{E,\kappa} | \hat{\mathbf{p}} | \varphi_{\text{in}} \rangle$ connect the initial state φ_{in} to continuum states $\varphi_{E,\kappa}$ of energy E . Because of selection rules, only some of the matrix elements are nonzero. For the photoeffect, implying weak perturbations $\mathcal{A}(t)$, Eq. (3) allows for an explicit solution, since one may assume that $a_{\text{in}}(t) = 1$ for all times.

For dynamic interference, however, the dynamic Stark shift and the depletion of the initial state become relevant. As long as the laser envelope varies slowly compared to the laser cycle, the system remains in an adiabatic regime where one may average the response of the system to the laser field over the laser cycle to arrive at a formulation solely expressed in terms of the laser envelope $g(t)$. Incorporated into (3), one obtains a modified coefficient $a_{\text{in}}(t)$ which still allows for a solution (at least in terms of a stationary-phase approximation) as before [9,10]. Hereby, the phase of a_{in} becomes time dependent:

$$a_{\text{in}}(t) = e^{-i\phi_{\text{in}}^{\delta\gamma}(t)} \quad (4a)$$

$$\phi_{\text{in}}^{\delta\gamma}(t) = [\delta - i\gamma/2] E_p T G(t), \quad (4b)$$

with the ponderomotive energy E_p and the dimensionless function G , respectively, defined by pulse parameters

$$E_p \equiv \frac{\mathcal{A}_0^2}{4} = \frac{I}{4\omega^2}, \quad (4c)$$

$$G(t) \equiv \frac{1}{T} \int^t dt' g^2(t'). \quad (4d)$$

The derivative $d\phi_{\text{in}}^{\delta\gamma}/dt$ can be interpreted as the complex, frequency-dependent energy of the initial state in the

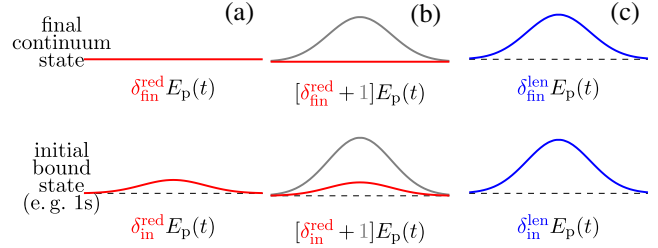


FIG. 1. Sketch showing the relation of the dynamic Stark shifts $\Delta(t)$ of both the initial bound and final continuum states for (a) the reduced velocity gauge $\hat{H}^{\text{red}} = \frac{1}{2} \hat{\mathbf{p}}^2 + \mathcal{A}(t) \hat{\mathbf{p}} + V(\hat{\mathbf{r}})$, (b) the velocity gauge $\hat{H}^{\text{vel}} = \frac{1}{2} [\hat{\mathbf{p}} + \mathcal{A}(t)]^2 + V(\hat{\mathbf{r}})$, and (c) the length gauge $\hat{H}^{\text{len}} = \frac{1}{2} \hat{\mathbf{p}}^2 + V(\hat{\mathbf{r}}) - \frac{d}{dt} \mathcal{A}(t) \cdot \hat{\mathbf{r}}$, respectively. See also Sec. 2 of Supplemental Material [14].

laser pulse, proportional to the (peak) ponderomotive energy E_p . Thereby, δ accounts for the Stark shift Δ [the Stark shift is indeed the time derivative of the phase $\Delta(t) = \delta E_p T (d/dt) G(t) = \delta E_p(t)$ with $E_p(t) = E_p g^2(t)$], and the decay width γ accounts for the depletion. Both constants depend on the laser frequency ω and can be derived from the 2nd-order time-independent perturbation theory (see Sec. 1 of Supplemental Material [14]) or equally extracted from a numerical propagation [10]. Obviously, the minimal description (4) is valid only as long as the Stark shift and decay width are linear in E_p .

In Eq. (4), the dynamic Stark shift has been introduced only to the initial state but not to the final state in the continuum. This is legitimate only in the reduced velocity gauge, where each state has just an intrinsic dynamic Stark shift as shown in Fig. 1(a), with the one for the continuum being in general negligible. This also applies to the velocity gauge [Fig. 1(b)], where all states have an *additive* ponderomotive shift, which can be easily removed by a global phase in the wave function as done in Eq. (2). By contrast, in the length gauge the ponderomotive shift *cannot* be easily separated, as indicated in Fig. 1(c). There, to a good approximation [17], the Stark shift of continuum electrons is given by the ponderomotive energy. Consequently, the intrinsic Stark shift of any continuum state in the reduced velocity gauge is rather small and negligible, rendering the description with Eq. (4) adequate.

As just emphasized, in the length gauge the (trivial) ponderomotive shift cannot be split off the initial or final states and therefore has to be covered by any convergent numerical calculation, which is typically much more demanding than in the velocity gauge (e.g., many more partial waves are required). This is the reason why it has been noted in long-wavelength strong-field physics that the velocity gauge is preferable for numerical calculations [18,19].

From Fig. 1, one obtains an intuitive understanding regarding the mechanism behind dynamic interference independent of the gauge: The initial-state energy increased

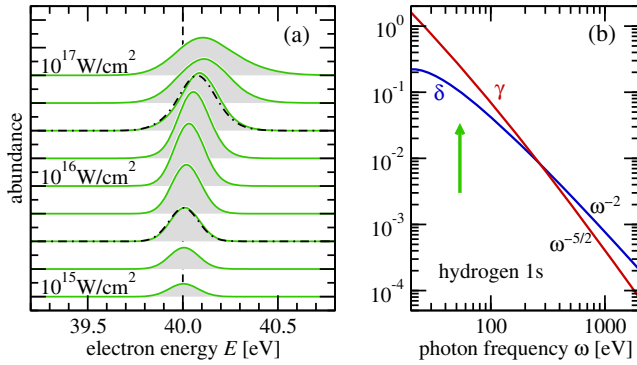


FIG. 2. (a) Photoelectron spectra for $1s$ hydrogen exposed to 10 fs pulses with a carrier frequency of $\omega = 53.60$ eV for intensities $I_k = 10^{k/4} \times 10^{15}$ W/cm 2 with $k = 0, 1, \dots, 8$. The dashed line marks the energy $E_\omega = E_{1s} + \omega = 40$ eV. The result of the minimal model (4) is shown for two intensities by dot-dashed lines. (b) Dimensionless parameters $\delta(\omega)$ and $\gamma(\omega)$ for the hydrogen $1s$ state as introduced in Eq. (4) and defined in detail in Sec. 1 of Supplemental Material [14]. The asymptotic behavior of them for $\omega \rightarrow \infty$ is given for both [20]. The green arrow marks the frequency $\omega = 53.6$ eV used in the left panel and in previous publications [9,10].

by ω for single-photon absorption may intersect the energy E of the final state at two points (in time). These time instants are stationary-phase points where the time derivative of the phase in the integral (3) vanishes; hence, the amplitudes at these two dominate the integral constituting the two-slit scenario. Dynamic interference will be most pronounced if the Stark shifts of initial and final states are very different. Quantitative details as well as the possibility for dynamic interference in the first place depend, of course, on the parameters entering the phase, namely, the electronic response properties $\delta(\omega)$ and $\gamma(\omega)$ in connection with the pulse properties E_p and T , which we will analyze next.

From the minimal model (4), it is easy to see that two conditions must be fulfilled for dynamic interference: (i) The Stark shift must be larger than the bandwidth of the pulse with length T in order to be energetically resolved, and (ii) depletion should be sufficiently weak in order to have ionization in the rising *and* falling wings of the pulse. In order to quantify these conditions, we note that the bandwidth of the pulse (1b) is $\sqrt{2}/T$ and that $G(0) = \sqrt{\pi/2}$. Thus, on one hand, condition (i) is satisfied if

$$\delta E_p > \sqrt{2}/T \quad \text{or} \quad E_p T > \frac{\sqrt{2}}{\delta}. \quad (5)$$

On the other hand, condition (ii) is fulfilled if

$$\frac{\gamma}{2} E_p T G(0) < 1 \quad \text{or} \quad E_p T < \frac{\sqrt{2/\pi}}{\gamma}. \quad (6)$$

These two conditions give lower and upper limits for the product $E_p T$. Apparently, they can be met simultaneously—thus allowing for dynamic interference—only if

$$\delta > \sqrt{\pi} \gamma, \quad (7)$$

which implies that in the competition between the Stark shift and depletion the former should dominate. This condition holds for any atom or molecule. As a consequence, we can predict the laser parameters for which one will observe dynamic interference, provided the response parameters $\delta(\omega)$ and $\gamma(\omega)$ are known. They are shown in Fig. 2(b) for the ground state of hydrogen as an example.

Having condition (7) in mind, one sees from Fig. 2(b) that this requires frequencies larger than $\tilde{\omega} = 265$ eV [21], where $\gamma(\tilde{\omega}) = \delta(\tilde{\omega})$ holds. This is confirmed by the numerical photoabsorption spectra in Fig. 2(a) determined by the direct propagation of the time-dependent Schrödinger equation in the (reduced) velocity gauge for the photon frequency ω marked by the green arrow in Fig. 2(b). Numerical details are given in Sec. 3 of Supplemental Material [14]; the parameters used are $\ell_{\max} = 4$, $r_{\max} = 3000 a_0$, $n = 3000$, and $E_{\max} \approx 134$ eV. As one can see from Fig. 2(a), the spectrum has a single photoelectron peak which gets Stark-shifted and broadened for increasing intensities while keeping the pulse length fixed at $T = 10$ fs. The results have been confirmed with two other packages [22,23] for the numerical propagation of the time-dependent Schrödinger equation.

In contradiction to these results, dynamic interference has been reported for the hydrogen ground state [9,10,24,25]. Even more puzzling, the numerical findings (obtained in the length gauge) seem to be supported by corresponding results from the minimal model in the length gauge appearing in the same papers. It is very rare and unfortunate that two independent mistakes, both related to a faulty usage of the length gauge, one in the model and one in the numerical calculation, should lead to agreeing results, seemingly reassuring the findings regarding dynamic interference [26].

The analytical mistake is easy to identify and originates from using the minimal model (4) in the length gauge but leaving out the Stark shift in the continuum state [see Fig. 1(c)]. Thereby, the difference between the time-dependent energies of initial and final states is artificially increased, leading to the appearance of stationary-phase points of the integrand in (4) at the wrong intensities. The numerical calculations were carried out in the length gauge with a limited number of partial waves, not enough to properly describe the ponderomotive motion of the electron. This parallels the mistake in the analytical model, where the ponderomotive energy dependence was left out and therefore leads to the accidental agreement of numerical and analytical calculations in those papers [9,10,24,25]. A detailed comparison of numerical calculations in the length and velocity gauges for different maximal angular

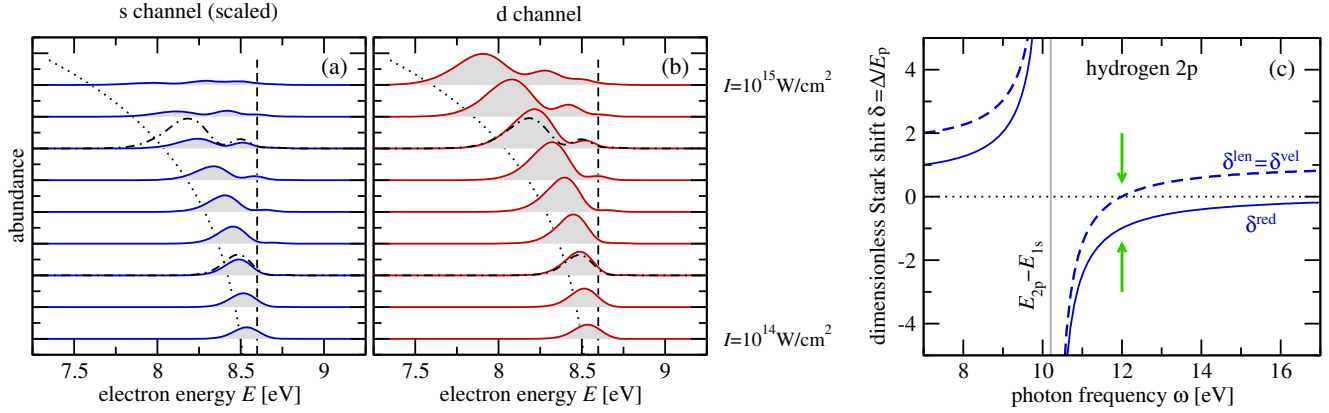


FIG. 3. Photoelectron spectra for 10 fs pulses with a carrier frequency of $\omega = 12$ eV for hydrogen in the $2p$ state. Ionization into the s channel (a) and the d channel (b) is shown. The intensities from bottom to top are $I_k = 10^{k/8} \times 10^{14}$ W/cm 2 with $k = 0, 1, \dots, 8$. The dashed line marks the energy $E_\omega = E_{2p} + \omega = 8.59875$ eV, and the dotted line interpolates between the energies $E_{\omega,k} = E_{2p} + \omega - E_p(I_k)$. The s channel is scaled by the factor f in order to make the height of the peak equal in the perturbative limit: $f = |\mathbf{p}_{E_{\omega,s}}|/|\mathbf{p}_{E_{\omega,d}}| \approx 6.137$. The result of the minimal model (4) is shown for two intensities by dot-dashed lines. (c) Dimensionless Stark shift δ for the $2p$ state of hydrogen as a function of the photon frequency ω in units of the ponderomotive energy $\delta \equiv \Delta/E_p$. We show $\delta^{\text{len}} = \delta^{\text{vel}}$ (blue solid curves) and $\delta^{\text{red}} = \delta^{\text{vel}} - 1$ (blue dashed curves); cf. Sec. 2 of Supplemental Material [14]. The green arrow marks the photon frequency $\omega = 12$ eV, where the Stark shift vanishes and which was used in the two left panels. The Stark shift diverges at the transition energy $E_{2p} - E_{1s}$ (gray solid line).

momenta can be found in Sec. 3 of Supplemental Material [14].

Extrapolating from the conditions for dynamic interference in ionizing hydrogen, it seems very difficult to realize this phenomenon with state-of-the-art laser systems. Yet, this can be easily achieved starting from the $2p$ excited state, which at the same time highlights the relevance of considering the proper Stark shifts. Starting from $2p$ and choosing appropriate intensities and frequencies of the laser, we can prepare an effective initial state which has no Stark shift in the length gauge (or a large negative Stark shift in the reduced velocity gauge). This implies a large difference in the Stark shifts between the continuum and initial bound states and therefore offers excellent conditions for dynamic interference.

At a frequency of $\omega = 12$ eV, the dynamic Stark shift of the $2p$ state vanishes (in the length and velocity gauges) and is therefore given by $\Delta(t) = -E_p(t)$ in the reduced velocity gauge, as can be seen in Fig. 3(c). At this frequency, the coupling to the $1s$ state fully compensates the coupling to all other states such that the polarizability and the Stark shift vanish. We have performed a propagation with the same parameters as before and obtained the spectra shown in Figs. 3(a) and 3(b); cf. details in Sec. 2 of Supplemental Material [14]. Since we start from a p state, photoelectrons are emitted into s and d channels, where the yield for the latter is larger due to the larger dipole matrix element. However, qualitatively, both angular momentum channels exhibit the same behavior for increasing intensities I . For low intensities, the spectrum is Gaussian-shaped and slightly redshifted with respect to $E_\omega = E_{2p} + \omega$. This shift increases with larger I , and for $I \gtrsim 5 \times 10^{14}$ W/cm 2 one clearly sees dynamic

interference in both channels. We note in passing that, according to earlier publications [9,10], one would not expect any dynamic interference at all here. Moreover, in contrast to the blueshift predicted previously, we observe a redshift increasing with intensity which follows directly from $\Delta(t) < 0$ mentioned above.

In summary, by formulating single-photon ionization in the 1st-order time-dependent perturbation theory with phases obtained from the 2nd order, we have derived quantitative conditions under which dynamic interference can occur. The approach is tailored towards the dynamic regime of nonperturbative single-photon ionization, characteristic of interactions with intense soft x rays, and has allowed for the separation of the frequency-dependent response of the electronic system in terms of Stark shift $\delta(\omega)$ and depletion $\gamma(\omega)$ and the time-dependent laser pulse envelope. This separation facilitates the determination of the electronic response by electronic structure calculations and helps to accurately assess experimental conditions for dynamic interference.

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