General Theory of Carrier-Envelope Phase Effects

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We present a general framework for understanding carrier-envelope phase (CEP) effects in a quantum system interacting with an intense, short laser pulse. We establish a simple connection between the CEP and the wave function that can be exploited to obtain the full CEP dependence of an observable given the wave function at a single CEP. Within this framework, all CEP effects are interpreted as interference between different photon amplitudes which, in turn, can be used to put limits on the pulse lengths and intensities required to see significant CEP effects.

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Progress in producing and manipulating intense, few-cycle laser pulses has made it possible to study laser-matter interactions in a qualitatively new regime where the carrier phase relative to the pulse envelope maximum—the carrier-envelope phase (CEP)—becomes an important parameter. It has been experimentally demonstrated, for instance, that the CEP leads to an asymmetry in the ionization of Kr atoms by linearly polarized infrared laser pulses [1]. More recently, a similar asymmetry was found in ionization of Rydberg states of Rb by few-cycle radio frequency pulses [2]. Strong CEP effects have also been predicted for dissociation of H_2^+ and HD^+ [3] and have recently been measured in dissociation of D_2 [4]. Many other theoretical CEP studies have appeared [5–7], including a prediction of weaker CEP effects in the excitation and ionization of Cs atoms [8,9]. Theory has also shown CEP effects in molecular isomerization [10], and high harmonic generation has shown clear CEP dependence experimentally [11].

Despite so much interest, CEP effects are not yet understood theoretically in any general way. Qualitative explanations have been given for many of the studies mentioned above, and several of these support their explanations with solutions of the time-dependent Schrödinger equation (TDSE), but few have gone further. One interesting interpretation [12] describes CEP effects in atomic ionization as two-slit interference in the time domain. In the end, nearly all CEP effects appear by breaking some spatial symmetry present for longer pulses, generally resulting in more localized wave functions. In this respect, the CEP plays a role much like the relative phase in long-studied two-color schemes (see, for example, Ref. [13]).

Our goal is to present a general, rigorous formulation of CEP effects that combines some ideas that have already been applied in this context with some that have not. In particular, our picture reveals a general, simple, and exact relationship between the laser phase and the wave function for short pulses that provides a unifying framework within which previously identified CEP phenomena can be understood both qualitatively and quantitatively.

The key step in our approach is separating the rapid oscillation of the carrier wave from the slower variation of the pulse envelope by combining the 2D-time formalism with a Floquet representation [14–16]. This combination allows the CEP to be eliminated by a simple transformation that, in turn, permits one to calculate the *complete* CEP dependence of any observable from the wave function calculated for a *single* CEP. Recent work has used a geometric transformation to accomplish the same goal for circularly [5] or bi-circularly [6] polarized light. While our formulation can also be applied to these cases, we will assume here that the laser is linearly polarized for notational simplicity.

We start from the TDSE, stressing the CEP, φ, dependence (atomic units are used unless stated otherwise),

$$i\frac{\partial}{\partial t}\Psi(\varphi;t) = [h_0 + V(\varphi;t)]\Psi(\varphi;t). \tag{1}$$

h_0 is the field-free Hamiltonian, and the laser-matter interaction is $V(\varphi;t) = -\mathbf{d}\cdot\boldsymbol{\mathcal{E}}(t)\cos(\omega t+\varphi)$ in the dipole approximation, with $\mathbf{d}$ the dipole operator, $\boldsymbol{\mathcal{E}}(t)$ the pulse envelope, and ω the carrier frequency. The carrier-envelope representation is reasonable for Gaussian pulses so long as the pulse length is longer than roughly one cycle; shorter pulses increasingly violate the requirement that $\int \mathcal{E}(t)\cos(\omega t+\varphi)dt = 0$ [17].

In the 2D-time representation, one introduces a second time s, such that the interaction becomes

$$V(\varphi;s,t) = -\mathbf{d}\cdot\boldsymbol{\mathcal{E}}(s)\cos(\omega t+\varphi), \tag{2}$$

and the TDSE, Eq. (1), becomes

$$\left(i\frac{\partial}{\partial s} + i\frac{\partial}{\partial t}\right)\Psi(\varphi;s,t) = [h_0 + V(\varphi;s,t)]\Psi(\varphi;s,t). \tag{3}$$

If $\Psi(\varphi;s,t)$ satisfies (3), then the solution of (1) can be written as $\Psi(\varphi;t) = \Psi(\varphi;s,t)|_{s=t}$. More details of the 2D-time formalism can be found in Refs. [14,15].

Since Eq. (2) makes Eq. (3) periodic in t, we can utilize the Floquet representation [15,16] to expand $\Psi(s,t)$ as

$$\Psi(\varphi; s, t) = \sum_{n=-\infty}^{\infty} \phi_n(\varphi; s) e^{in\omega t}. \quad (4)$$

The coefficients $\phi_n(\varphi; s)$ are the n-photon emission ($n > 0$) and absorption ($n < 0$) amplitudes. Substituting this Ψ into Eq. (3) yields

$$i\frac{\partial}{\partial s}\phi_n = (h_0 + n\omega)\phi_n - \frac{1}{2}\mathbf{d}\cdot\boldsymbol{\mathcal{E}}(s)(e^{i\varphi}\phi_{n-1} + e^{-i\varphi}\phi_{n+1}). \quad (5)$$

Propagating this equation to large s and evaluating the resulting Ψ at $s = t$ yields the exact solution of Eq. (1) from which any desired observable may be calculated.

The main advantage of Eq. (5) is that the CEP dependence can be eliminated by a simple unitary transformation. To see this, we define the vector $\mathbf{\Phi}(\varphi; s)$ of n-photon amplitudes $\phi_n(\varphi; s)$ and rewrite Eq. (5) as

$$i\frac{\partial}{\partial s}\mathbf{\Phi}(\varphi; s) = \mathbf{H}(\varphi; s)\mathbf{\Phi}(\varphi; s). \quad (6)$$

Defining $[\mathbf{U}(\varphi, \varphi_0)]_{mn} = \delta_{mn}e^{in(\varphi-\varphi_0)}$, it can be verified that the φ dependence can be transformed out of $\mathbf{H}$:

$$\mathbf{H}(\varphi; s) = \mathbf{U}(\varphi, \varphi_0)\mathbf{H}(\varphi_0; s)\mathbf{U}^{\dagger}(\varphi, \varphi_0). \quad (7)$$

This equation establishes our main result: namely, that the complete CEP dependence of the wave function—and thus any observable—can be obtained from

$$\mathbf{\Phi}(\varphi; s) = \mathbf{U}(\varphi, \varphi_0)\mathbf{\Phi}(\varphi_0; s), \quad (8)$$

given $\mathbf{\Phi}$ at a single CEP, $\varphi = \varphi_0$ (hereafter, we will take $\varphi_0 = 0$ for simplicity). Equation (8) implies that the wave function in physical time ($s = t$) is

$$\Psi(\varphi; t) = \sum_{n=-\infty}^{\infty} e^{in\varphi}\tilde{\phi}_n(0; t), \quad (9)$$

where $\tilde{\phi}_n(0; t) = e^{in\omega t}\phi_n(0; t)$. In practice, we only apply this relation after the laser pulse is off at $t = t'$, since the $\tilde{\phi}_n$ then evolve freely as $\tilde{\phi}_n(t) = e^{-ih_0(t-t')}\tilde{\phi}_n(t')$.

Since these results have been obtained under quite general assumptions, we can conclude that *all* CEP effects can be interpreted as interference between different n-photon channels. Such an interpretation has been discussed before for particular cases [2,8,18] as has the conjugate relation (9) between CEP and photon number for two-color pulse trains [18]. The present results, however, apply to all cases, including single Gaussian pulses.

Because the CEP dependence is explicit in our wave function, we can also find the explicit CEP dependence of any given observable O. Using (9) for times $t > t'$, we can write the expectation value of O as the Fourier series

$$\langle O\rangle(\varphi, t) = \frac{\alpha_0}{2} + \sum_{k=1}^{\infty}[\mathrm{Re}(\alpha_k)\cos k\varphi - \mathrm{Im}(\alpha_k)\sin k\varphi], \quad (10)$$

where $\alpha_k(t) = 2\sum_n \langle\tilde{\phi}_{n-k}(0; t)|O|\tilde{\phi}_n(0; t)\rangle$. As the notation suggests, α_k and $\langle O\rangle$ are, in general, time-dependent depending on the specific observable under consideration.

Equation (10) shows immediately that the φ-averaged value of O is $\overline{\langle O\rangle}(t) = \alpha_0(t)/2$. The variance of $\langle O\rangle(\varphi, t)$ with φ—which provides, of course, a convenient measure of the magnitude of the CEP effect—is

$$\sigma(t) = \left[\frac{1}{2\pi}\int_0^{2\pi}(\langle O\rangle - \overline{\langle O\rangle})^2 d\varphi\right]^{1/2} = \left(\frac{1}{2}\sum_{k=1}^{\infty}|\alpha_k|^2\right)^{1/2}. \quad (11)$$

Equation (10) also suggests that a Fourier transform of the CEP dependence of O might provide a means to measure the amplitude and phase of its matrix elements.

To demonstrate how our formulation works in practice, we will consider a simple two-level system governed by

$$i\frac{\partial}{\partial t}\begin{pmatrix} a \\ b \end{pmatrix} = \begin{pmatrix} 0 & V(t) \\ V(t) & \Delta \end{pmatrix}\begin{pmatrix} a \\ b \end{pmatrix}, \quad (12)$$

where $V(t) = V_0 e^{-(t/\tau)^2}\cos(\omega t + \varphi)$ and $\sqrt{2\ln 2}\tau$ is the intensity FWHM pulse duration. Figure 1(a) shows the excitation probability $P_b(\varphi)$ obtained by direct numerical integration of Eq. (12) assuming $a(-\infty) = 1$ and $b(-\infty) = 0$. The parameters correspond roughly to the $1s\sigma_g$ and $2p\sigma_u$ states of H_2^+ at a *fixed* internuclear distance of $R = 4.56$ in a 7 fs (FWHM), 790 nm laser pulse of peak intensity 1.1×10^{14} W/cm^2.

Figure 1(a) also shows $P_b(\varphi)$ obtained from the solution of Eq. (5) for $\varphi = 0$ and extended to all φ using Eq. (10)

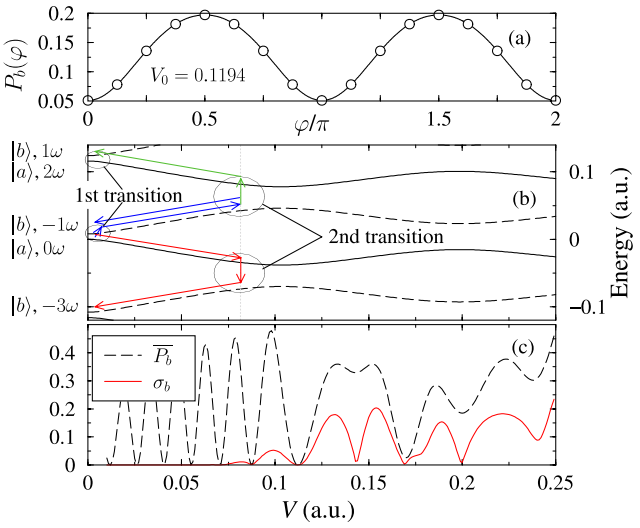

FIG. 1 (color online). For the two-level system of Eq. (12) with $\Delta = 0.066$, $\omega = 0.058$, and $\tau = 248$: (a) Excitation probability from Eq. (14) (solid line) and from solving Eq. (12) (circles). (b) Floquet Hamiltonian eigenenergies adiabatic in V. The arrows indicate the main pathways for $V_0 \approx 0.075$ (vertical dashed line). (c) The CEP-averaged excitation probability $\bar{P}_b$ and the magnitude of the CEP effect σ_b from Eq. (11).

with $O = |b\rangle\langle b|$. Explicitly,

$$P_b(\varphi) = |\langle b|\Psi\rangle|^2 = \left| \sum_{n\text{odd}} e^{in\varphi} \tilde{b}_n(t') \right|^2. \quad (13)$$

For the parameters used, we included $n = -15, \ldots, 15$ and chose $t' = 6\tau$ to ensure convergence. The $n = -3$, -1, and 1 amplitudes dominate the rest, giving

$$P_b(\varphi) = 0.1299 - 0.073\,07\cos 2\varphi - 0.005\,864\cos 4\varphi. \quad (14)$$

Since no approximations have been made besides truncating the series in Eq. (10), the essentially perfect agreement between the direct solution of the TDSE and our 2D-time solution is not surprising.

Besides providing a convincing demonstration of our approach, this example also illustrates the utility of our formulation for understanding CEP effects more qualitatively. For instance, how do the peak intensity and the pulse duration affect the magnitude of CEP effects?

It is now clear that at least two n-photon components of $|b\rangle$ must be populated after the pulse. Since this requires multiphoton transitions, there must be a minimum intensity that produces a clear CEP effect. Figure 1(b) illustrates this point using the eigenenergies of the Floquet Hamiltonian $\mathbf{H}$, adiabatic in V. Starting from the state $|a\rangle$ with no photons ($|a\rangle$, 0ω) at $V = 0$ before the pulse, the system follows all possible pathways along the curves as V increases, with some probability of making nonadiabatic transitions at each avoided crossing. As V decreases on the trailing edge of the pulse, the avoided crossings are again traversed with the same probabilities of transitions. In fact, nonadiabatic transitions are necessary in order to see CEP effects since these are the only ways to populate multiple n-photon states in this picture.

These arguments allow us to understand why the $\tilde{b}_1$, $\tilde{b}_{-1}$, and $\tilde{b}_{-3}$ amplitudes are dominant for the parameters of Fig. 1(b). The arrows in Fig. 1(b) trace the evolution along the most important paths during the pulse. Nonadiabatic transitions occur at the avoided crossings near $V = 0$ and 0.1 on both the rising and falling edges of the pulse, populating the $n = 1$, -1, and -3 states which, in turn, interfere to give the calculated CEP dependence. By the same argument, values of V_0 greater than the next crossing at 0.2 would populate a larger range of n, leading to more complicated CEP dependence.

We can thus set a lower limit on the peak intensity: no interference can happen if V_0 does not approach the second crossing. This observation is illustrated in Fig. 1(c) which shows the magnitude of the CEP effect σ_b. Only for V_0 above the second crossing is σ_b significant.

High intensities alone are not enough to produce CEP effects, however. The field should also pass rapidly through the crossings, otherwise the system follows the adiabatic path and no population transfer occurs. This requirement provides the means to understand the pulse duration dependence of CEP effects.

As we have seen, the nonadiabatic second transition limits the magnitude of the CEP effect. Although it does not strictly apply here, we can use the Landau-Zener model to roughly estimate this transition probability—and thus the size of CEP effects—as a function of τ:

$$P_{\text{LZ}} = 2e^{-2\pi\delta}(1 - e^{-2\pi\delta}), \qquad \delta = \frac{|V_{12}|^2}{|\dot{V}_{11} - \dot{V}_{22}|} \propto \tau. \quad (15)$$

Here, V_{12} is the coupling at the crossing and $\dot{V}_{ii} = dV_{ii}/dt$, which is generically proportional to τ^{-1}. Since σ_b is proportional to the transition amplitude from Eq. (11), i.e., $|\alpha| \propto \sqrt{P_{\text{LZ}}}$, it follows that CEP effects should decrease exponentially with τ. This result is shown in Fig. 2(a) together with σ_b calculated directly from Eqs. (5) and (11) for each τ. The striking qualitative agreement between this simple estimate and the exact result gives us confidence that our interpretation is correct.

It is important to emphasize that this τ dependence is a property of the combined laser-matter system and not the laser pulse alone. Since CEP effects are the result of interference, the greatest modulation will result when the contributing amplitudes have similar magnitude. If one is much larger, as at a resonance, then there will be comparatively small CEP effects [8]. Consequently, a judicious choice of parameters can yield significant CEP effects even for pulses that are much longer than one cycle, as shown in Fig. 2(b). We see that even for pulses as long as $\tau = 15T$, $\sigma_b = 0.1$ (compared with $\bar{P}_b \approx 0.4$).

Although this discussion has centered on a simple two-level system, our conclusions about the influence of the intensity and pulse length are quite general. Further, our picture rigorously justifies the collective conclusions that have emerged from other CEP studies to date—namely, that a laser pulse must be both short and intense in order to produce significant CEP effects.

Being general, our picture applies equally well to processes with a continuum final state such as ionization and dissociation. The CEP effects in H_2^+ dissociation, for

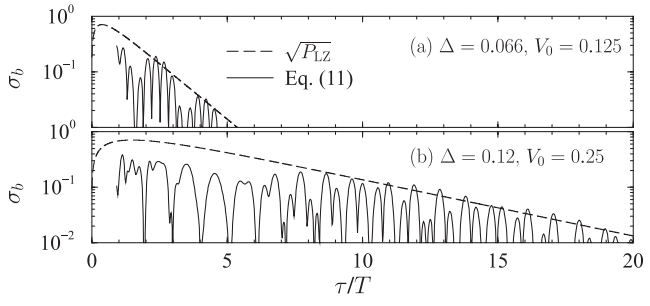

FIG. 2. Magnitude of the CEP effect σ_b for excitation in a two-level system as a function of the pulse duration in units of the laser period $T = 2\pi/\omega$ for two different sets of parameters.

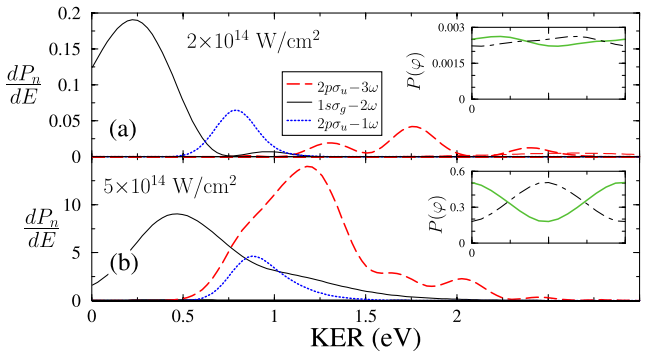

FIG. 3 (color online). Nuclear kinetic energy release (KER) spectra for each n-photon component $\tilde{\phi}_n(R, t')$ for dissociation of $H_2^+(v = 0)$ in a 10 fs, 790 nm laser pulse. The insets show the CEP effect on dissociation to $p + H$ and $H + p$.

instance, appear as asymmetries between the experimentally distinguishable $p + H$ and $H + p$ channels (see insets in Fig. 3) [3]. Since this asymmetry can only result from interference of the $1s\sigma_g$ and $2p\sigma_u$ molecular channels, the nuclei dissociating in these channels must contribute at the same kinetic energy [2]. Further, our formulation shows that each channel must also contribute with different numbers of photons—which is guaranteed by dipole selection rules. Figure 3 shows the kinetic energy distributions for the dominant n-photon states $\tilde{\phi}_n(R, t')$ from Eq. (5) including only the $1s\sigma_g$ and $2p\sigma_u$ channels and neglecting nuclear rotation. For strong, short pulses, Fig. 3(b), the laser bandwidth and intensity-induced shifting and broadening of the initial state [2] lead to the requisite overlap of nuclear wave packets. For weak or long pulses, Fig. 3(a), this overlap vanishes.

As a practical matter, this approach requires the n-photon amplitudes ϕ_n—either from Eq. (6) or by extracting them from solutions of the original TDSE (1). For the simple problems described in this Letter, the increased computational burden implied by Eq. (6) is not a barrier, and the former route is preferable. For more complex problems, this increase may simply not be possible. In this case, solving Eq. (1) for several φ and Fourier transforming the wave function using Eq. (9) will provide the necessary amplitudes to use the rest of our formalism. Moreover, given the relation (9) between n and φ, the number of φ required can be estimated *a priori* using the physics of the problem.

In summary, we have presented a formalism for treating and understanding CEP effects in short laser pulses. Our approach allows the full CEP dependence of any observable to be calculated from a single solution of the TDSE in a Floquet representation. Further, it clearly establishes the interpretation of short pulse CEP effects for *any* system as interference between different n-photon pathways. Because it is completely general, it incorporates previous work as special cases while simultaneously providing the qualitative framework to understand how the CEP effects in these disparate cases stem from the same underlying physics. Finally, the two-level example discussed demonstrates how our formulation can be used to obtain estimates of the pulse duration and intensity dependence, showing, in particular, that CEP effects can persist to surprisingly long pulse durations for a proper choice of parameters for the laser-matter system.

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