Heterodyne Mixing of Laser Fields for Temporal Gating of High-Order Harmonic Generation

Thomas Pfeifer,* Lukas Gallmann, Mark J. Abel, Phillip M. Nagel, Daniel M. Neumark, and Stephen R. Leone

Departments of Chemistry and Physics, University of California, Berkeley, California 94720, USA and Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

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The concept of heterodyne mixing of laser fields is theoretically applied to the process of high-harmonic generation to enhance and modulate the kinetic energy of the active electron on subcycle time scales. A very small amount of intensity in the heterodyne field creates a significant modification of the electron kinetic energy, due to its amplification by the strong fundamental field in the kinetic-energy term, in which the heterodyne mixing occurs. Quantum calculations are carried out to verify the predictions of the classical results, demonstrating very good qualitative and quantitative agreement. Applications of the heterodyne-mixing concept are the extension of the harmonic cutoff to higher photon energies and the temporal gating of attosecond pulse production.

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High-order harmonic generation (HHG) is one of the most intriguing processes in nonlinear optics, both from a fundamental physics perspective as well as for its applications. Applications of HHG now include the production of the shortest pulses of light, with durations less than one femtosecond [1,2], nonlinear optics with soft x-rays [3,4], and tomographic imaging of molecular orbitals [5]. On the fundamental side, HHG is a fascinating example of nonlinear physics, where simple pictures can be used to describe strongly nonperturbative processes. For example, the highly nonlinear process leading to the emission of high-energy photons in HHG can be conceptually understood by classical mechanics [6]: the outermost electron of an atom in a highly intense laser field is ionized, driven away from and back to its parent ion by the alternating electric field of the laser that accelerates it, eventually to recombine, emitting the sum of its kinetic energy and the ionization potential into a high-energy photon that is perfectly synchronized (coherent) with the laser field. In this Letter, we show that key aspects of HHG can be enhanced through heterodyne mixing of laser fields.

Heterodyne mixing is commonly used in various areas of science and engineering as a phase sensitive means of detecting and characterizing small oscillatory signals. Examples of applications in quantum optics are the measurement of the quantum state of light [7] and the spectral phase analysis of nonlinear optical signals [8]. The heterodyne technique is based on the superposition of a weak signal wave with a strong coherent reference wave, termed the local oscillator. Mixing of the two light fields occurs in the intensity of the combined fields by virtue of the quadratic dependence of the intensity on the total electric field. The cross term is a product of the field amplitudes of both the local oscillator and the signal field and it depends on their relative phase, resulting in a characteristic intensity beating if the two fields do not coincide in frequency. Since the intensity modulation is proportional to the strong local oscillator field, the heterodyne detection of the signal field can be far more sensitive than the signal field alone.

We present theoretical evidence that heterodyne mixing, when applied to HHG, leads to enhancement of the harmonic cutoff energy and efficient subcycle control of the active electron. The fundamental field acts as the local oscillator for the weak superimposed signal field. The relevant mixing of the two copolarized fields occurs through the kinetic energy of the active electron as opposed to the combined intensity of the two fields, which is the case in conventional heterodyne mixing. Because of the increased sensitivity of the heterodyne method, a low-intensity signal field at a different frequency is sufficient to result in significant enhancement and modulation of the kinetic energy of the returning electron. Since the heterodyne mixing of the two fields is sensitive to the relative phase of the fundamental (local oscillator) and signal fields, it breaks the dynamical symmetry that results in the half-cycle periodic emission of attosecond pulses in the case of a monochromatic fundamental. For the same reason, it is possible to employ the signal field (which we term control field in the following) as an efficient means for temporal gating of the harmonic emission, as will be detailed below. Harmonic heterodyne mixing is a new concept employed to understand bichromatic harmonic generation [9]. Moreover, the recently predicted phenomenological result of period doubling [10] in an attosecond pulse train by adding a second harmonic field to the fundamental field can be explained as a special case of the heterodyne-mixing mechanism considered here. The appearance of even-order harmonics reported in earlier studies of high-harmonic generation with two-color laser fields [11]—even at very low intensities of the second color field—can now be physically understood by the mechanism presented here.

We discuss the general concept of high-harmonic heterodyne mixing and present two important applications: HHG at photon energies exceeding the standard cutoff energy and the temporal gating of harmonic emission. The latter can lead to isolated attosecond pulses with femtosecond multicycle duration driver pulses that are

directly available from commercial Ti:sapphire chirped-pulse amplification systems, without the need for further pulse compression. Moreover, HHG is only one application of the presented heterodyne-mixing mechanism. The conceptual framework can be transferred to any process involving strong-field driven electron dynamics—including but not limited to nonsequential double ionization, above-threshold ionization, relativistic electron motion in ultraintense laser fields, and plasma dynamics—due to the classical nature of the control mechanism.

Considering first the quasiclassical picture of HHG [6], we can calculate the electron kinetic energy by integrating the classical equation of motion with the initial condition of the electron being at rest immediately after ionization that occurs at time t_0. The fundamental and control fields are $E_1(t) = \hat{E}_1 \cos(\omega_1 t)$ and $E_2(t) = \hat{E}_2 \cos(\omega_2 t + \varphi)$ [atomic units (a.u.) are used throughout], respectively, with amplitudes $\hat{E}_i$ and different frequencies ω_i. We limit ourselves to the case of a weak control field $\hat{E}_2 = a\hat{E}_1$, with $a \ll 1$. The relative phase between the two fields is denoted by φ. The classical velocity v of the electron at the time of recombination with the ion t_r is

$$v = -\int_{t_0}^{t_r} [E_1(t) + E_2(t)]dt = v_1 + v_2, \tag{1}$$

where v_1 and v_2 denote the velocity components due to acceleration in the fields $E_1(t)$ and $E_2(t)$, respectively, for an electron ionized at time t_0 and recombining at time t_r:

$$v_1 = \frac{\hat{E}_1}{\omega_1}[\sin(\omega_1 t_0) - \sin(\omega_1 t_r)] \tag{2}$$

$$v_2 = \frac{\hat{E}_2}{\omega_2}[\sin(\omega_2 t_0 + \varphi) - \sin(\omega_2 t_r + \varphi)] \tag{3}$$

Heterodyne mixing of the two fields occurs through the kinetic energy of the electron in the presence of the two fields, which can be written as

$$E_{\text{kin}} = \frac{1}{2}(v_1 + v_2)^2 = \frac{1}{2}(v_1^2 + v_2^2 + 2v_1 v_2). \tag{4}$$

While the kinetic energy of returning electrons produced by the control field alone ($v_2^2/2 = E_{\text{kin},2}$) can be very small compared to the contribution of the fundamental field alone ($v_1^2/2 = E_{\text{kin},1}$), the cross term ($v_1 v_2$) is significantly larger. This is the essence of the heterodyne effect: The cross term "amplifies" the small velocity component v_2 arising from the weak control field by the large velocity component v_1 originating in the strong local oscillator field of the fundamental. Note that the heterodyne mixing here occurs in the square of the velocity components (vector potentials or integrals of the electric fields) rather than the square of the electric fields of signal and local oscillator, as is the case in conventional heterodyne mixing.

In the following, we discuss the case of a control field taken as the subharmonic of the fundamental at one half of the fundamental frequency. Typically in HHG, the kinetic energy of the fastest electron and thus the cutoff energy of the high-harmonic spectrum is $E_c = I_p + 3.17U_p$ (I_p and U_p being the ionization potential and the ponderomotive potential $U_p = E^2/4\omega^2$, respectively). This quantity depends linearly on laser intensity and thus quadratically on the electric field of the light. However, for the case of kinetic-energy heterodyne amplification of a weak control field ($a \leq 0.1$), Eq. (4) indicates a linear dependence of the cutoff energy on the control field velocity v_2. Because of the linear dependence of the velocities v_i on the electric field amplitude $\hat{E}_i$ of the laser [see Eqs. (2) and (3)], we should therefore expect linear scaling of the cutoff energy with the weak laser control field amplitude $\hat{E}_2$, instead of its intensity. This is shown in Fig. 1(a), where the maximum kinetic energy of the returning electron is calculated classically and shown as a function of the ratio a of the control field with respect to the copolarized fundamental (local oscillator) field for a fixed relative phase of $\varphi = 0$. A clear linear dependence is visible. For larger control field amplitudes the time of ionization t_0 and recombination t_r become significantly dependent on the control field, resulting in a very slight deviation from linearity as can be seen in the figure.

To verify the results quantum mechanically, we perform a one-dimensional numerical integration of the time-dependent Schrödinger equation. The high-harmonic spectrum is obtained by Fourier transforming the time-dependent dipole acceleration expectation value according

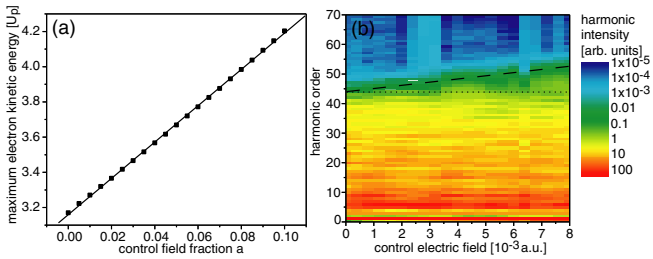

FIG. 1 (color online). Demonstration of linear heterodyne weak-field amplification of electron kinetic energy (a) and high-harmonic cutoff photon energy (b) by varying the control field strength. The weak control field is assumed to be the subharmonic of the fundamental field at half its frequency and at a relative phase of $\varphi = 0$. The classical calculation (a) shows a linear scaling of the energy of the fastest electron returning to the parent ion. The solid line is a linear fit to the calculated data points. The same linear scaling is apparent in the quantum simulation (b) (details see text), where the cutoff harmonic order (photon energy) (dashed line) scales linearly with the control electric field strength, which is varied up to 1/10th of the fundamental field strength. The horizontal dotted line shows the original position of the cutoff harmonic order at zero control field. While the ponderomotive potential of the largest control field is only a minor fraction (1/25) of the fundamental one, the changes in kinetic and cutoff energy beyond the ionization potential are ~30% due to the heterodyne mixing through the kinetic energy of the electrons, Eq. (4).

to Ehrenfest's theorem [12,13]. A model argon atom is considered to interact with the combined field of two Gaussian laser pulses of 24 fs FWHM (full width at half maximum) duration at a center wavelength of 800 nm (fundamental) and 1600 nm (control field). The field strength of the 800 nm pulse was kept constant at 0.08 a.u. and the field strength of the 1600 nm control field was varied from 0 to 8×10^{-3} a.u., the upper limit thus again corresponding to the case $a = 0.1$. Plotting the harmonic spectrum versus the control electric field [Fig. 1(b)] shows the linear dependence of the harmonic cutoff energy, clearly indicating the validity of the heterodyne effect as observed in the classical case. This effect can therefore be applied to extend the harmonic cutoff to higher photon energies without having to significantly increase the laser pulse energy (conversion of only $<0.1^2 = 1\%$ intensity to the control field is needed) or to decrease the pulse duration. At the same time, the experimental challenge to produce the second light field is minimal, since only a very small quantity of fundamental light has to be converted. The amount of 1% intensity is far below the typical conversion efficiency achieved in experiments on parametric down-conversion [14], especially at the degeneracy point ($\omega \rightarrow \omega/2$) considered in the above example.

We now turn to another application of this effect, which is the temporal gating [15] of high-harmonic generation. In Fig. 2(a) we plot the classically calculated kinetic energy of the electron that returns to the parent ion versus time of return. If only a single-frequency fundamental field is applied, the well-known [6] maximum of $\sim 3.17U_p$ is observed. If we consider conversion of 1% fundamental intensity into a subharmonic at half of the initial frequency with 0 relative phase and recalculate the kinetic energies

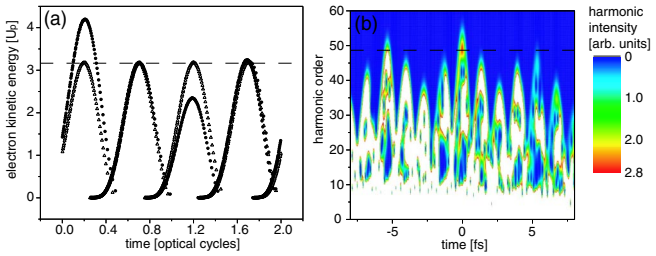

FIG. 2 (color online). Kinetic energy of the electrons returning to the parent ion as a function of time. A classical calculation is shown in (a) for the case of a single fundamental field (open triangles) and after conversion of 1% intensity into the subharmonic at half the fundamental frequency and a relative phase of 0 (solid dots). A clear symmetry breaking of the kinetic energy in adjacent cycles is observed, creating electron energies up to $4.2U_p$. The conventional (single-frequency limit) of $\sim 3.17U_p$ is indicated by the dashed line. A quantum-mechanical simulation (details see text) of HHG in argon atoms is performed to verify the validity of the classical picture (b). The Gabor transformed dipole acceleration response of the single-atom system shows that the photon energy emitted as a function of time is in very good agreement with the classical calculation.

for the returning electrons, a significantly different scenario is observed. The symmetry of adjacent half-cycles of the fundamental field is broken by the subharmonic field and the energies of the returning electrons are different in neighboring half-cycles. In particular, the kinetic energy of the fastest returning electrons occurs only one out of four fundamental half-cycles (corresponding to one optical cycle of the subharmonic) and has 32% higher electron energy upon return to the parent ion compared to the highest other half-cycles. According to the mechanism of HHG [6], this allows the production of the highest photon energies of harmonics only once within two optical cycles. This complete half-cycle symmetry breaking for the highest photon energies also provides physical insight into why even at low values of the field ratio $a \sim 0.1$ even harmonic orders emerge at equal strength in the cutoff region of the harmonic spectrum observed in earlier works [11].

To demonstrate the validity of the classical consideration, the single-atom high-harmonic emission is simulated for the same conditions as used in Fig. 1(b). To analyze the photon energy emitted from the atom as a function of time, we perform a Gabor transform (windowed Fourier transform) as was previously applied for the analysis of HHG [16,17]. It is defined as

$$G(t, \omega) = \left| \int_{-\infty}^{+\infty} \hat{a}(\omega') \exp\left(-\frac{(\omega' - \omega)^2}{\Omega^2}\right) \times \exp(i\omega' t) d\omega' \right|^2, \quad (5)$$

where $\hat{a}(\omega')$ is the dipole acceleration expectation value in the frequency domain and Ω is the spectral window width, which needs to be chosen large enough to obtain sufficient (subcycle) temporal resolution. We chose $\Omega = 2$ eV$/\hbar$ and the result is given in Fig. 2(b). Different maximum harmonic orders are produced in adjacent half-cycles, as is expected from the classical calculation. The agreement of the classical [Fig. 2(a)] and quantum-mechanical calculation [Fig. 2(b)] is remarkable and underlines once again the validity of the classical approach to understand high-harmonic generation. In addition, this calculation points out that even *control* of attosecond pulse generation can be understood from a classical perspective, which will help towards the future goal of inventing and applying control techniques in the soft x-ray attosecond region.

For the conventional method of isolated attosecond pulse generation, the femtosecond driver pulse needs to be short enough to permit only half an optical cycle to generate high-energy photons in the harmonic spectrum. High-pass filtering of the harmonic spectrum separates these photons to produce an isolated attosecond pulse from the attosecond pulse train consisting of lower-order harmonics [2]. Applying the heterodyning technique explained here, the restriction on the driver pulse duration can be relaxed by a factor of 4, since now only once within two fundamental cycles are the higher-energy harmonics emitted, using the weak subharmonic as a gating field. Instead

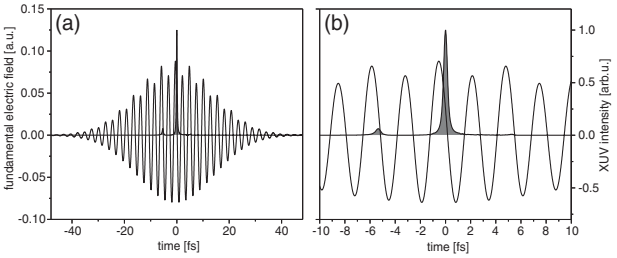

FIG. 3. Application of the heterodyne technique for temporal gating yields an isolated attosecond pulse (shaded area) produced with a field (solid line) consisting of an 800 nm fundamental field of 24 fs FWHM pulse duration with 1% intensity of the subharmonic at 1600 nm. The intensity of the weak satellite pulse is less by more than an order of magnitude. An enlarged view of (a) is shown in (b).

of having to use a ~6 fs pulse of the commonly used 800 nm central wavelength for isolated attosecond pulse production, a 24 fs pulse is sufficient if ~1% of the fundamental is converted to the 1600 nm subharmonic intensity. The consequence of this approach is that pulse compression methods would no longer be necessary for single attosecond pulse generation, since ~24 fs pulse durations are readily accessible directly from commercial Ti:sapphire multipass amplification systems.

Figure 3 shows the calculated result for the attosecond pulse emitted after high-pass filtering above a frequency corresponding to the 48th harmonic as indicated by the dashed line in Fig. 2(b). An isolated attosecond pulse of 370 as FWHM duration is generated, only accompanied by a minor satellite pulse which is less intense by more than an order of magnitude.

In conclusion, the concept of kinetic-energy heterodyne mixing of a weak control field with the fundamental field as a local oscillator is developed for high-harmonic generation. While the control field alone would not significantly accelerate the active electron in high-harmonic generation, the mixing and amplification of the weak field with the strong fundamental field in the electron kinetic-energy term allows for significant modulation of the kinetic energy of the returning electron, resulting in an enhancement of the harmonic cutoff energy. Another application of the mechanism is the temporal gating of high-harmonic generation, possibly enabling the production of isolated attosecond pulses with driving pulses that are longer than the conventional limit by a factor of 4. It should be noted that the case of the subharmonic field as the control field is just one example and the heterodyne-mixing process can be used with arbitrary frequencies that might be beneficial for producing and shaping attosecond pulses and pulse trains in the future. In most general terms, the presented heterodyne approach—due to its classical nature—can be applied in any strong-field process to control electron dynamics.

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*Electronic address: tpfeifer@lbl.gov

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