Ideal Waveform to Generate the Maximum Possible Electron Recollision Energy for Any Given Oscillation Period

L. E. Chipperfield,* J. S. Robinson, J. W. G. Tisch, and J. P. Marangos
Imperial College London, London SW7 2BW
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We present the perfect waveform which, during a strong field interaction, generates the maximum possible electron recollision energy for any given oscillation period, over 3 times as high as that for a pure sinusoidal wave. This ideal waveform has the form of a linear ramp with a dc offset. A genetic algorithm was employed to find an optimized practically achievable waveform composed of a longer wavelength field, to provide the offset, in addition to higher frequency components. This second waveform is found to be capable of generating electron recollision energies as high as those for the perfect waveform while retaining the high recollision amplitudes of a pure sinusoidal wave. Calculations of high harmonic generation demonstrate this enhancement, by increasing the cutoff energy by a factor of 2.5 while maintaining the harmonic yield, providing an enhanced tool for attosecond science.

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Following strong field ionization in an intense laser field, the laser driven electron can recollide with its parent ion with a kinetic energy orders of magnitude greater than the photon energy of the field [1,2]. These recollisions are highly localized in both space and time. Therefore, combined with their high recollision energies, which can approach the keV range [3], they are an ideal tool for probing attosecond time scale electron dynamics, either directly [4–6] or through the attosecond light pulses they generate [7], and for mapping out the field of the drive pulse [8,9]. Increasing the maximum electron recollision energy without reducing the recollision amplitude is a challenge of great importance to attosecond science [10,11]. It is important for methods utilizing the electron recollision itself for probing attosecond electron dynamics [4] as well as for the generation of higher energy harmonics, which has applications to time-resolved extreme ultraviolet spectroscopy [12] and attosecond pump-probe experiments [13].

For a monochromatic wave the peak electron recollision energy depends on the laser intensity, I, and wavelength, λ, scaling as $\propto I\lambda^2$. Increasing the intensity of the driving laser pulse provides a relatively simple route to increasing the recollision energy. However, photoionization depletion of the ground state provides a fundamental limit to the maximum energies that can be achieved before the recollision amplitude drops exponentially. This limit is present even for few-cycle pulses [14]. Alternatively, the use of longer wavelength fields has been identified as a promising route to increasing the electron recollision energy. Longer wavelength driving fields, however, have the serious disadvantage that the harmonic yield is greatly reduced compared to shorter wavelengths. This reduction is a consequence of the increased trajectory duration, $\tau \propto \lambda$, which results in extra electron wave packet spreading. This reduces the overlap with the ground state upon recollision, $\sim\tau^{-3}$, and contributes toward an unfavorable scaling of harmonic yield (e.g., $\lambda^{-5} - \lambda^{-7}$ scaling has been reported [15,16]).

To overcome these fundamental limits the efficiency with which the laser field energy is used to transport and accelerate the recolliding electron must be increased, without increasing the time the electron spends in the continuum. This can be achieved by intracycle shaping of the laser waveform. For example, small gains are found through the addition of a second harmonic, which can deform the waveform such that the peak electron recollision energy is increased by 13% [17,18], and similar gains can also be generated through the addition of a weak low-frequency field [19]. However, in both cases this is accompanied by an increase in the excursion time and a reduction in the ionization rate.

In this Letter, we take a first principles approach to determining the best waveform for driving efficient electron recollisions. We report two new results of importance to high harmonic generation (HHG) light sources and attosecond technology: (i) we identify the ideal waveform that provides the highest electron recollision energy for a fixed oscillation period and a fixed average power over that period, leading to recollision energies a factor of 3.11 larger than a sinusoidal wave; (ii) we find a practically realizable route to synthesizing a waveform that produces a similar enhancement in the recollision energy while maintaining a high HHG yield compared to a pure sinusoidal field.

The peak electron recollision energy generated by any particular waveform will be raised by increasing its period T, or the field fluence per period, F, given by

$$F = \int_0^T I(t)dt = \frac{\epsilon_0 c}{2}\int_0^T E(t)^2 dt, \quad (1)$$

where ϵ_0 is the permittivity of free space and c is the speed of light. Therefore, the waveform which most efficiently uses the field energy to accelerate the recolliding electron will maximize the recollision energy for any given T and F. The maximum recollision energy, $\mathcal{E}$, is calculated using the classical three-step model [2,20], in which the strong field approximation is taken. Once ionized at time $t_I = 0$, the electron propagates as a free particle driven solely by the electric field of the laser, $E(t)$. The electron's velocity upon ionization is zero, $v(t_I) = 0$, and the recollision time, $t_R \leq T$, is defined to occur when, and if, the electron returns to its origin, $x(t_R) = x(t_I)$.

By employing the above considerations, a formal analysis [21], or numerical optimization [22], leads to a waveform consisting of a linear ramp with a dc offset,

$$E(t) = \pm\sqrt{\frac{F}{T}\frac{2}{\epsilon_0 c}}\left(\frac{3t}{T} - 1\right) \quad \text{for } 0 \leq t \leq T, \tag{2}$$

as shown in Fig. 1(a). This perfect wave drives the electron along a trajectory, with $t_I = 0$ and $t_R = T$, that generates the maximum recollision energy possible for any given period, T, and per period fluence, F.

The maximum electron recollision energy generated by the perfect waveform is

$$\mathcal{E}_{\text{opt}} = \frac{e^2}{m}\frac{FT}{4\epsilon_0 c}. \tag{3}$$

For a monochromatic wave with a period of T, given by $E(t) = E_0 \cos(2\pi t/T)$, for which the fluence per cycle is $F = \epsilon_0 c E_0^2 T/4$, the maximum recollision energy generated is

$$\mathcal{E}_M = 3.17 U_P = \frac{3.17}{\pi^2}\frac{e^2}{m}\frac{FT}{4\epsilon_0 c}, \tag{4}$$

where $U_P = (eE_0T/4\pi)^2/m$ is the ponderomotive potential. Therefore, compared with a monochromatic field with the same fluence per cycle, the perfect wave generates electron recollisions with over 3 times as much energy,

$$\mathcal{E}_{\text{opt}} = \frac{\pi^2}{3.17}\mathcal{E}_M \simeq 3.11\mathcal{E}_M. \tag{5}$$

The waveform that most efficiently uses the field energy to accelerate an electron is a dc field. However, such a waveform does not satisfy the condition $x(t_R) = x(t_I)$; to do so the electron must first be transported away from the core before it can be accelerated back again. Consequently, one may consider the electron's excursion in the continuum as consisting of three sections: (i) acceleration away from the core, (ii) deceleration to a standstill at some distance from the core, (iii) acceleration back into the core. The electron recollision energy is determined solely by the integral over this final section of the electric field. The perfect wave, given in Eq. (2), maximizes this integral primarily by minimizing the amount of field energy used within the first two sections for transporting the electron away from the core. It achieves this because the field energy used to accelerate the electron back to the core is concentrated close to the time of recollision, maximizing the amount of energy transferred to the electron while minimizing the distance it travels. Conversely, the field energy used to accelerate the electron away from the core is concentrated close to the time of ionization, minimizing the amount of energy consumed while maximizing the distance traveled. This can be illustrated by comparing the perfect wave to a sinusoidal wave with a period of $1.54T$, plotted in Fig. 1(b) along with the trajectory taken by its highest energy recolliding electron. In both cases the trajectory duration and fluence within the interval $0 < t < T$ are the same; however, the recollision energy for the perfect wave is higher. This is because the field energy of the sinusoidal waveform is not concentrated close to the time of recollision. Consequently, the electron must be transported farther from the core, 27% farther than for the perfect wave, which consumes a higher fraction of the field energy and reduces the final velocity of the recolliding electron.

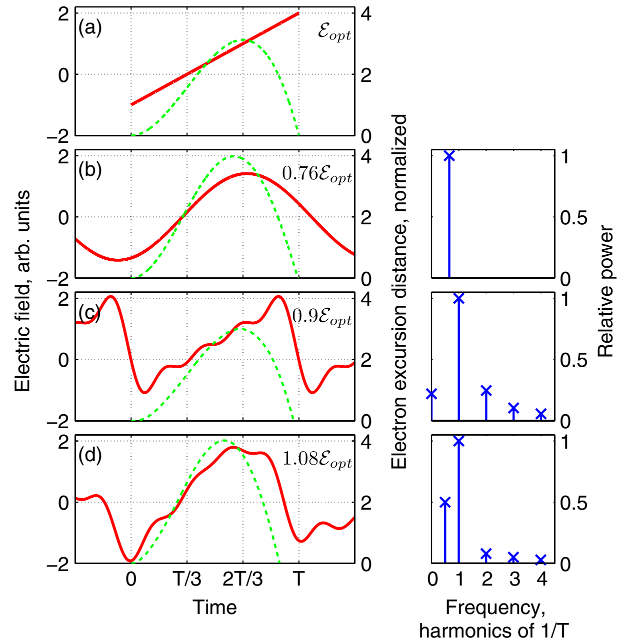

FIG. 1 (color online). The waveforms (solid lines) along with the most energetic recolliding electron trajectories they generate (dashed lines) are plotted on the left with the recollision energy displayed in the top right corner. Where appropriate, the waveform's spectrum is plotted on the right. The waveforms plotted are (a) the perfect wave; (b) a sinusoidal wave with a period of $1.54T$, chosen so that the highest energy trajectory has a duration of T; (c) a sawtooth waveform generated using the first four harmonics of $1/T$ plus a dc field; (d) the optimized waveform found using a genetic algorithm, maximizing both the field amplitude at $t = 0$ and the recollision energy, given the first four harmonics of $1/T$ and a field with a period of $2T$. The average power is fixed for all waveforms, and the fluence within the period $0 < t < T$ is also equal for (a), (b), and (c). However, for the optimized waveform in (d) the fluence within this period is 1.5 times that of the others, due to beating between the $1/T$ and $1/2T$ frequency components, leading to a recollision energy which is greater than (a).

There are two main challenges hindering the creation of the perfect wave: the large comb of frequencies required to generate a linear ramp, and the presence of a very high dc field required for the asymmetry in the field amplitude. Fortunately, neither of these is essential for generating recollision energies very close to the optimum. To find a practically achievable solution, a genetic algorithm [23] was used to converge upon the optimum waveforms, given a finite number of frequency components. The conditions used to derive Eq. (2) were maintained, with the fluence, F, within the interval T fixed and the fitness taken to be the electron recollision energy. Given the first four harmonics of $1/T$ plus a dc field this algorithm produces a sawtooth-like waveform, plotted in Fig. 1(c). Although this waveform is not linear, the recollision energy is very close to the optimum, primarily because the field energy is still concentrated close to the time of recollision. To remove the need for a dc field, a field with a period significantly greater than T is required. It is particularly beneficial to use a field with a period of $2T$, because the separation between its extrema of opposite polarity is T, which is a requirement of the perfect wave. For $T \sim 2.7$ fs this corresponds to a field with $\lambda \sim 1600$ nm. Few-cycle pulses of such fields have been generated with energies high enough to produce intensities of over 10^{15} W cm^{-2} [24]. An added advantage of combining a field with a period of T with another of $2T$ is that the beat frequency between them can act to concentrate the field fluence into alternate cycles.

To generate electron recollisions with high probabilities the field needs to be sufficiently intense to drive the ionization that initiates the recolliding electron. To prevent depletion before recombination the remainder of the field must, therefore, be weaker than at the time of ionization. Unfortunately, this condition is not satisfied by the perfect wave, for which the amplitude at the time of ionization is half that reached before recombination [25]. To avoid this problem a deviation from the perfect wave is required: the first section of the field must be deformed into a peak around the time of ionization, at the expense of the amount of field energy used to accelerate the electron back to the core. To determine such a waveform the field amplitude at the time of ionization was incorporated into the fitness function of the genetic algorithm. This algorithm was used to find the optimum waveform given the first four harmonics of $1/T$ and a field at half the fundamental frequency, $1/2T$, limited to half the power of the fundamental, to replace the dc component. Because of the inclusion of a field with a period of $2T$, normalizing the fluence during $0 < t < T$ is no longer suitable, so it is normalized over the period $0 < t < 2T$ instead. The optimized waveform returned is displayed in Fig. 1(d), with the phase and relative fluence of each component given in Table I. Recent advances in synthesizing ultrashort phase stable pulses across the UV and IR (see, for example, [26,27]) illustrate the feasibility of this approach. Note that for long pulses the absolute time delay between the envelope and carrier waveform is unimportant. Therefore, so long as the relative time delays between the component carrier waves are constant, carrier-envelope phase stabilization is unnecessary.

The second main result of this Letter is summarized in Fig. 2(a). A single active electron calculation of the high harmonic spectrum, including the effects of wave packet spreading and neutral depletion through ionization, was performed for a number of waveforms for neon, with an $I_P = 21.6$ eV, using the strong field approximation [28]. The same pulse duration of 16 fs full width at half maximum (FWHM) intensity was used for all waveforms, and the total laser energy was fixed equal to that of a pure sinusoidal pulse with a peak intensity of 3×10^{14} W cm^{-2}. The HHG spectra from pure sinusoidal fields at wavelengths of 800 nm, 1200 nm, and 1600 nm are shown as a solid black line, solid light gray (green) line, and dashed (pink) line, respectively. The cutoffs of these spectra increase $\propto \lambda^2$ from 79 eV for the 800 nm pulse to 250 eV for the 1600 nm pulse, as expected. This is accompanied by a steep drop in harmonic yield, $\propto \lambda^{-7}$ for the cutoff harmonics. In contrast, the optimum synthesized waveform, shown in Fig. 1(d), gives rise to an HHG spectrum, solid dark gray (red) line, with a cutoff at 198 eV, a factor of 2.5 greater than that generated by the 800 nm waveform but without an associated drop in yield. This increase is equivalent to lengthening the driving wavelength from 800 nm to 1430 nm, which would increase the travel time of the electron trajectory by a factor 1.8. For the optimized waveform, on the other hand, the trajectory duration increases by a factor of only 1.3. Because of wave packet spreading the recollision amplitude is highly sensitive to the electron excursion time, $\sim\tau^{-3}$, so this corresponds to a significant enhancement in the harmonic yield compared to a 1430 nm field. Alternatively, the waveform composed of just the 800 nm and 1600 nm field, without the higher harmonics, generates a spectrum, dotted (blue) line, with a yield

TABLE I. The relative fluence and phases of the frequency components for the optimized waveform in Fig. 1(d). The actual energies used for the propagation calculation, which generated the dark gray (red) HHG spectrum in Fig. 2(b), are presented in the fourth column.

Frequency	Fluence, fraction of total	Phase, in radians	Actual energies used for the propagated pulse
0.5	0.30	3.0	58 μJ (1600 nm)
1	0.60	0.0	91 μJ (800 nm)
2	0.048	-1.9	6.7 μJ (400 nm)
3	0.031	2.2	4.3 μJ (267 nm)
4	0.017	0.1	2.4 μJ (200 nm)

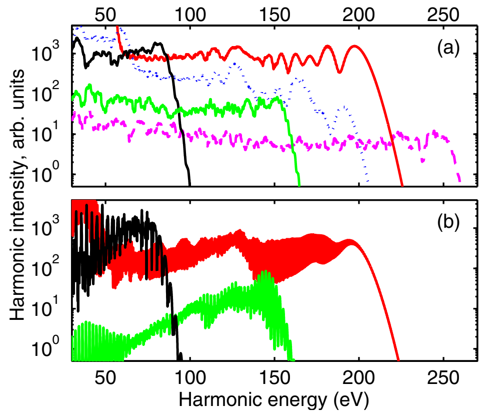

FIG. 2 (color online). Calculated HHG spectra generated by pure sinusoidal fields with wavelengths of 800 nm plotted as a solid black line, 1200 nm as a solid light gray (green) line, and 1600 nm as a dashed (pink) line; the optimized waveform plotted as solid dark gray (red) line; and a field composed solely of the 800 nm and 1600 nm components of the optimized field plotted in a dotted (blue) line. All spectra were generated using pulses of the same duration and pulse energy, normalized to a 16 fs FWHM 800 nm pulse with a peak intensity of 3×10^{14} W cm^{-2}. (a) Single active electron calculations. For clarity a 5 eV wide moving average has been used. (b) Full propagation calculations after radial integration.

approximately 10 times lower than the pure 800 nm field. Therefore, the optimized waveform is an important result, as it demonstrates that by using a synthesized waveform, which can be generated with current laser technology, the electron recollision energy can be more than tripled without reducing the harmonic yield.

As HHG is inevitably a multiatom process, it is necessary to take the effects of phase matching into account. To this end the propagation of the fields through a Ne gas jet was simulated, assuming cylindrical symmetry, by factorizing the second order wave equation to efficiently implement a forward-only approximation (see, e.g., [29]). All of the frequency components were focused at the same point in space with beam waists of $w_0 = 40\ \mu$m and 16 fs FWHM intensity profiles. The gas jet was placed 2 mm downstream from the focus and had a $\cos^2$ density profile with a FWHM of 0.5 mm and a peak density of 10^{16} cm^{-3}. The amplitudes and phases of the fields were such that on axis at the center of the gas jet they formed a waveform with a total fluence equal to that of a sinusoidal 800 nm field with the same pulse duration and a peak intensity of 3×10^{14} W cm^{-2}. As the shorter wavelengths diffract less than the longer wavelengths, they need less energy to generate the required intensity within the gas jet. The actual energies used for the optimized waveform are shown in the fourth column in Table I. The results are presented in Fig. 2(b). Comparing the HHG spectrum generated by the optimized waveform with those generated by the 800 nm and 1200 nm sinusoidal fields reveals that propagation preserves the gains shown in Fig. 2(a), demonstrating a robustness to variations in the amplitude and phase of the field components.

In conclusion, for any given trajectory duration, we have found, analytically, the optimum waveform which maximizes the electron recollision energy: this is a linear ramp with a dc offset. A field composed of repeating units of this waveform generates recollision energies over 3 times greater than those of a monochromatic field with the same oscillation period and average power. Aided by a genetic algorithm we have found a waveform that approximates this optimum field by utilizing harmonics up to the fourth order plus a field with twice the wavelength. This waveform generates electron recollision energies as high as the optimum waveform, while maintaining the same harmonic yield as a pure sinusoidal pulse with the same periodicity.

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*Luke.Chipperfield@imperial.ac.uk

[1] J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).
[2] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[3] J. Seres *et al.*, Nature (London) **433**, 596 (2005).
[4] J. Itatani *et al.*, Nature (London) **432**, 867 (2004).
[5] S. Baker *et al.*, Science **312**, 424 (2006).
[6] R. Torres *et al.*, Phys. Rev. Lett. **98**, 203007 (2007).
[7] M. Uiberacker *et al.*, Nature (London) **446**, 627 (2007).
[8] E. Goulielmakis *et al.*, Science **305**, 1267 (2004).
[9] C. A. Haworth *et al.*, Nature Phys. **3**, 52 (2007).
[10] T. Pfeifer *et al.*, Chem. Phys. Lett. **463**, 11 (2008).
[11] M. F. Kling and M. J. Vrakking, Annu. Rev. Phys. Chem. **59**, 463 (2008).
[12] E. Seres and C. Spielmann, Appl. Phys. Lett. **91**, 121 919 (2007).
[13] S. X. Hu and L. A. Collins, Phys. Rev. Lett. **96**, 073004 (2006).
[14] A. Gordon and F. Kärtner, Opt. Express **13**, 2941 (2005).
[15] J. Tate *et al.*, Phys. Rev. Lett. **98**, 013901 (2007).
[16] P. Colosimo *et al.*, Nature Phys. **4**, 386 (2008).
[17] J. Mauritsson *et al.*, Phys. Rev. Lett. **97**, 013001 (2006).
[18] Z. Zeng *et al.*, Phys. Rev. Lett. **98**, 203901 (2007).
[19] T. Pfeifer *et al.*, Phys. Rev. Lett. **97**, 163901 (2006).
[20] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett. **70**, 1599 (1993).
[21] L. E. Chipperfield (to be published).
[22] S. B. P. Radnor, L. E. Chipperfield, P. Kinsler, and G. H. C. New, Phys. Rev. A **77**, 033806 (2008).
[23] S. Forrest, Science **261**, 872 (1993).
[24] C. Vozzi *et al.*, Opt. Lett. **32**, 2957 (2007).
[25] These waveforms can still be of practical use if an appropriately timed attosecond duration soft x-ray pulse is applied at $t = 0$ to drive the ionization step.
[26] G. Cirmi *et al.*, J. Opt. Soc. Am. B **25**, B62 (2008).
[27] P. Tzankov *et al.*, Opt. Lett. **31**, 3629 (2006).
[28] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[29] G. Genty, P. Kinsler, B. Kibler, and J. M. Dudley, Opt. Express **15**, 5382 (2007).