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CAN ELECTRONIC WAVEPACKETS GENERATE ATTOSECOND PULSES OF LIGHT?[1]

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We investigate quantum mechanical effects in the recollision picture of high–harmonic generation, relate these to its classical counterpart and discuss the generation of attosecond pulses from recollision Bremsstrahlung.

1. Introduction

The interaction of atoms with intense laser light has been a rich area of research with many important discoveries [1]. One of th major findings has been high–harmonic generation (HHG); in this process the strongly driven atomic system reradiates energy at odd multiples of the driving frequency. Using HHG it has been possible to generate very short wavelength light upto XUV frequencies [2-6] with excellent coherence properties and with the potential for many applications.

All HHG spectra show the same generic behaviour: there is a sharp fall from the driving frequency fundamental peak to a plateau, followed by a cut–off for the maximal attainable energy of coherent light. The sharp fall is typically from the fundamental to the 7^{th} or 9^{th} harmonic, while the plateau extends out to very high harmonics (e.g. the 135^{th}). A quasi–classical approach first proposed by Kulander, Schafer and Krause and by Corkum [7,8] was found to predict accurately the cut–off energy as $I_p + 3.17U_p$, where I_p is the ionization potential of the atom and U_p the ponderomotive energy of the electron in the incident field. The ponderomotive energy is associated with the quiver motion of a free electron in a strong laser field E of frequency ω:

$$U_p = \frac{e^2E^2}{4m\omega^2} \tag{1}$$

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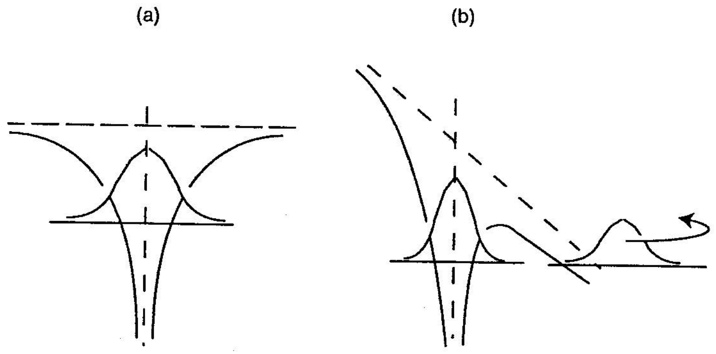

Fig. 1. Bound atomic wavepacket (a) in a Coulombic potential and (b) after (1/4) cycle application of an intense field, so that a tunnelling wavepacket escapes, but is ponderomotively accelerated back towards the atomic core.

and if the intensity I is expressed in Wcm^{-2} and the wavelength of the laser field λ in μm, then $U_p \sim 10^{-13} I(Wcm^{-2})(\lambda(\mu m))^2$. The laser field will dominate the dynamics of a bound electron if its electric field E exceeds the Coulomb binding field $E_C = e/4\pi\epsilon_0 a_0^2$, where a_0 is the Bohr radius; the Coulomb field experienced by an electron in the ground state of atomic hydrogen is $5.14 \times 10^9 Vcm^{-1}$, equivalent to an atomic unit of intensity $3.51 \times 10^{16} Wcm^{-2}$.

An initial atomic wavepacket representing the ground state wavefunction can tunnel [9,10] out of the Coulomb potential well if a sufficiently strong laser field is applied (Fig. 1). The quasi-static tunnelling rate $\Gamma(t)$ is given by [11]

$$\Gamma(t) \sim (I_p/E(t)) \exp\{-2/3E(t)\}. \tag{2}$$

In a very strong laser field, the tunnelling wavepacket quivers and recollides with the atomic core; this happens repeatedly every half-cycle. The collisional encounter with the core creates such strong acceleration that it radiates; the repeated recollisions every half-cycle ensure harmonics are generated by the periodic encounters, whilst the brevity of each encounter guarantees a broad spectral range for the harmonic emission. A simple classical model shows that the maximum kinetic energy allowed for a recolliding electron is $3.17U_p$, so that the cut-off rule can be interpreted in terms of the maximum energy a recolliding electron can give up in radiation: the kinetic energy plus the recombination ionization potential energy I_p.

2. The Recollision Picture

The recollision picture is quasi-classical in that the ionization process is purely quantum mechanical; however, once ionized the electron wavepacket is treated as a free electron

in a laser field. Harmonics are then generated by a sequence of single collisions of these electron wavepackets formed by the tunneling process near the peaks of the incident laser electric field [12]. One has for the complete electron acceleration

$$a(t) = \sum_{l=1}^{2N} a_l(t) \tag{3}$$

where $2N$ is the total number of collisions during an N-cycle laser pulse. The individual component accelerations $a_l(t)$ over half a cycle and including one recollision, are assumed to be identical, but with different phases due to the different birth times of the wavepackets, i.e., $a_l(t) = a_B(t)e^{il\pi}$, for $t_{l-1} < t < t_l$, and zero otherwise. The l-th collision occurs between the times t_{l-1} and t_l, where $t_l = lT/2$ and T is the laser period. The Fourier amplitude from the initial time t_0 to the final time t_f, is then

$$\tilde{a}(\omega) = \int_{t_0}^{t_f} dt\, e^{i\omega t}\, a(t) = \sum_{l=1}^{2N} e^{i\omega t_{l-1}}\, e^{il\pi}\, \tilde{a}_B(\omega) \tag{4}$$

Here, $\tilde{a}_B(\omega)$ is the Fourier amplitude of the acceleration during a single collision event,

$$\tilde{a}_B(\omega) = \int_0^{T/2} d\tau\, e^{i\omega\tau}\, a_B(\tau), \tag{5}$$

i.e. it is the strength of the Bremsstrahlung radiation at frequency ω [13,14]. Then the spectrum is:

$$S(\omega) = |\tilde{a}(\omega)|^2 = 2N|\tilde{a}_B(\omega)|^2 \left\{ 1 + \frac{1}{N} \sum_{\substack{n,m=1 \\ n<m}}^{2N} \cos\left[\left(\frac{\omega}{\omega_L} + 1\right)(n-m)\pi\right] \right\} \tag{6}$$

where $\omega_L = 2\pi/T$ is the frequency of the incident laser. For odd harmonics, $\omega/\omega_L = 2s+1$, where $s = 0, 1, 2, \ldots$ one finds maxima $S[(2s+1)\omega_L] \sim N^2|\tilde{a}_B(\omega)|^2$, while for even harmonics the sum in Eq.6 tends to zero for large N, yielding $S(2s\omega_L) \sim N|\tilde{a}_B(\omega)|^2$. This would mean that as the number of collisions increases then the spectrum would become better defined.

In the above, we have assumed that the recollisions are identical to each other. This is appropriate for a constant amplitude laser field excitation, but not for pulsed excitation, where the quiver excursion amplitude and frequency varies throughout the pulse. We have examined the case of pulsed excitation using a generalization of the above model [15] and show how blueshifts of the harmonics [16] and a smearing-out of individual components results from this modification of the periodicity.

3. Classical Recollision

In order to obtain insight into the underlying physics of the recollision process we consider it both in the classical and then in the quantum mechanical picture. For our classical calculations we start from the classical Hamiltonian [17,18] :

$$H = \frac{1}{2}m(\frac{dq(t)}{dt})^2 - eE_0 q(t)\cos(\omega t) - \frac{e}{\sqrt{1+q(t)^2}} \tag{7}$$

where q, e and m represent the electron position, charge and mass and E_0 the maximal electric field strength. From this we obtain a Newtonian equation of motion for an electron moving in the laser field and soft-core potential

$$m\frac{d^2q(t)}{dt^2} = e\left[E_0\cos(\omega t) - \frac{eq(t)}{(1+q(t)^2)^{3/2}}\right]. \tag{8}$$

This equation of motion requires two initial conditions. Insight into the dynamics of the recollision picture is gained by eliminating the ionization process: we therefore start with an electron initially some distance away from the *bare* atomic core with initial conditions such that no drift is imposed on the electron motion. This can be realized for example by placing the electron at the classical turning point of the ponderomotive motion with velocity zero for the initial laser phase as chosen in the above Hamiltonian. We then follow this system under the influence of a monochromatic laser field which involves various encounters of the electron with the nucleus and evaluate the acceleration and the radiation at the various stages in time. In Fig. 2 we show what happens after 1, 2 and 8 collisions. As expected from our previous calculations we find Bremsstrahlung after the first encounter and with increasing number of collisions interference shapes this into a proper harmonic spectrum.

However, to truly model a wavepacket an ensemble number of these individual classical electron trajectories must be considered and the results averaged. This approach is able to model wavepacket spreading but is, of course, unable to simulate quantum mechanical interference effects. In Fig. 3 we have the calculated classical HHG spectrum for $E_0 = 0.1a.u.$, $\omega = 0.038a.u.$ and an initial gaussian wavepacket simulated by an ensemble in phase space centered at $70a.u.$ away from the core in the spatial direction with width $10a.u.$ and at momentum 0 with width $0.1a.u.$, in a field which is phased such that it initially accelerates towards the core with maximal strength. This should produce a spectrum which has a cut–off at $I_p + 2U_p$ i.e. at the 91st harmonic. The reason why this is determined by $2U_p$, rather than $3.17U_p$ is that starting in such an initially–displaced position generates such a quiver energy for the returning electron. This cut–off rule is approximately true; the cut–off however is far less pronounced than in the quantum calculations which follow.

4. Quantum Wavepacket Recollisions

To investigate the quantum mechanical effects associated with the recollision process we have chosen to integrate the Schrödinger equation. This is achieved by transforming into the rest frame of the electron, or the Kramers–Henneberger (KH) frame, which significantly simplifies the numerics and can also provide a useful handle on the behaviour of the system [19,20]. Within the KH frame the split–step operator method is used to evolve the initial state under the chosen pulse. It is then possible to extract time–dependent quantities such as the dipole acceleration (which is used to calculate

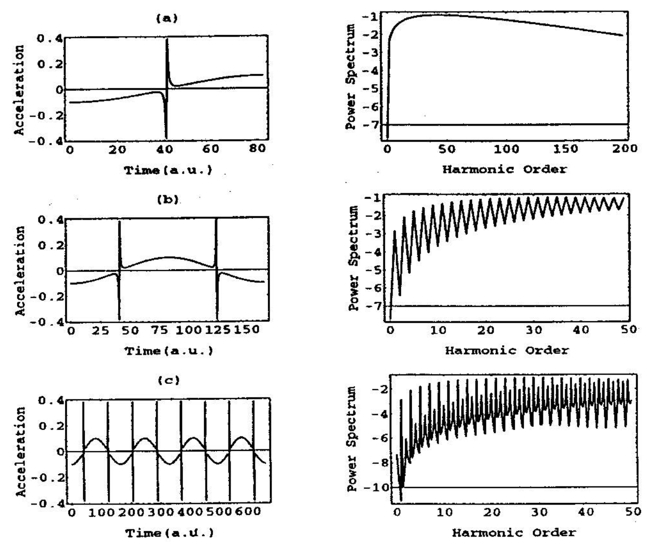

Fig. 2. The dipole acceleration of a single electron with the corresponding power spectrum on a logarithmic scale for (a) 1, (b) 2 encounters and (c) 8 encounters with the nucleus. The electric field strength was 0.1 a.u., the angular frequency 0.038 a.u. and the electron was initially placed 70 a.u. away from the nucleus with zero velocity. Note that for all the power spectra the fundamental frequency elastically radiated directly from the laser field has been removed.

the harmonic emission [21]), or ionization from the time–dependent wavefunction. Because of the Coulomb singularity it is not possible to use the Coulomb potential in a one dimensional approach, however, a smoothed potential,

$$V(x) = -\frac{1}{\sqrt{1+x^2}} \tag{9}$$

can be used, which behaves asymptotically like the Coulomb potential, but with a binding energy of -0.67 a.u. compared to -0.5 a.u. for the real hydrogen atom.

Assuming the same parameters and initial condition as our classical study we find an interesting wavepacket evolution which is shown in Fig. 4 . As expected the wavepacket

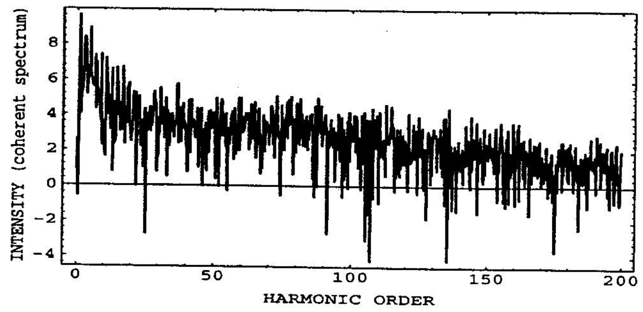

Fig. 3. The classical Monte Carlo spectrum for a laser field of electric field strength $E = 0.1a.u.$, frequency $\omega = 0.038a.u.$ with a 1D "soft" atomic core and an electron in an initial gaussian wavepacket simulated by an ensemble in phase space centered at $70a.u.$ away from the core in the spatial direction with width $10a.u.$ and at momentum 0 with width $0.1a.u.$. The laser field is phased initially such that it the electrons initially accelerates towards the core with maximal strength.

spreads and encounters the core; however, for the first few encounters the quantum mechanical interference is not very large and these collisions are 'classical–like'. However, once the wavepacket has spread enough, the core interaction leads to the wavepacket acquiring sharp, fine–structured features. The underlying physics of the additional structures can be understood as arising from the interferences between those parts of the wavepacket which are *reflected* by the core and those parts which are still incoming. This is a purely quantum mechanical effect and is missing from the previous classical treatment.

In order to highlight the importance of quantum mechanical interference, we show in Fig. 5 the calculated dipole acceleration. After the first three collisions, the accelel-eration resembles quite closely that of a single electron trajectory, with, however, a fast amplitude decay due to wavepacket spreading. A windowed Fourier transform of the first collision shows that the associated power spectrum is that of Bremsstrahlung (Fig. 5(a)). After the second collision, we see the beginning of harmonic structure (Fig. 5(b)) but only for the low order harmonics (the wavepacket spreading is the cause for this Bremsstrahlung mechanism being inefficient). Only when the high frequency

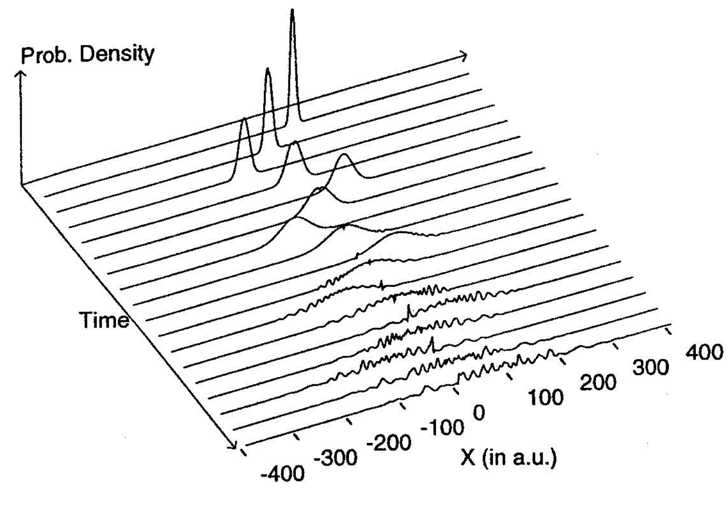

Fig. 4. The quantum mechanical evolution of the wavepacket for the same parameters as for Fig. 3, but taken over 4 full cycles of the incident laser field. The first snapshot is the initial wavepacket of width 10 a.u., and consequent snapshots are taken every quarter of a cycle.

components of the dipole acceleration are included do we begin to see anything resembling a true harmonic spectrum (Fig. 5(d)). If we take the spectrum of the whole dipole acceleration, this now has all the features associated with standard HHG (Fig. 5(f)). It is possible to observe, following the trend from (Fig. 5(a)) to (Fig. 5(f)), how the overall efficiency is enhanced, producing a very *clear* cut–off around the expected harmonic order. Thus, essentially classical dynamics affects the spectrum during the first few encounters of the electron wavepacket with the nucleus; quantum interference effects become crucial once the width of the wavepacket has reached a certain extent.

It would seem that to obtain a 'good' harmonic spectrum it is important to have these fast oscillations in the dipole acceleration. The question is how do they occur. The answer lies in the wavepacket evolution (Fig. 4) and the form of the dipole acceleration operator [21],

$$\langle a(t)\rangle = -\int \frac{\partial V(x)}{\partial x}|\psi(x,t)|^2\, dx, \tag{10}$$

where $V(x)$ is the atomic potential and $\psi(x,t)$ the wavefunction. A highly structured spatial wavepacket generates via Eq.(10) high temporal frequencies [22,23] in the radiated field. Therefore, any wavepacket with many features will produce an acceleration which has fast oscillations and, consequently, efficient harmonic generation. These spa-

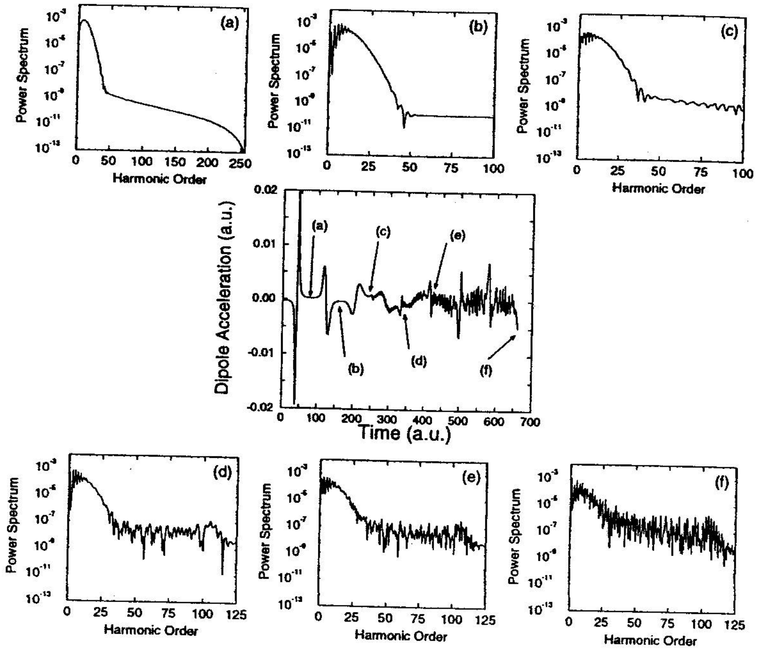

Fig. 5. The dipole acceleration (center) and the corresponding power spectra at various points in time as indicated by the labels (a) - (f). The parameters are the same as in Fig. 4 .

tial features only occur when the previously mentioned interference between incoming and outgoing parts of the wavepacket takes place. This idea can be corroborated by observing that the fast oscillations in the dipole acceleration appear at the same time as the spatial features in the wavepacket. If it were possible to control this interference it would provide a very powerful way of controlling the harmonic spectrum.

5. Attosecond Generation

We have seen that a single encounter of a recolliding electron with an atomic core can generate very broad-band radiation. If a single encounter can be engineered then supercontinuum radiation with such characteristics can be produced instead of the comb of high harmonics. The problem i s how to arrange this single encounter. Ivanov et al. [24] have shown how this can be achieved by controlling the atomic wavepacket quiver using a time-dependent elliptical polarization of the laser field. Harmonic generation is produced by linearly-polarized light: only for such linear polarization do the tunnelling wavepackets recollide upon ponderomotive acceleration. For an elliptically-polarized field the electronic wavepackets can tunnel out but miss altogether the atomic core upon phase reversal of the field. Dietrich et al. [25] have shown how the intensity of a

chosen harmonic (the 41^{st} of the Ti:Sapphire pump laser employed) diminishes rapidly when the ellipticity (the ratio of the y-component to the x-component of the laser field) is increased: a 4% elliptically polarized field reduces the chosen harmonic efficiency to 10^{-2} of its value for linearly polarized fields.

Ivanov et al. [24] proposed to use two laser fields, one x-polarized with frequency $(\omega+\Delta\omega)$, the other y-polarized with frequency $(\omega-\Delta\omega)$, each with the same amplitude $E_0 f(t)/\sqrt{2}$ where $f(t)$ describes the pulse shape:

$$\underline{E} = \frac{E_0}{\sqrt{2}} f(t)\{\cos[(\omega+\Delta\omega)t+\phi_1]\hat{\underline{x}} + \cos[(\omega-\Delta\omega)t+\phi_2]\hat{\underline{y}}\}. \qquad (11)$$

If the axes are rotated by 45^o to (x', y')

$$\underline{E} = \frac{E_0}{\sqrt{2}} f(t)\{\cos(\omega t)\cos[\Delta\omega(t-t_l)]\hat{\underline{x}}' + \sin(\omega t)\sin[\Delta\omega(t-t_l)]\hat{\underline{y}}'\}. \qquad (12)$$

At time t_l the light is linearly-polarized ($t_l = 0$ if $\phi_1 = \phi_2$). If $\Delta\omega/\omega$ is chosen to be 0.07, then the ellipticity is $E_{x'}/E_{y'} = 0.2$, at exactly half an optical period later. If this is engineered at the peak of the pulse, then only one packet will return and will radiate a very broad-band supercontinuum. This elliptic switch acts as a kind of atomic Pockels cell [24], so that the supercontinuum is radiated on the attosecond time-scale associated with a single collision.

6. Conclusions

In summary, we have shown that HHG is simply an interference effect of single periodic Bremsstrahlung events that are associated with the recollisions. We have demonstrated that the classical dynamics of the free electron recollision is capable of reproducing many aspects of the harmonic spectrum. It reproduces the increased clarity of the harmonics with rising number of recollisions and simulates the wavepacket spreading. However, it cannot, naturally, take into account the quantum interferences within the wavepacket. These interferences are quite important for the very high harmonics, as we have pointed out, and lead to significant differences between the classical and quantum results. We have developed this idea further to include the dynamics induced by real pulse shapes, and where the quiver motion changes in time. Blue shifts of the harmonics are predicted as a consequence [16]. Finally, we discuss how a single recollision encounter, if one can be engineered, can generate a broad-band supercontinuum on an attosecond time-scale.

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