Subfemtosecond pulses

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High-harmonic emission is very sensitive to the polarization of a fundamental laser beam. By combining two short perpendicularly polarized pulses with frequencies centered at ω_1 and ω_2, where $(\omega_1 - \omega_2)/\omega_1 \ll 1$, we can create a pulse with polarization that sweeps from circular through linear back to circular. It is possible to make the duration of the segment of the pulse capable of emitting short-wavelength radiation so small that emission is possible only for a time less than the period of the fundamental.

Since the development of the laser, obtaining shorter and shorter pulses has been a major emphasis of laser science. To approach the limit of 10 fs, at which short-pulse lasers currently operate,[1,2] nonresonant (Kerr) nonlinearities are used. However, the development of short-pulse coherent sources of less than 1-fs duration will require shorter-wavelength radiation and a new nonlinearity. In this Letter we introduce a method for producing pulses with durations approaching 1 atomic unit of time based on high-harmonic emission from atomic gases.

High harmonics have been extensively studied.[3] Their discrete nature and broad envelope spectrum implies that, if the harmonics are appropriately phased, a train of very short (~10 as) pulses separated by one half of the fundamental period will result.[4] In analogy with lasers, it is not sufficient to have equally spaced modes of an oscillator; they must be locked in the appropriate relative phase.

Aside from phase-matching issues, the harmonic emission of a quantum system is determined by the squared modulus of the second time derivative of the dipole moment. Therefore the problem is to find the dipole moment as a function of time. It was shown[5,6] that, in the intense field regime in which the energy of the free-electron oscillation in the laser field is greater than the ionization potential, one can understand the high-order nonlinear response of an atom by considering the following three steps: (1) In the process of ionizing, the atom releases an electron wave packet into the continuum. (2) In the direction of the laser field the electron follows a trajectory that is first away from the ion and then comes back toward the ion. Quantum effects (spreading of the electron wave packet) dominate in the direction perpendicular to the field. (3) Those electrons that return to the vicinity of the ion emit harmonic radiation in a coherent interaction between the portion of the wave function remaining in the ground state and the returning portion (coherent spontaneous recombination to the ground state). In what follows we use the semiclassical version of this model[5] to describe the approach to producing attosecond pulses. The more comprehensive quantum treatment[6] of the induced dipole moment gives substantially the same results, but, although it is more rigorous, the quantum treatment loses some of the transparency of the semiclassical description.

Both the semiclassical and the quantum analyses indicate that, if viewed through a filter that passes only the highest harmonics, a sequence of short pulses [Fig. 1(a)], would be seen, much like the output of a mode-locked laser. We performed this calculation, assuming a 25-fs pulse of 800-nm light interacting with neon with a peak laser intensity of 6×10^{14} W/cm^2. The filter had the transmission characteristics of 200-nm-thick silver film.[7] The filter ensures that frequencies below 100 eV are filtered out. The dependence of the square of the electron wave function in the direction transverse to the electric field of the laser was taken as Gaussian $\exp\{-[r/r_0(t)]^2\}$, with $r_0(t) = 0.7t$ nm, where t is the time difference, in units of femtoseconds, between the moment of birth of the electron into the laser electric field and the moment of its reencounter with the ion. This $r_0(t) = 0.7t$ value is consistent with the experiment described below and with quantum calculations.[6,8] Figure 1(a) shows $|\ddot{d}(t)|^2$ [$d(t)$ is the time-dependent dipole moment] plotted as a function of time for a few periods near the peak of the pulse. The instantaneous spectrum of any of the pulses is chirped, starting at the filter cutoff, increasing to a maximum frequency of ~140 eV, and finally decreasing again to the filter cutoff.

To obtain a single subfemtosecond pulse from this train, we need to modulate the induced dipole moment on a time scale of the period of the fundamental wavelength, and surprisingly this is relatively simple to do. Returning to the analogy of a mode-locked laser, we introduce an atomic Pockels cell.

The semiclassical model outlined above predicts a strong dependence of the high-harmonic emission on the ellipticity of the fundamental. This can be easily understood. In an elliptically polarized field an electron born (released) at rest into the continuum when the instantaneous electric field is near maximum will be displaced in the direction of the minor axis of the polarization ellipse at the time of its first reencounter with its parent ion. If this displacement exceeds an amount that can be compensated for by the initial transverse momentum distribution of the ionizing electron, then harmonic production ceases. We can therefore achieve the required modu-

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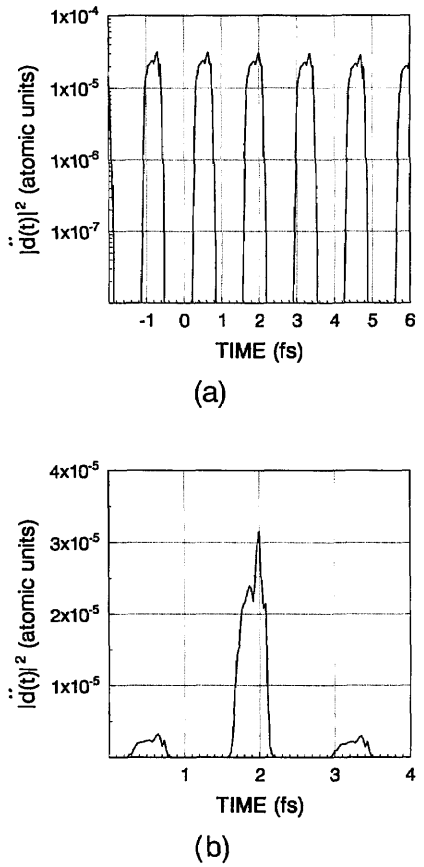

Fig. 1. Square modulus of the second derivative of the time-dependent dipole moment plotted for 800-nm light interacting with neon at an intensity of 6×10^{14} W/cm^2 as seen through a 200.0-nm-thick silver filter. The fundamental field was taken as $\mathbf{E} = E_0 f(t)[\cos(\omega_1 t)\mathbf{i} + \cos(\omega_2 t)\mathbf{j}]$, where $f(t) = \exp[-(t/T)^2]$, $T = 10$ periods of the fundamental, and (a) $\omega_2/\omega_1 = 1.0$ and (b) $\omega_2/\omega_1 = 0.9$.

lation of the induced dipole moment by modulating the ellipticity of the fundamental. This can be done by the combination of two orthogonally polarized pulses of slightly different frequencies. Such a pulse is described by

$$\mathbf{E} = E_0 f(t)[\cos(\omega_1 t)\mathbf{x} + \cos(\omega_2 t)\mathbf{y}]/\sqrt{2}. \quad (1)$$

The field in Eq. (1) can be constructed by use of femtosecond pulse-shaping techniques.[9] In Eq. (1), $f(t)$ describes the pulse envelope. In a reference frame x', y', with x' as the direction of the laser polarization at time $t = 0$, we can write the laser electric field as

$$\mathbf{E} = E_0 f(t)[\cos(\omega t)\cos(\Delta\omega t/2)\mathbf{x}' + \sin(\omega t)\sin(\Delta\omega t/2)\mathbf{y}'], \quad (2)$$

where $\omega = (\omega_1 + \omega_2)/2$ and $\Delta\omega = \omega_1 - \omega_2$. If $\Delta\omega \ll \omega$, this pulse has time-dependent ellipticity.

At times $t = 0, n\pi/\Delta\omega$, where n is an integer number, the pulse is linearly polarized. For other times the pulse is elliptically or circularly polarized. If we are to obtain a single high-frequency pulse, then the envelope $f(t)$ must describe an ultrashort pulse. Before we discuss how short the fundamental pulse must be, we introduce results of experiments with elliptically polarized light (see also Refs. 8 and 10). Because the strong ellipticity dependence of the high harmonics is so critical to attosecond pulse generation we include our most recent measurements. These measurements permit us to determine the rate of transverse spread of the electron wave packet in the continuum, which the semiclassical model requires as a parameter to predict quantitatively the induced dipole moment.

Using the approach described previously,[8] we show in Fig. 2 the harmonic yield for the 37th harmonic as a function of the ellipticity ε. The curve plots theoretical results from the quasi-static model that we discussed above and is completely consistent with the quantum model.[6]

Based on the experimental results we can now state the necessary values of ω and $\Delta\omega$ in Eq. (2). If $\Delta\omega \ll \omega$, with time-dependent ellipticity, then $\varepsilon \equiv E_{y'}/E_{x'} = \tan \Delta\omega t/2$. If we choose $\Delta\omega/\omega = 0.1$, then the ellipticity $\varepsilon = 0.15$ is reached after one half of a laser period. According to experimental results, for 800-nm light interacting with neon (see Fig. 2) at $\varepsilon = 0.15$ the emission will drop by a factor of 5 compared with that at $\epsilon = 0$. With respect to the pulse duration, we choose $f(t)$ so that $f(t) = 0.7$ when linear polarization reappears at $|t| = \pi/\Delta\omega$. With $f(t) = 0.7$ the harmonics are emitted less efficiently, since the ionization rate has fallen by approximately a factor of 10 from its maximum, and the maximum frequency of the harmonic emission has fallen by a factor of 2. The latter ensures that the already-weak emission is not transmitted through the filter.

We now show results of calculations of the square of the time-dependent dipole moment, using the pulse described by Eq. (1). With the parameters outlined above, we use the semiclassical model described above with the expansion of the wave function transverse to the laser electric field that is determined by the quantum theory[6] and the above experiment. Figure 1(b) shows $|\ddot{d}(t)|^2$ versus t for the same conditions as in Fig. 1(a), except that now the pulse has a time-dependent ellipticity determined by $\Delta\omega/\omega = 0.1$. To ensure that only high-harmonic radiation is present, we filtered out low harmonics by

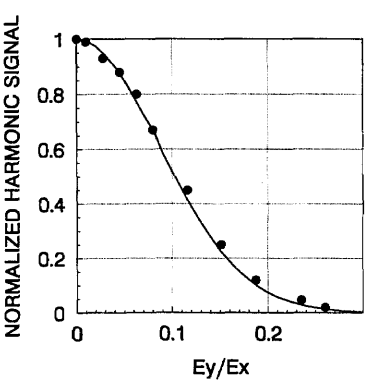

Fig. 2. Ellipticity dependence of the harmonic emission observed from helium by use of 800-nm light. The data points plot the experimental results, and the solid curve plots the results of the semiclassical theory presented here.

using the same 200-nm-thick silver filter as that in Fig. 1(a). The time dependence of the instantaneous emission frequency is chirped, just as described for the individual pulses in Fig. 1(a).

It is not sufficient to fabricate a subfemtosecond pulse; we must ensure that the pulse will emerge undistorted from the nonlinear medium and the filter. In the medium there are two issues of concern, i.e., group-velocity and phase-velocity dispersion. In experiments on high-harmonic generation there are two main sources of phase-velocity dispersion: (1) the phase advance that is due to the focusing geometry of the incident laser and (2) the dispersion in the laser-produced plasma.[11] Whereas in principle the first problem is technical, the second problem is unavoidable; the high laser intensities necessary for efficient high-harmonic generation always lead to ionization. Indeed, the two are inherently related. We now show that, if the phase-matching condition is satisfied, no significant increase in the duration of the ultrashort pulse will occur.

In low-density plasmas the refractive index is $n(\Omega) = 1 - \omega_{pl}^2/2\Omega^2$, where Ω is the optical frequency in question. Therefore the change in the phase velocity of $\delta v_{ph}(\omega_L)$ at the fundamental frequency ω_L is N^2 times larger than at $N\omega_L$. One can also show that, for low plasma densities, the change in the group velocity in the plasma is $\delta v_{gr}(\Omega) = -\delta v_{ph}(\Omega)$. Hence we conclude that (1) the main factor involved in increasing the duration of the high-frequency pulse is the difference between the group velocity of the generated signal and the phase velocity of the fundamental wave and (2) this effect is of the same order as the phase dispersion between the incident and harmonic waves. The phase-matching condition requires that the dispersion between incident and harmonic waves not exceed one half of the harmonic radiation wavelength. Therefore, even for poor phase matching, the dispersion of the ultrashort pulse is much smaller than the wavelength of the fundamental radiation. Thus dispersive broadening can never approach an optical period of the fundamental.

Another source of dispersive broadening is the dispersion in the filter that is used to eliminate low harmonics. In our calculations we used a 200.0-nm-thick silver filter, which is transparent for the radiation with Ω exceeding 100 eV. All dispersion characteristics of this filter are known,[8] and a 200.0-nm-thick silver film will increase the pulse duration by a maximum of 0.1 fs.

We emphasize that the conditions assumed above are experimentally feasible. Phase-related beams with frequencies ω_1 and ω_2 can be selected from an ultrashort laser pulse. The 800-nm light was chosen because, at this wavelength, pulses with durations of only ten optical periods are now generated[1,2] and amplified.[12] Two frequency bands near ω_1 and ω_2 can be selected by use of standard techniques.[9] The bandwidth of the two components can easily be broad enough to permit the synthesized pulse to fall sufficiently in intensity before the linear polarization condition can reappear, thus ensuring that harmonic emission is very weak at all times except near $t = 0$. Measurement of the time duration of subfemtosecond pulses also seems feasible. Since it is as complex as the production process it will be addressed elsewhere.[13]

Our approach to attosecond pulse generation is a special case of a much more general phenomenon, namely, coherent control of strong-field atomic processes. Here we have described how control over the trajectory of a continuum electron can modulate the high-order susceptibility in time. The same physics can be applied to the control of the atomic susceptibility in space. In fact, a relatively weak (control) beam of circularly polarized light propagating along the direction of the electric-field vector of the fundamental light will deflect an electron so that it misses the ion. Therefore the control beam can ensure that any atom illuminated by the control beam will be unable to emit harmonic radiation, thus permitting quasi-phase matching. We foresee its application to harmonic generation using ions.

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