Single attosecond pulse generation from asymmetric molecules with a multicycle laser pulse

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A new scheme for single attosecond pulse generation from asymmetric molecules with a multicycle laser pulse is proposed. It is shown that both even and odd harmonics are generated from the asymmetric molecules, and the attosecond pulses are produced every full cycle of the driving laser field rather than each half-cycle. By filtering the harmonics in the cutoff of the spectrum, a single attosecond pulse can be obtained with a multicycle laser pulse with a duration of 2 times longer than the few-cycle pulse conventionally used.

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The advent of attosecond pulses makes it possible to study the dynamic of electrons in atoms and molecules directly and in real time, allowing scientists to investigate deeper into the burgeoning field of attoscience. Thus the generation of attosecond pulses has attracted much attention, and many schemes based on high-harmonic generation (HHG), stimulated Roman scattering, Thomson scattering, and the plasma mirror have been proposed in the past years.

So far HHG is the unique way to produce attosecond pulses in experiments. In the breakthrough work, a train of attosecond pulses separated by a driving laser half-cycle is produced by superposing five harmonics generated in argon onto a multicycle laser pulse with a duration of 40 fs full width at half-maximum (FWHM). However, straightforward attosecond metrology prefers an isolated attosecond pulse, and thus much effort is expended to extract an isolated pulse from the attosecond pulse train. It is proposed that a single attosecond pulse can be generated with a few-cycle laser pulse. In this scheme, the highest harmonics (the so-called cutoff) are confined in the short time when the laser intensity reaches its maximum, and the superposition of these harmonics leads to the generation of an isolated attosecond pulse. However, the requirement for the laser pulse is rather stringent; it requires a few-cycle laser pulse with a stabilized carrier-envelope phase, which is achievable only by using state-of-the-art laser technology. Therefore much effort has been expended to produce single attosecond pulse generation with multicycle laser pulses. It is shown that HHG depends sensitively on the ellipticity of the driving light. When a laser pulse with time-dependent ellipticity is used, the harmonics are only efficiently emitted for a short time, and then a single attosecond pulse can be generated with a multicycle laser pulse. Furthermore, it is proposed that a single attosecond pulse can be produced in the multicycle pulse regime with a two-color driving laser field. Very recently a single attosecond pulse was produced using a multicycle pulse with a very high intensity.

In this Letter we propose a new method for single attosecond pulse generation in the multicycle pulse regime with less stringent experimental conditions based on HHG from asymmetric molecules. As is well known, only odd harmonics are generated in the interaction of a laser field with atoms and symmetric molecules because of the symmetry of the system. Therefore, in the time domain attosecond pulses are produced periodically every half-cycle of the driving field. To extract an isolated pulse from the attosecond pulses train, a few-cycle laser pulse is required. However, for asymmetric molecules, the symmetry is broken; then both odd and even harmonics can be generated. In the time domain, attosecond pulses are produced every full cycle of the driving field rather than each half-cycle. In this case, by applying the same filtering method used in the conventional scheme of few-cycle laser pulse, a single attosecond pulse can be generated by using a multicycle laser pulse with a duration of 2 times longer than the few-cycle pulse conventionally used. In the following, we first investigate the HHG from asymmetric molecules in a plane wave and show that attosecond pulses are produced every full cycle of the driving field. Afterward, the interpretation of this process is presented. Finally, we show the results of single attosecond pulse generation with a multicycle laser pulse.

As the simplest heteronuclear asymmetric molecule, HeH$_2^+$ has been the subject of many previous works. In this Letter, we preform a quantum simulation of HeH$_2^+$ in an intense laser field. The three-dimensional time-dependent Schrödinger equation is $i\frac{\partial}{\partial t}\Psi(r,t) = [\mathbf{p}^2/2 + \mathbf{A}(t) \cdot \mathbf{p} + V(r)]\Psi(r,t)$. The Coulomb potential $V(r) = -Z_1/|r+\mathbf{R}/2| - Z_2/|r-\mathbf{R}/2|$, where $Z_1=2$ and $Z_2=1$ are the electric charges of He$^2+$ and H$^+$, respectively, and atomic units (a.u.) are used. The internuclear distance $R$ is 3.89, $A(t) = A_0 f(t) \sin(\omega_0 t + \phi_0) \mathbf{Z}$, is the vector potential of the laser pulse, $A_0$ is the amplitude, $f(t)$ is the pulse envelope, $\omega_0$ is the laser frequency, $\phi_0$ is the carrier-
envelope phase. The Schrödinger equation is solved in the cylindrical coordinates. We use a Ti:sapphire laser pulse with an intensity of $2 \times 10^{15}$ W/cm$^2$ and a wavelength of 800 nm.

The harmonic spectrum of HeH$_{2}^{+}$ in a plane laser field is shown in Fig. 1(a). We assume that the laser field is turned on and off linearly over 3 optical cycles. In contrast to the HHG from atoms and symmetric molecules, both even and odd harmonics are produced from asymmetric molecules. Since the harmonics in the cutoff are locked in phase, the superposition of these harmonics will give rise to a train of attosecond pulses. The temporal profiles of the attosecond pulses are shown in Fig. 1(b). In contrast to the atoms and symmetric molecules, attosecond pulses from asymmetric molecules are produced every full cycle of the driving field rather than each half-cycle. This characteristic can be easily understood via the close analogy to the mode-locked laser. For atoms and symmetric molecules, the HHG results in a series of models spaced by $2\omega_0$, and in the time domain the interval between the adjacent attosecond pulses is $2\pi/(2\omega_0) = T_0/2$, where $T_0$ is the optical cycle of the driving laser field. However, for asymmetric molecules both even and odd harmonics are produced, the frequency difference between the adjacent harmonics is $\omega_0$, and then the time interval of the attosecond pulses becomes $T_0$.

Although the temporal characteristics of the attosecond pulses generated from asymmetric molecules have been discussed in the previous paragraph, the full explanation of the underlying physics is somewhat deeper. HHG is well understood in terms of the three-step model. In detail, the electron is first ionized from the nucleus, then accelerated freely in the laser field, and finally recaptured by the nucleus, emitting an energetic photon. As the first step, the ionization process play an important role in HHG. For asymmetric molecules, the ionization process is strikingly different in adjacent half-cycles. In the first half-cycle the electric field is parallel to the permanent dipole of the molecule (PDM), while in the subsequent half-cycle the electric field changes its direction and is antiparallel to the PDM. The combined Coulomb and laser field potential $V(t) + E_z(t)$ along the molecular axis in the parallel and antiparallel orientation cases are illustrated by the solid and the dotted curves in Fig. 2, respectively. The arrows are drawn at the energy level of the dressed ground state of HeH$_{2}^{+}$. In the parallel orientation case the energy of the electron in the deeper well at He$_{2}^{+}$ can be approximated by $-Z_1^2/2 - Z_0/R - E_0 R^2/2 = -2.7$, and in the antiparallel orientation case the electron energy is approximately $-Z_1^2/2 - Z_0/R + E_0 R^2/2 = -1.8$. This result implies that single attosecond pulse can be obtained by using a multicycle laser pulse with a duration 2 times longer than the few-cycle pulse by applying the same filtering method conventionally used. To verify this idea, we use a multicycle pulse with the sine squared envelope $A(t) = \sin^2(\pi t/T)\sin[\omega_0(t - T/2) + \phi_0]$. Then $E(t) = -\partial A(t)/\partial t$, and the pulse with $\phi_0 = \pi$ is defined as a cosine pulse, which with $\phi_0 = \pi/2$ is defined as a sine pulse. As shown in Fig. 3(a), a single attosecond pulse is generated with a 12 fs cosine laser pulse by filtering the harmonics in the cutoff

![Fig. 2. (Color online) Combined Coulomb and driving field potential $V(t) + E_z(t)$ for HeH$_{2}^{+}$ along the molecular axis. The arrows are drawn at the energy level of the dressed initial state of HeH$_{2}^{+}$.](image-url)
(270th–280th). The satellite pulse is 1 order of magnitude lower than the main pulse. Since the duration of the driving laser pulse is greater than 10 fs, the attosecond pulse depends less sensitively on the carrier-envelope phase of the driving laser pulse. The temporal profile of the attosecond pulse generated with a 12 fs sine laser pulse is shown in Fig. 3(b). It is clearly shown that a single attosecond pulse still can be generated by filtering the harmonics from 270th to 280th. Note that the intensity of the satellite pulse is less than 10% of the main pulse when \( \phi_0 \) varies from \( \pi/2 \) to \( \pi \).

In the conventional scheme for single attosecond pulse generation using the few-cycle laser pulse, a cosine pulse is required and the carrier-envelope phase must be stabilized.\(^{10}\) In contrast to the conventional scheme, the advantage of our scheme is that the required pulse duration can be extended to 12 fs, and then the attosecond pulse depends less sensitively on the carrier-envelope of the driving laser field. The required laser system in our scheme is relatively easier to realize in experiments. Note that molecules should be first aligned in our scheme. This can be achieved by using a prepulse or static electric field,\(^{19}\) which has been extensively investigated previously.\(^{20}\)

Our scheme for single attosecond pulse generation is based on the distinct difference between the ionization of asymmetric molecules in the parallel and antiparallel orientation cases. It is shown that this effect occurs at other laser intensities and molecular internuclear distances. In addition, it is indicated that this effect is present in any asymmetric polar molecules. Possible candidates for experiments are alkaline hydrides (LiH\(^+\) etc.) and halogen compounds (HCl\(^+\) etc.), which are stable molecular systems with asymmetric potentials and a single valence electron. Therefore, even though our results are obtained with HeH\(^2+\), it is expected that single attosecond pulse still can be produced with other asymmetric molecules. Also, our scheme for single attosecond pulse generation with asymmetric molecules is robust.

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