Yield and temporal characterization of high-order harmonics from intense midinfrared excitation of a cesium vapor

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Cesium vapor interacting with a tightly focused, intense midinfrared (3.4 μm) source produces high harmonic radiation in the visible/UV spectral range. The measured yields of harmonic orders 9–17 are found to yield good quantitative agreement with numerical simulations that include both the single-atom and the macroscopic response. The 5th–9th harmonic orders are generated with sufficient pulse energies (~100 pJ) for direct temporal measurements using an autocorrelation method and when correlated with bandwidth measurements are found not to be transform limited. A blueshift connected to a strong time-dependent ionization appears to be the cause of this spectral broadening.

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I. INTRODUCTION

The discovery of high harmonic generation (HHG) [1,2] has spurred over a decade of intense experimental and theoretical work. High harmonics are commonly generated in high ionization potential atoms (rare gases) at intensities around \(10^{14} \text{ W/cm}^2\) [3], so that the Keldysh parameter [4] defined as \(\gamma = (I_p/2U_p)^{1/2} \approx 1\). Here \(I_p\) is the ionization potential and \(U_p\) is the ponderomotive energy defined as the cycled-averaged energy of a free electron in a laser field. \(U_p\) is proportional to the intensity and quadratic with wavelength. The Keldysh parameter provides a scalable gauge of the interaction dynamics and defines the limits between tunnel (\(\gamma \ll 1\)) and multiphoton (\(\gamma \gg 1\)) ionization. It has been shown [5] that alkali metal vapors interacting with an intense 3.4 μm midinfrared (MIR) source results in a scaled interaction with a Keldysh parameter also close to one. This scaled interaction is nonperturbative and produces both above-threshold ionization [6] and high harmonics [5] spectra. High harmonics generated from a MIR fundamental field have the practical advantage of radiating in the visible/UV spectral range where standard temporal metrology, e.g., autocorrelation, can be used, provided the harmonic production has sufficient intensity.

Previously, we reported [7] on the characterization of harmonic orders 5–9 produced by a MIR source interacting with cesium atoms, with special emphasis placed on their coherence properties. The present work is motivated by a need for a quantitative test of our understanding of high harmonic generation in alkali-metal atoms excited by a long wavelength MIR pulse. In a broad sense, our strategy for utilizing a scaled interaction as a roadmap for synthesizing a subfemtosecond pulse hinges upon the ability to measure and predict. Even though the current scaled system achieves a Keldysh parameter close to one, there remains significant differences with other investigation. First, alkali-metal atoms differ from inert gas atoms in their atomic structure and most notably, the alkali atoms are good one-electron systems (hydrogen-like) while inert gas atoms have a closed shell multielectron configuration. Consequently, the single-atom response could be different. Second, there are practical differences in the experimental geometry that will influence the macroscopic response. In particular, we study harmonic generation in a cesium vapor inside a heatpipe interacting with a focused 3.4 μm beam. In this arrangement the harmonics are generated from a tightly focused geometry since the medium length is larger than both the beam’s confocal parameter and coherence length. Furthermore, the intensity is large enough within the confocal length to completely ionize the medium before the end of the pump pulse. Under these conditions, we have measured the absolute yields of the harmonic orders 5 through 17. In parallel, nonperturbative calculations that include both the single-atom and the macroscopic response were performed. Good quantitative agreement between the calculated and measured yields has been achieved.

We have also measured the pulse durations and spectral bandwidths of the harmonic orders 5–9 and found that the time-bandwidth products are greater than twice the transform limit. Analysis shows that the most probable cause of this
spectral broadening is a blueshift of the fundamental field due to the rapid change in the medium’s free-electron density, and not a chirp due to an intrinsic intensity dependent phase [8,9].

II. EXPERIMENTAL AND THEORETICAL METHODS

Scaling the Keldysh parameter to approximately one for the alkali metals requires a MIR light source capable of producing an intensity greater than 1 TW/cm². This is accomplished in the current experiment by difference frequency mixing an amplified Ti:Sapphire laser system (pump beam) with an amplified Nd:YLF laser system (signal beam) in a potassium titanyl arsenate (KTA) crystal. The idler produced in this process is tunable (3–4 μm) with a near Gaussian spatial mode. The system operates at a 1 kHz repetition rate and produces ~100 μJ pulse energy with an ~2.5 ps pulse duration. The MIR pulse duration is measured using a home-built second harmonic generation (SHG) autocorrelator. The linearly polarized MIR light is focused by a 150 mm focal length CaF₂ lens producing a 60 μm beam diameter (1.7 mm confocal parameter) in a heat pipe containing a 2 cm long vaporized cesium column. The heat pipe is operated at a Cs vapor pressure near 18 torr and a variable argon buffer gas pressure of 30–75 torr. For the current measurements, the 3.4 μm beam is focused towards the end of the cesium column in order to minimize reabsorption and dispersion.

The harmonic light exits the heat pipe through a UV grade fused-silica window and enters two separate achromatic systems where temporal and spectral measurements are performed. The frequency spectrum is obtained in a 0.18 m flat-field monochromator equipped with an intensified charge-coupled device (CCD) camera. A 147 g/mm grating provides a broad low resolution spectrum of the high harmonic distributions. The absolute energy measurements are accomplished by comparing the harmonic’s frequency spectrum directly to the value measured on a calibrated photodiode for the 5th and 7th harmonics. Once this calibration is established the higher harmonic (orders > 7) energies are extrapolated from the response function of the monochromator/ICCD system. A 2400 g/mm grating with a resolution of <0.4 Å/pixel is used for the spectral width measurements. The pulse duration measurements were performed using an autocorrelator based on noncollinear SHG. Two replica pulses resulting from a two-arm interferometer are frequency doubled in a type-I beta-barium borate (BBO) crystal and the SHG signal is detected on a filtered photomultiplier tube. The wavelength and intensity of 5th and 7th harmonics are sufficient for autocorrelation. However, the shorter wavelength of the 9th harmonic could not be phasematched for SHG in BBO and as a result had to be cross correlated with the fundamental or 5th harmonic. Both cross-correlation schemes gave similar results although referencing to the 5th harmonic allowed for a more precise measurement.

In order to realistically simulate the experimental conditions, calculations of the harmonic distributions are performed that include both the response of individual cesium atoms to the intense MIR field, and the propagation and phase matching of the generated harmonics in the macroscopic medium. First, the time-dependent Schrödinger equation within the single active electron approximation [10] is integrated numerically to obtain the dipole spectrum and the ionization rate for a quasicontinuous intensity of the fundamental field. The calculation is repeated for several intensities. We use the pseudopotential of Stevens et al. [11] to model the interaction of the valence electron with the ionic core.

We then solve the Maxwell wave equation in the paraxial and slowly varying envelope approximations, using the single-atom dipole moments as a source of the nonlinear polarization [12]. Ionization is incorporated into the macroscopic calculation through a slowly varying depletion of the neutral medium, and through the resulting time-dependent density of free electrons which influences the propagation of the fundamental field. Absorption cross sections for harmonic order 9 through 17 are calculated from the oscillator strengths of the 6s→np transitions using the potential of Ref. [11]. For the 5th and 7th harmonic orders, whose energies are between the first excited state and the continuum limit, we have experimentally estimated the absorption cross section by measuring the harmonic yield as a function of the position of the focus relative to the center of the heat pipe. Atomic dispersion is also taken into account; the refractive index at the fundamental and harmonic frequencies are calculated using the Sellmeier equation [13]. This computational approach has been shown to produce excellent agreement with experimentally determined absolute photon yields for xenon atoms irradiated by 1.06 μm pulses [12].

For cesium atoms irradiated by 3.4 μm radiation with a peak intensity of 1 TW/cm² the Keldysh parameter is 1.3, which indicates that the dynamics is a mix of multiphoton and tunnel ionization. The single-atom calculations show the typical tunneling-dominated behavior for the harmonics above the ionization threshold (≥11-order); the dipole amplitudes exhibit a clear cutoff and plateau behavior as a function of intensity. The phase of these high harmonics shows a dependence on the intensity which is consistent with contributions from several quantum paths [8], although we do not at present have enough data to resolve the different paths [9]. However, the harmonics below threshold show no systematic phase behavior as a function of the intensity.

III. HIGH HARMONIC YIELDS IN THE TIGHT-FOCUSING REGIME

Figure 1(a) shows the absolute pulse energies (pJ) for high harmonic orders 5–17 for a cesium heat pipe temperature of 673 K (18 torr) excited by the 3.4 μm fundamental pulse. The results are for two different argon buffer gas pressures, 30 torr (solid circles) and 76 torr (open circles). The spectra show the general strong-field characteristic of a plateau in the high harmonic distribution. The shortest wavelength detected in the current experiment is limited to 0.200 μm (17-order) since our optical/detection system is airbased and blind to shorter wavelengths. The 3U_p+I_p cut-off law [14] would predict that the harmonic plateau terminates in the vacuum ultraviolet (<0.2 μm) beyond our detection limit. The energy conversion efficiency from the fundamental pulse varies from 10⁻⁶ to 10⁻⁹ from the 5th
harmonic into the plateau region, corresponding to $10^8$–$10^9$ photons/pulse over the same range. The plot also shows a dependence of the high harmonic yield on the argon buffer gas pressure. The dotted line in Fig. 1(a) indicates the position of the ground-state binding energy, the high harmonics produced to the right of this line are bound-free transitions.

Also shown in Fig. 1(a) is the calculated harmonic yields ($\diamondsuit$) for a fundamental peak intensity of 1 TW/cm$^2$. The macroscopic parameters in the calculation are the same as those used in the experiment; a 2 mm confocal parameter, a 2 cm medium length, and an 18 torr cesium pressure. The agreement between the experiment and theory is good, capturing the relative shape of the plateau although overestimating the absolute strength. Examination of Fig. 1(b), which plots the ratio of the theory to experiment (76 torr measurement) as a function of harmonic order, shows that in the plateau region for orders $\geq 9$ the two differ only by a factor of 10. There are a number of potential sources for this discrepancy that can be traced to experimental uncertainties and the idealized modeling in the calculations. For one, the model assumes a Gaussian focal distribution which is probably not the case for the experiment and at present beyond our limited diagnostic capabilities at 3.4 $\mu$m pulse. In addition, as shown in Fig. 1(a), the observed harmonic distribution is dependent on the buffer gas pressure and suggests a more complex behavior in the gas dynamics, e.g., molecular condensation, than in uniform density distribution, as assumed in the calculations. Considering that small uncertainties in the experiment can result in large changes in the harmonic yield, since the emission is quadratic in density and highly nonlinear with intensity, the agreement for the high-order harmonics is remarkably good. However, for lower orders ($\leq 7$) the difference becomes larger. The fact that the experimental yields for the low harmonics are less sensitive to the buffer gas pressure indicates that there is some balance between gas density and the absorption and phase matching in the medium, and this is not mimicked in the macroscopic calculation. A significant difference between the earlier study [12] on inert gas atoms that achieved better agreement between experiment and theory than the current study on cesium could be a reflection of the atomic structure, i.e., closed-shell versus hydrogenic. In cesium the presence of low-lying excited states can strongly influence the yields of the 5th and 7th harmonic orders since both are close to the two lowest $p$ states which have large oscillator strengths. Consequently the contributions to phase matching from linear dispersion and absorption could be more important for these low harmonics than for the higher harmonics. For instance, our estimate of the linear susceptibility parameters does not account for dynamic effects caused by the strong MIR field [15]. This is an interesting problem which will be the subject of a future study.

IV. TEMPORAL AND SPECTRAL PROPERTIES

Temporal and spectral characterization of harmonic orders 5–9 are performed using a cesium vapor pressure of 10 torr. Figure 2 shows the autocorrelation (a)–(c) and frequency spectrum (d)–(f) for these harmonics and the (a) autocorrelation of the 3.4 $\mu$m pulse. The width of the autocorrelation trace ($\tau_{ac}$) for the fundamental 5th and 7th harmonics is analyzed by fitting a Gaussian intensity profile and the duration $(\Delta \tau_q = \tau_{ac}/\sqrt{2})$ evaluated assuming a Gaussian temporal profile. The duration of the 9th harmonic is deconvoluted from the cross-correlation trace using the 5th harmonic as a reference pulse. Before each cross-correlation measurement, an autocorrelation is recorded to characterize the reference pulse. The bandwidth, $\Delta f_q$, of the high resolution wavelength spectrum also shows the ratio of theory to experiment (76 torr data) as a function of harmonic order.
scales as $1/q$, and can therefore be neglected. The linear fit to the experimental data, shown in the Fig. 3 with solid circles, yields a blueshift of 5 nm. The blueshifts and thereby the bandwidths of the harmonics then scale as $1/q$ in wavelength, or with $q$ in frequency. Note that the direct blueshift of the $q$th-harmonic from the time-dependent refractive index scales as $1/q^3$, and can therefore be neglected.

Figure 3 plots the measured blueshift ($\Delta \lambda$) for the 5th through 17th harmonics. The shift increases nearly linear with harmonic order, in good agreement with the prediction for the bandwidth discussed above. The linear fit to the experimental data, shown in the Fig. 3 with solid circles, yields a blueshift of the fundamental of $\sim 4$ nm, which is in very good agreement with our simple estimate of 5 nm.

V. SUMMARY

An intense 3.4 $\mu$m fundamental pulse used to generate visible/UV high harmonics from a strongly focused geometry and a rapidly ionizing Cs vapor has been spectrally and temporally analyzed. The experiment reports the first direct measurement of the pulse duration for high harmonics up to 9th order, as well as spectral measurements up to 17th order. The harmonic distribution is governed by the nonperturbative nonlinear dipole response and the dispersion/absorption properties of the medium. Moreover, the strong time-dependent ionization induces a blueshift of the fundamental radiation. The observed blueshift of the 5th–17th harmonic orders range from 0.5 to 2 THz which is in good agreement with that induced by the shifted fundamental pulse.
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