Enhanced High Harmonic Generation from an Optically Prepared Excited Medium

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We investigate high harmonics generated from rubidium atoms irradiated simultaneously by an intense 3.5 μm fundamental field and a weak cw diode laser. When $5p$, $5d$, and $4d$ excited states are populated through cascade excitation or deexcitation, orders-of-magnitude increases in harmonic yield as compared with the ground state are observed. It appears that, quite unexpectedly, the population accumulated in the $4d$ state alone is responsible for the observed enhancement.

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One dramatic manifestation of nonlinear optical physics arises from the response of a ''single'' atom to an intense, ultrafast laser pulse. The interaction produces energetic electrons [1] and photons [2]. Over the past two decades extensive work has been directed toward understanding these basic physical processes. More recently the emphasis has been on the practical implications of producing a tabletop photon source capable of converting visible light into coherent, short wavelength radiation. In an intense field a comb of odd-order high harmonics of the fundamental field is emitted by the atom. Harmonic orders of several hundreds have been observed [3]. A major prospect [4] is the synthesis hyperfast extreme ultraviolet (XUV) pulses with attosecond durations (10^{-18} s). Recently, the principles of these new techniques have been demonstrated [5,6].

High harmonic generation (HHG) results from both the single atom (quantum) and macroscopic (phase-matching) response. To date, investigations of the atomic response have been limited to selecting a ground state atom, generally a rare gas. Theory has examined HHG under conditions where excited states play a role [7,8]. In this Letter we report on the first observation of high-order harmonics from an optically prepared excited state atom. In our work the two fields, and hence the atomic populations, are incoherent. Specifically, Rb atoms are exposed to an intense 3.5 μm midinfrared (MIR) laser pulse in the presence of a weak, cw laser field tuned to the $5S_{1/2} \rightarrow 5P_{3/2}$ (D_2 line) transition. The experiments show that exciting the medium results in large enhancements of the harmonic yields (orders 5 to 11). The increased yields depend strongly on the MIR intensity and the cw resonant laser wavelength. However, the observed enhancements appear to originate from a completely unanticipated source: a population that cascades into and accumulates in the $4d$ excited state.

Compared to ground state atoms, excited states interacting with an intense laser field can provide effective single atom control of the HHG process due to the change in atomic polarizabilities, ionization rates, and modifications in the continuum dynamics. Unfortunately, the large excitation energy of rare gases has hindered the experimental exploration of excited states since vacuum ultraviolet (vuv) light is needed for state preparation. This difficulty can be avoided by using a scaled approach that mimics the behavior of an inert gas atom interacting with intense near-infrared light. In the language of Keldysh [9], an equivalent interaction can be realized by exciting a lower binding energy atom, e.g., alkali metal, with an intense lower frequency field, e.g., MIR. The benefit in the current context is that the alkali transitions are accessible with visible diode lasers. The scaled approach for achieving an equivalent and strongly nonperturbative interaction for the production of high-energy electrons [10] and high harmonics [11] has been established.

The cw pumping, spontaneous emission, and collision processes produce independent (incoherent) populations in several different Rb initial states so that we must treat each of them separately in the calculations. Ionization rates and dipole strengths are calculated by solving the time-dependent Schrödinger equation within a single-active-electron approximation [12]. The intense laser pulse temporal profile is simulated by a linear field ramp for 5 cycles (20 cycles for the $5d$ state), followed by 30 cycles at constant intensity. To minimize the transient effects created during the ramp, we evaluate the ionization rates and the dipole strengths only over the last 10 cycles of the pulse [13].

Figure 1 shows the calculated single-atom dipole emission strengths for the harmonics H3–H9 for four different Rb initial states. For brevity the notation Hq references the

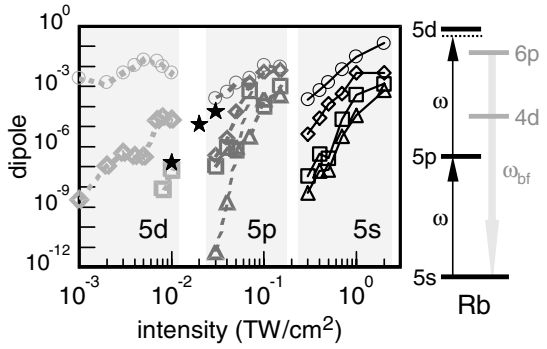

FIG. 1. Calculated Rb dipole strengths for orders H3 (circles), H5 (diamonds), H7 (squares), and H9 (triangles) as functions of the MIR intensity. The gray bands are the $5s$, $5p$, and $5d$ initial states. The star symbols are the results for H7 from the $4d$ state. Relevant Rb states are shown on the right. The arrow labeled ω_{bf} is the 0.42 μm blue fluorescence detected in the experiment.

qth-order harmonic. The $5S_{1/2}$ ground state emission results from transitions originating and returning to the $5s$ valence orbital. Since in the experiment the weak diode laser (ω) transfers the population into several excited states, we present yields from those states we believe to be the most important: the $5p$ orbital of the $5P_{3/2}$ state and the more weakly bound $5d$ orbital of the $5D_{3/2}$ state, along with the $4d$ orbital from the $4D_{3/2}$ state.

As expected, for the more weakly bound excited states, a given harmonic saturates at a substantially lower intensity: a decrease of about 1 order of magnitude between the $5s$ and the $5p$ and between the $5p$ and the $5d$ states. The MIR intensity must exceed 0.5 TW/cm^2 to produce harmonics from the ground state. However, below this threshold, large HHG yields appear achievable from the excited states by introducing the weak cw diode laser. We find that for each state the individual harmonic orders rise rapidly then saturate as the ionization itself becomes saturated and the HHG distribution differs from each initial state. The calculations show that the significant decrease in the dipole strength between adjacent orders makes the $5d$ state an ineffective generator compared with the $5p$ or $5s$ states. In fact, Fig. 1 shows that at the lower saturation intensity of the $5d$ state no higher orders ($q \geq 7$) are generated since the field energy transfer to the ionizing electron during the essential one-cycle time interval following its release is small [14].

Thus the calculations suggest that, below the $5s$ threshold ($<$0.5 TW/cm^2), only the $5P_{3/2}$ state or another of the strongly bound valence states would produce high harmonics ($q \geq 7$); the $5d$ state, and other Rydberg states (studied, but not shown) ionize too easily. From these results we conclude that the MIR intensity must be sufficient to excite the $5p$ or the $4d$ states to produce high harmonic orders ($q > 5$), i.e., $I_{\mathrm{MIR}} > 0.05$ TW/cm^2.

Our calculations do not include phase matching but some general statements can be made. The MIR confocal length, b, is approximately equal to the target length, L. In this case [2], the harmonic yield scales as the density squared. Assuming equal dipole strength for the different states at their respective saturation intensities, one expects that the HHG yield from the excited medium exceeds the ground state emission since the contributing volume to the macroscopic fields is larger. However, for $b \simeq L$, the change in the geometric volume beyond the saturation intensity, I_s, depends only on the radial growth which for a Gaussian spatial distribution increases as $\ln(I/I_s)$, where $I \geq I_s$. Indeed, significant ionization which can shorten the coherence length occurs during the MIR pulse. For the ground state experiments, ionization is expected to become most important at the highest intensities. This would then lead to a less than N^2 density dependence but not occur. Phase-matching effects are not expected to change the important conclusions of this Letter. Their inclusion would be required, however, if a quantitative agreement with measurements were sought, which is beyond the scope of this analysis.

In the experiment, 2.5 ps, 3.5 μm MIR pulses are produced by difference frequency mixing the outputs of titanium sapphire ($\sim$0.81 μm) and Nd:YLF (1.05 μm) kilohertz repetition rate laser systems in a phase-matched potassium titanyl arsenate nonlinear crystal [15]. The resulting MIR pulse has energy in excess of 100 μJ in a near-Gaussian mode.

The linearly polarized MIR light is focused by a $f =$ 150 mm lens into a vacuum chamber at a distance of $\sim$1 mm downstream of the aperture of a Rb oven. At this location, the MIR beam diameter is $\sim$50 μm with a confocal length of 1.1 mm, nearly equal to the target length along the propagation direction. The oven temperature controls the Rb vapor pressure (0.1–10 Torr). The maximum MIR peak intensity is $\sim$1 TW/cm^2. The high harmonics generated on axis are propagated through air and 1:1 imaged onto the entrance slit of a 0.3 m flat-field monochromator equipped with a gated, intensified charge-coupled device camera. Only harmonic orders $\geq$5 are detected.

A temperature stabilized cw diode laser operating on the Rb D_2 line prepares the excited state target. Control of the diode's temperature and current allows wavelength tuning near the 0.78 μm resonance, which is independently monitored using a low-density (10^{10} cm^{-3}) Rb fluorescence cell. The weak diode laser counterpropagates at a small angle with respect to the intense MIR light. In the focal region, where the two beams spatially overlap, the diode intensity is $\geq$ 100 W/cm^2, which is well above saturation for the D_2 line [16]. The diode laser's linewidth is matched to the Doppler broadened Rb absorption profile ($\sim$700 MHz at 1 torr) by applying a high-frequency modulation to the diode's forward current.

The main experimental results are presented in Figs. 2 and 3. Figure 2(a) shows a portion of the Rb HHG distribution excited by a 3.5 μm MIR pulse alone and with the addition of the cw diode laser tuned to the D_2 line. The background resulting from the cw diode laser alone is subtracted from the latter spectrum. Other than the addition

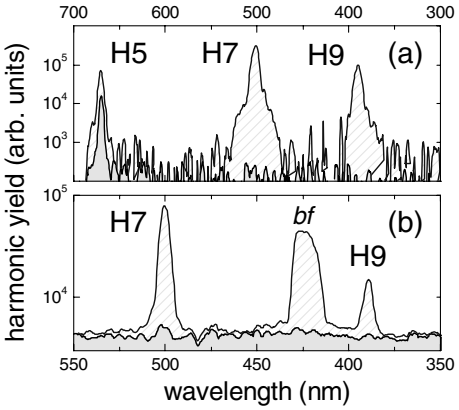

FIG. 2. (a) Harmonic spectra taken at 0.25 torr with 0.8 TW/cm^2, with 3.5 μm pulses only (gray shaded areas) and with the cw diode laser (gray hashed areas) tuned to the D_2 transition. (b) Spectra taken at 0.75 torr with 0.1 TW/cm^2, with 3.5 μm pulses, and with the cw diode laser "on" (gray hashed areas) and "off" (gray shaded area) resonance.

of the diode laser, the conditions for both spectra are identical. They are recorded after optimizing the harmonic yield by varying the position of the MIR focus relative to the Rb beam. The distribution is then recorded by introducing the diode laser. Clearly the cw diode laser significantly enhances the harmonic yield for $q \geq 5$ and depends on q. In fact, the H7 and H9 harmonics are barely detectable

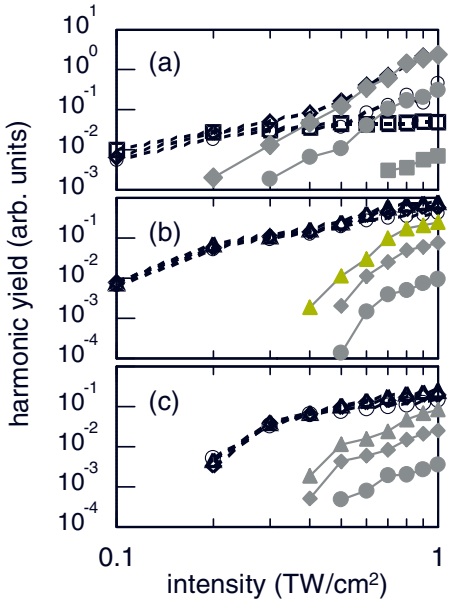

FIG. 3 (color online). Harmonic yields for (a) H5, (b) H7, and (c) H9 as a function of the MIR intensity and density. Filled symbols–solid lines: MIR alone. Open symbols–dashed lines: diode laser "on" resonance. The Rb densities are 0.2×10^{16} cc^{-1} (squares), 1.2×10^{16} cc^{-1} (circles), 2.2×10^{16} cc^{-1} (diamonds), and 3.7×10^{16} cc^{-1} (triangles). The corresponding open and filled symbols are at the same density. The excited state harmonics show no variation with density, consequently the open symbols are nearly indistinguishable.

from the MIR excitation of the ground state alone but become substantial with the diode laser present. In addition, higher resolution spectra (not shown) reveal that the enhanced harmonics, for all orders, increase in bandwidth relative to the ground state harmonics by a factor of 3.

The dependence of this enhancement on the diode laser wavelength is shown in Fig. 2(b). Both spectra are recorded under identical conditions and show that the enhancement strongly depends upon the diode laser being resonant with the $5s \rightarrow 5p$ transition. When the diode laser is off resonance, the harmonics disappear into the noise. The peak labeled *bf* (also shown in Fig. 1), which originates from the diode laser only, is the broadened cw fluorescence (discussed below) of the $6p \rightarrow 5s$ Rb transition (a fine structure doublet, $J = 1/2, 3/2$).

Figure 3 quantifies the enhancement for H5 (a), H7 (b), and H9 (c) yields for the ground and diode-coupled excited states as a function of the MIR intensity at three different Rb densities. The intensity range extends below the threshold where the $5s$ ground state alone produces these harmonics, and within the range calculated for harmonics from the $5P_{3/2}$ state. First, we observe that the enhancement factors are smallest at the highest MIR intensity (1 TW/cm^2) but are still significant (a factor of 40 for H9). As the MIR intensity decreases, the enhancement rapidly increases, reaching a maximum measured value of 10^4. In fact, the enhancement for the higher orders ($q \geq 7$) persists to such low intensity that the ground state harmonics, and therefore the enhancement factors, cannot be measured. Second, the harmonic yield as a function of density is different for the ground state and the excited target. Harmonics 5, 7, and 9 from the ground state increase with the density N as N^m with m (uncertainty in parentheses): H5, $m = 2.2$ (0.1); H7, $m = 2.7$ (0.3); H9, $m = 2.5$ (0.3). Although this scaling is higher on average than the N^2 dependence expected from phase-matching arguments [2], the statistics is not sufficient to make the difference significant and refute the quadratic power law. More surprisingly, the enhanced harmonic yield is independent of density over the Rb density range where the experiment can be conducted. This is related to the collisional quenching of the d states as discussed hereafter. The exception to this is shown in Fig. 3(a) for the two highest densities. Here the ground state H5 emission overtakes and dominates that from the excited target as the intensity increases. Finally, for our experimental conditions ($b \simeq L$), the expected geometric increase in the HHG yield, for $I > I_s(5s)$, can be estimated. Figure 3 shows that the excited state harmonics saturate at an intensity $\sim$10 times less than the ground state, which would cause a geometric increase in yield of a factor of $\sim$30. Obviously the measured enhancements are significantly larger.

The connection between the excited target and the observed enhanced harmonics is unequivocally established. Several other key observations pointing to the existence of an "essential excited state" other than the $5P_{3/2}$ can be

made. (1) A necessary condition for detecting the enhancement is the observation of the blue fluorescence [labeled "bf" in Figs. 1 and 2(b)]. (2) The $6p \rightarrow 5s$ blue fluorescence transition results from the weak cw diode laser excitation alone. (3) Correlated with the blue light is the appearance of red (0.776 μm) fluorescence from the $5d \rightarrow 5p$ transition. (4) The fluorescence line strengths depend upon the diode laser's intensity and wavelength and the Rb pressure. (5) For the conditions in Fig. 3, these fluorescence lines are essentially independent of density (changes by less than a factor of 2) and have a maximum value of 1% of the D_2 line strength. (6) Experiments conducted with saturated $5s \rightarrow 5p$ conditions but no observable blue fluorescence, e.g., low Rb density or diode fluence, result in no harmonic enhancement. Obviously, attributing the enhancement to the population in the $5p$ state is incorrect.

These observations provide clues into the origin of the harmonic enhancement. The observed $5d \rightarrow 5p$ fluorescence establishes that the presence of the *cw* diode laser can promote population into the $5d$ state. As illustrated in Fig. 1, pumping the $5d$ state is efficient for Rb due to the accidental near degeneracy of the $5s \rightarrow 5p$ and the $5p \rightarrow 5d$ transition energies. Thus a quasi-two-photon resonant transfer can occur via a three-body process involving an excited $5P_{3/2}$ atom, a ground state atom, and the red-detuned diode photon. For typical conditions, 99% of the collisions have the required 8 meV energy needed to compensate for the detuning from the $5d$ state. The $5d$ level can also be populated by the near resonant, energy pooling reaction $\mathrm{Rb}(5p) + \mathrm{Rb}(5p) \rightarrow \mathrm{Rb}(5s) + \mathrm{Rb}(5d)$ [17]. Furthermore, the fluorescence density dependence discussed in item (5) is consistent with the observed density independent enhanced harmonic yields. Our experiment shows that for the density range that results in a measurable harmonic yield (Fig. 3), the collisional quenching of the $5d$ excited state is beginning to compete with the fluorescence resulting in a nearly constant $5d$ population. At lower densities, the fluorescence yield is quadratic with density while at higher densities the high collision rate depopulates the $5d$ state. Consequently, the small population for these two limiting cases prohibited any quantitative harmonic measurements except at the reported densities. However, this does establish a crucial link between the observed enhancement and the population that passes through the $5d$ state.

According to the calculations presented in Fig. 1, the $5d$ state and most of those levels it can radiatively decay into, e.g., $6p$, are too weakly bound to produce the higher harmonic orders observed in the experiments. One (and the only) potential candidate is the more deeply bound $4d$ state which can be populated by a fluorescence transition from the $6p$ state. To explore this possibility, we have calculated the ionization rates and the dipole strengths for this state (see Fig. 1) and found that the harmonics saturate near the low intensity end plotted in Fig. 3 and that the higher orders are sufficiently strong to be consistent with the measurements.

In summary, we report the first experimental observation of HHG from excited state atoms using the scaled interaction of Rb atoms with an intense MIR fundamental field. Orders-of-magnitude enhancements in the harmonic yields have been measured from the excited medium compared with the ground state. We argue that the $4d$ state, which is populated radiatively from states that the observed fluorescence shows are excited, is the likely "essential state" for the enhancement. The formation of Rb dimers could be another source of the enhancement, but we observe no spectroscopic evidence [17] of any significant molecular formation.

The unresolved issue of this work is that the population directly promoted to the $5P_{3/2}$ state by the cw diode laser does not result in any significant enhancement contrary to single-atom calculations suggesting that this should be a very effective path for enhanced yields. Currently, we have no explanation for this unexpected observation. Additional investigations are needed to reach more rigorous comparisons with our conclusions and to resolve the disagreement between our expectations and observations.

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