Enhancement of many high-order harmonics via a single multiphoton resonance

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In the strong-field regime a single multiphoton resonance can enhance many harmonics of the fundamental laser frequency simultaneously. We demonstrate that an alkali-metal atom driven by an intense, ultrafast midinfrared pulse is an ideal system for observing this effect by calculating the response of potassium atoms to radiation that is nearly three or five photon resonant with the $4s$-$4p$ transition. We find that all the harmonics through at least the 17th are enhanced over a wide range of intensities, and that the resonance width of the enhanced harmonics is governed by the lifetime of the multiphoton resonance.

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With the observation of high-order harmonic generation in the late 1980s, nonlinear optics was expanded to include the strong-field regime [1]. This regime is characterized by the nonperturbative response of a medium to an intense driving field, when the nonlinear polarization giving rise to the qth harmonic increases with the laser intensity I much more slowly than I^q. The most striking manifestation of this nonperturbative behavior is the existence of a plateau in the harmonic spectrum in which many harmonics up to a characteristic cutoff energy all have approximately the same strength [2,3]. A more traditional route to a nonperturbative nonlinear optical response is the use of atomic resonances to enhance harmonic generation and/or frequency-mixing processes. First explored in the 1970s by Miles and Harris [4] and others, this has become a widely used means of generating coherent vacuum ultraviolet (vuv) radiation [5]. However, with the exception of a few cases in which a single harmonic was enhanced [6,7], atomic resonance effects have not been observed in the numerous studies of strong-field harmonic generation in the rare gases using visible and IR lasers. This stands in stark contrast to studies of strong-field photoionization, where clear signatures of n-photon intermediate resonances have been seen in many above-threshold ionization peaks [8,9].

In this paper, we present a theoretical study of the resonant enhancement of high-order harmonics generated in an alkali-metal atom, potassium, subject to intense 300-fs pulses of mid-infrared (MIR) radiation. As we explain below, the alkali metals at MIR wavelengths are much better suited to the observation of strong-field resonant enhancement than the rare gases previously used. We calculate the atomic response for wavelengths close to the three- or five-photon resonance between the strongly coupled ground $4s$ and first excited $4p$ states. We use intensities up to 1 TW/cm^2. At these intensities the atomic response is strongly nonperturbative, and the harmonic spectrum extends to approximately the 31st order [10]. We find enhancement of the harmonics through at least the 17th order. This enhancement of harmonics far above the resonant one (up to 12 additional photons in our calculations) is the primary difference between the present study and previous work on resonant enhancement of harmonic generation in the rare gases at IR wavelengths. Our study also extends the large body of work on the resonant enhancement of low-order harmonics in alkali-metal atoms [4,5], in several ways: First, with the recent development of intense MIR (2.5–4.5 μm) lasers [11] it is experimentally feasible to drive a *multiphoton* resonance between the $4s$ and $4p$ states, which are separated by 1.5 eV. Second, with femtosecond pulses one can efficiently drive a nonlinear process without saturation or significant population redistribution. Finally, in the strong-field regime, many harmonics can be enhanced simultaneously.

Our calculations also exhibit two effects that are characteristic of strong-field atom-laser interactions. In potassium, we find that the $4s$-$4p$ multiphoton resonance wavelength increases with intensity. This decrease in the relative energy spacing of the ground and first excited states is the opposite of what one would expect from a two-level model atom. In addition, we find that the resonance width of the enhancement can be smaller than the laser bandwidth. This is an explicitly multiphoton effect.

In order to study in detail the origin and behavior of the resonant enhancement, we consider only the response of a single atom to the intense field. Our single-atom results do allow us to make predictions about the macroscopic behavior of the resonant harmonics, and hence their experimental observability. A resonant third or fifth harmonic can be expected to undergo strong absorption as it propagates through the nonlinear medium, and may therefore be difficult to observe directly in an experiment. However, all the harmonics above the resonance exhibit the same behavior with respect to the intensity and wavelength of the driving field as the resonant harmonic, and the behavior of the resonance could therefore be studied in any of the enhanced harmonics. Furthermore, the phase of the enhanced single-atom harmonics with respect to the driving laser intensity shows no marked difference from the nonresonant case which was the subject of a previous experimental study [10]. We therefore do not expect phase-matching effects to hinder the observation of the enhancement.

To calculate the full nonperturbative response of a potassium atom to an intense laser field, we numerically integrate the time-dependent Schrödinger equation within the framework of the single active electron approximation [12,13]. We treat the interaction between the valence electron and the potassium core using the pseudopotential of Stevens *et al.*

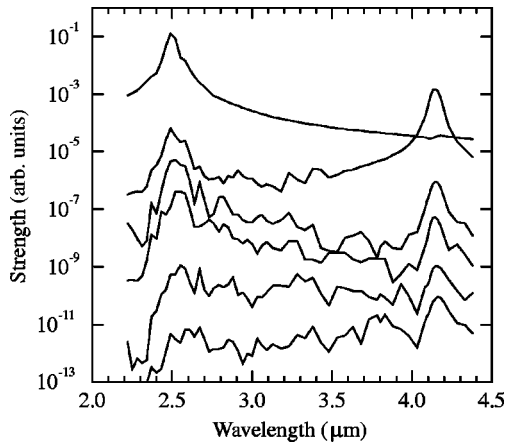

FIG. 1. Wavelength scans of several harmonics in potassium for 20-cycle FWHM Gaussian pulses with peak intensities of 0.35 TW/cm^2. From above, on the left, we show the 3rd, 5th, 7th, 9th, 13th, and 17th harmonics. Every curve consists of 63 data points, each representing the harmonic spectrum integrated over a one-photon-wide bin.

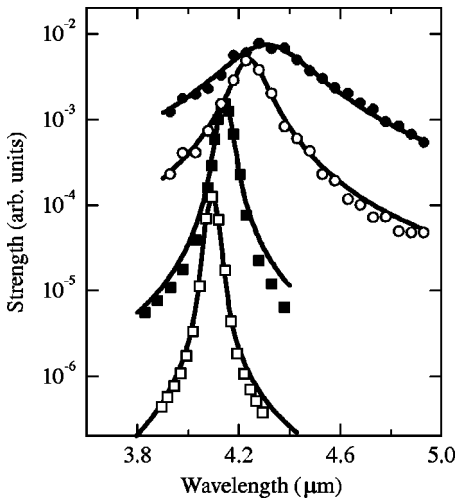

FIG. 2. The 5th harmonic yield as a function of wavelength for intensities of 0.2 TW/cm^2 (white squares), 0.35 TW/cm^2 (black squares), 0.65 TW/cm^2 (white circles), and 0.95 TW/cm^2 (black circles). The lines in the figure are calculated fits to the data points; see the text.

[14]. This potential reproduces the energy levels and transition matrix elements of the potassium atom well. The dipole spectrum is proportional to the Fourier transform of the time-dependent acceleration, calculated using the full Hamiltonian including the time-dependent laser field and the atomic potential [13]. We window the acceleration by applying a Hanning function of width equal to three times the full width at half maximum (FWHM) pulse length of the driving field [15]. The window is used to filter the laser-driven harmonic response from a strong oscillatory signal at the field free $4p$-$4s$ transition wavelength, caused by (a small amount of) population transfer to the excited state. Using the window function we can eliminate this component, which gives rise to a large background in the harmonic spectrum, without affecting the harmonic strength. Experimentally, this fluorescence signal is easily distinguished from the harmonic signal [10].

We use a linearly polarized, transform limited Gaussian laser pulse, with a FWHM pulse length of 20 optical cycles and peak intensities between 0.2 and 1 TW/cm^2. We integrate in time from -50 to 50 optical cycles, when the intensity has been reduced by more than 10^7 compared to the peak intensity. We use wavelengths ranging between 2.5 and 4.5 μm. The bandwidth $\Delta\lambda_1$ of our pulses therefore varies between 55 and 90 nm, and the pulse length varies between 150 and 300 fs.

Figure 1 shows wavelength scans of the harmonic strength at a peak laser intensity of 0.35 TW/cm^2, for selected harmonics between the 3rd and 17th. The wavelength scan includes 63 points, and each data point represents the harmonic spectrum integrated over a one-photon-wide bin around the harmonic. Resonant enhancement of harmonics 3 through 13 near 2.5 μm, and 5 through 17 near 4.2 μm, is clearly seen in the figure, corresponding to the three- and five-photon $4s$-$4p$ resonances. Both resonances occur at longer wavelengths than the field-free resonance wavelengths, which are 2.43 and 4.05 μm, respectively. Although both states are shifted toward lower energy with respect to the ionization threshold in the strong field, the downward shift of the upper state is larger (due to its interaction with the nearby $5s$ and $3d$ states), resulting in a longer resonance wavelength. At the five-photon resonance, the 5th harmonic is 50 times stronger than the nonresonant 3rd harmonic. Note however, that there is a resonant pathway to the 3rd harmonic involving the absorption of five laser photons and the emission of two laser photons. Since the 5th harmonic is much stronger than the 3rd harmonic close to resonance, the $+3\omega_1$ and the $+5\omega_1-2\omega_1$ contributions interfere and give rise to the dispersionlike shape of the 3rd harmonic curve at the five-photon resonance.

The resonant enhancement shown in Fig. 1 is built on the strong $4s$-$4p$ coupling, which accounts for more than 99% of the one-photon $4s$-np oscillator strength. We found no sign of a resonant enhancement at the much weaker $4s$-$5p$ five photon resonance (2.05 μm). It is also possible to enhance odd harmonics of the driving laser field using an even-parity resonance [4,6]. In potassium the $5s$ and $3d$ states are nearly degenerate, and have a six-photon ground- to excited-state field-free transition wavelength of 2.9 μm. We see the effect of these resonances in Fig. 1 as a shifting and broadening of the 7th and higher harmonics near the three photon $4s$-$4p$ resonance. We find that at lower intensity (not shown), the effects of three- and six-photon resonances are clearly separated, but the two resonances merge with increasing intensity.

We now examine the five photon $4s$-$4p$ resonance in more detail. Figure 2 shows the strength of the 5th harmonic as a function of wavelength, for different peak intensities of the laser field. The resonant enhancement is very large, and is still an order of magnitude at the highest intensity. The resonance wavelength changes as the intensity increases, and moves between 4095 and 4305 nm for the intensities shown. The shift is linear in the intensity, and amounts to about 6%

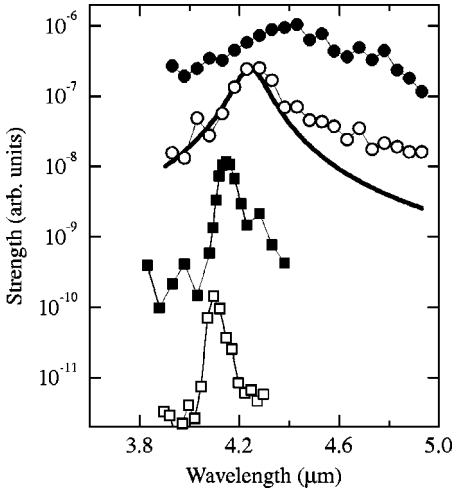

FIG. 3. Wavelength scans of the strength of the 11th harmonic, at different intensities with symbols as in Fig. 2. The data points are connected with thin lines. The thick line is the Lorentzian fit to the 5th harmonic at 0.65 TW/cm^2, scaled to match the peak of the 11th harmonic at 0.65 TW/cm^2.

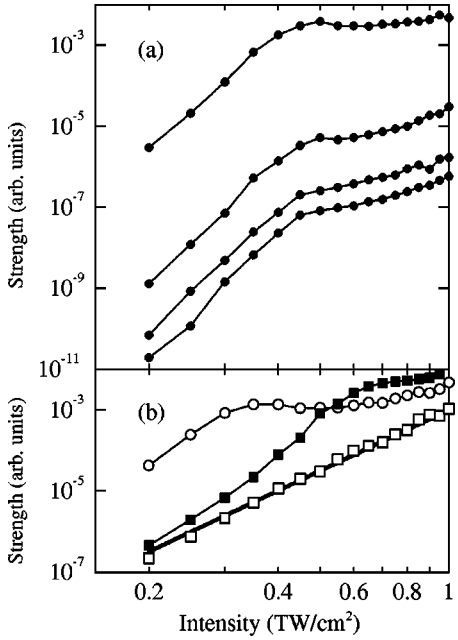

FIG. 4. In (a) the intensity dependences of the harmonics 5, 7, 9, and 11 are shown using a wavelength of 4180 nm. In (b) the 5th harmonic is shown at 3800 nm (white squares), 4130 nm (white circles), and 4280 nm (black squares). The thick solid line is proportional to I^5.

of the ponderomotive energy [16]. The width of the enhancement results from a combination of several factors. The lifetime of the dressed excited state due to ionization (and coupling to other states) will give rise to a Lorentzian line shape in the absence of other contributions. In general, however, the lifetime must be deconvoluted from the broadening induced by the laser bandwidth. Since we are considering a five-photon resonance, the bandwidth available to drive this process is on the order of $\Delta\lambda_1/\sqrt{5}$. This reduction in the effective bandwidth is clearly seen in the lowest intensity scan, which is significantly narrower (the FWHM is approximately equal to 50 nm) than the 90-nm bandwidth of the laser pulse. The thick lines shown in Fig. 2 are fits to the data points. For the two highest intensities (0.95 and 0.65 TW/cm^2) the lifetime is short, and the results are well fit by Lorentzian line shapes with widths of 356 and 260 nm, respectively. For the lower intensities, the deconvoluted Lorentzian widths are 34 nm at 0.35 TW/cm^2 and approximately 17 nm at 0.2 TW/cm^2. These correspond to lifetimes ranging from 27 to 500 fs.

The wavelength (and intensity) dependence of the resonant harmonic is repeated in the higher harmonics. As an example, in Fig. 3 we show wavelength scans of the 11th harmonic for the same peak intensities as in Fig. 2. The strength of the enhancement, as well as its position and width, correspond well to the results found in the resonant 5th harmonic. For comparison, the (scaled) Lorentzian fit to the 5th harmonic at 0.65 TW/cm^2 is superimposed as a thick line. The broadening of the harmonic to the long-wavelength side, and the additional structure, is a general trend seen in all the harmonics above the 9th, and is counter to the usual expectation that the harmonic efficiency decreases with increasing wavelength.

Figure 4 explores the intensity dependence of the harmonics close to resonance. In Fig. 4(a) the odd harmonics 5–11 are shown as a function of peak intensity at 4180 nm. At this wavelength, the five-photon resonance occurs at an intensity of 0.5 TW/cm^2. Again, all the harmonics exhibit the same intensity dependence. The behavior of the harmonics is very different above and below the resonant intensity. At low intensity they increase rapidly as a result of the intensity dependent shift of the resonant states—the detuning becomes gets smaller as the intensity increases toward the resonant value. Above the resonant intensity the harmonics become flat due to a balance between the states moving out of resonance, and the increase in strength of the nonlinear process with intensity. In Fig. 4(b) we show the 5th harmonic as a function of peak intensity for three different laser wavelengths. At 3800 nm, the $4s$-$4p$ transition is not resonant at any intensity, and the detuning from resonance becomes greater with increasing intensity. Not surprisingly, the fit to a fifth-order power law (shown as the thick line) is excellent [17]. The 4130- and 4280-nm curves again demonstrate the large change in intensity dependence near resonance. The resonant intensities inferred from the change of slope in the figure, 0.35 TW/cm^2 for 4130 nm and 0.65 TW/cm^2 for 4280 nm, are the same as were found from the wavelength scans in Fig. 2. We note that the 4280-nm intensity scan, which is further from the field-free resonance than the 4130-nm scan at zero intensity, increases as I^5 at low intensity and then steepens toward 0.65 TW/cm^2 where the resonance condition is fulfilled.

The near absence of resonant enhancement of high-order harmonics in previous strong-field studies can be understood by considering the differences between potassium at MIR wavelengths and the rare gases at IR and visible wavelengths. The excited states in the rare gases are weakly coupled to the ground state, and lie within 1–2 photons of the ionization threshold, which means that most harmonics

that could be resonantly enhanced lie well above threshold. Both of these facts are problematic in terms of achieving resonant enhancement of many harmonics, such as we have described here. A laser field which is sufficiently strong to couple two states in a rare-gas atom can also very efficiently drive tunnel ionization, the nonresonant process which gives rise to the plateau harmonics [18]. Since in harmonic generation resonant and nonresonant contributions add coherently, resonant enhancement of an above threshold harmonic is easily obscured by the large nonresonant contribution due to tunnel ionization. In resonant photoionization, on the other hand, nonresonant processes contribute only a smooth background to the photoelectron spectrum [8].

In the alkali metals, the strong coupling between the two lowest states means that one can efficiently drive a resonant coupling of the two states at intensities belonging to the multiphoton ionization (MPI) regime. This regime is characterized by the nonperturbative behavior of the high-order nonlinear polarizations at intensities where tunnel ionization is still negligible. Both working in the MPI regime *and* using a wavelength and an atom such that there are many harmonics above the resonant one that are still below or close to threshold, are crucial for obtaining the multiharmonic resonant enhancement that we find here.

In summary, we have demonstrated that by driving potassium atoms with strong MIR radiation near resonant with the $4s$-$4p$ transition, harmonics at least through the 17th (more than two photons above threshold) can be enhanced by several orders of magnitude. Strongly coupled, low-lying states such as we have considered are a feature of many atomic and molecular systems with ionization potentials substantially lower than the rare gases. The use of long wavelength (MIR) lasers in strong-field experiments can therefore be expected to lead to the observation of new multiphoton resonance effects (such as the counterintuitive increase of the resonance wavelength with intensity) in a variety of systems.

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