Intensity dependence of the phase of harmonics in one- and two-frequency laser fields

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The phase relationship between an induced atomic dipole moment and a driving laser field is an essential aspect of high harmonic generation (HHG) in a dense medium. A ''phase-matching'' criterion must be fulfilled to obtain coherent amplification of harmonic radiation, which is critical for practical HHG. We have performed numerical calculations on model atoms to survey the intensity dependence of the phase in the case of strong one- or two-frequency laser fields. The calculations show that multiphoton resonances significantly affect the yield of harmonic generation and that in the ''plateau'' regime, the phases exhibit only weak dependence upon the intensity of the driving field.

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I. INTRODUCTION

High harmonic generation (HHG) may lead to a practical source of short-wavelength coherent radiation if the conversion efficiency can be increased from that currently observed in laboratory experiments. A key factor limiting harmonic output is phase matching of the driving field and harmonic radiation. Effects of electromagnetic wave propagation and intrinsic atomic phase variation both contribute to phase mismatch of harmonic generation. Previous theoretical work has emphasized propagation effects [1], and intrinsic phase variation has begun to generate attention [2].

In particular, Shkolnikov *et al.* [3,4] have identified an optimal phase-matching condition in the case of high-order difference-frequency mixing. Their analysis relies upon the intensity dependence of the phase and amplitude of the single-atom response to a driving field. At present, these dependencies have not been systematically understood outside the perturbative regime, especially when two driving frequencies are applied. In this paper we investigate the variations of intensity-dependent phases for one- and two-frequency driving laser fields incident on a commonly used one-dimensional model atom [5–9], which has been popularized by the work of Eberly and co-workers [6,7]. This one-dimensional model incorporates the effects of intermediate dressed-state resonances, which are of key importance in understanding details of strong-field ionization [10,11,9]. We find that these resonances have a pronounced and complex effect on harmonic generation. However, in the plateau region, dependence of the atomic phase upon driving laser intensity is weak except for cases of isolated resonances.

II. ONE-DIMENSIONAL MODEL ATOM

The one-dimensional model atom has been used extensively in calculations of HHG and multiphoton ionization [5–9]. It describes an electron subject to an atomic potential of the form [in atomic units (a.u.), in which the electron mass m_e and the charge e, and the reduced Planck constant $\hbar$ have the numerical value 1]

$$U=-\frac{U_0}{\sqrt{x^2+1}}, \quad U_0=0.707\,325. \tag{2.1}$$

This potential exhibits the long-range Coulomb tail characteristic of real atomic systems, and supports an infinite series of bound states. Energies of the lowest eight states of this model atom are given in Table I. This choice of parameters gives an ionization potential identical to that of Xe, and also places an even-parity state at the same energy as a $4f$ state of Xe; this has been found to be useful in interpreting the strong-field photoionization process in that atom [9].

We have solved the time-dependent Schrödinger equation for this system in the presence of laser fields in the dipole approximation. Propagation of the wave function in time is calculated by use of the split-operator expression:

$$|\psi(t+\Delta t)\rangle=e^{-i\hat{H}\Delta t}|\psi(t)\rangle \\ =e^{-i\hat{V}\Delta t/2}e^{-i\hat{T}\Delta t}e^{-i\hat{V}\Delta t/2}|\psi(t)\rangle+O(\Delta t^3), \tag{2.2}$$

where T is the kinetic energy operator and V is the potential of Eq. (2.1) plus the potential of the laser fields $f(t)x$, treated in the electric dipole approximation. The kinetic energy contribution $e^{-I\hat{T}\Delta t}$ has been calculated by Richardson's split-operator method [12,13], in which T is approxi-

TABLE I. Energy levels of one-dimensional model atom.

Level designation	Energy (a.u.) Even parity	Odd parity
E_0	−0.445 932	
E_1		−0.158 850
E_2	−0.084 660	
E_3		−0.050 024
E_4	−0.034 018	
E_5		−0.023 960
E_6	−0.018 182	
E_7		−0.013 987

mated by a three-term finite difference formula with a grid spacing of 0.25 a.u. The box six is 1024 a.u. and an exponential-type absorbing potential [14] has been put near the boundary to suppress reflection of the wave function by the box walls. Convergence of calculation results has been positively tested by doubling the box size. The spectra are relatively stable with respect to changes of the grid spacing. The laser pulse duration D is 32 optical cycles, except where noted, with a pulse envelope shape of $\sin^2(\pi t/D)$.

We solve Eq. (2.2) over the duration of the pulse, and record the instantaneous values of the acceleration $a(t)$ given by

$$a(t)=\langle\psi(t)|-[[x,H],H]|\psi(t)\rangle=\left\langle\psi(t)\left|\frac{\partial U(x)}{\partial x}\right|\psi(t)\right\rangle+f(t). \tag{2.3}$$

In contrast to the dipole moment operator $d(t)$, Eq. (2.3) is dominated by contributions from regions near the center of potential. This feature makes a converge faster than d with respect to box size. The power spectra presented in this work are the squares of the absolute values of Fourier components of $a(t)$ averaged over the duration of the pulse. All calculations were done on a Connection Machine CM-5 parallel computer.

III. RESULTS OF CALCULATIONS

For a one-frequency driving field, results for laser frequency $\omega=0.073\,489$ a.u. ($\lambda\approx620$ nm) and $\omega=0.04554$ a.u. ($\lambda\approx1$ μm) are shown in Figs. 1 and 2, respectively. Two regimes are apparent in these power spectra. When the peak laser field strength F is small, the response is consistent with the predictions of perturbation theory, being proportional to F^{2n} where n is the harmonic order. The ramps of the seventh and ninth harmonics in Fig. 2 should come from near-resonance enhancements between ground state and E_1 and E_3, respectively. The phases of harmonics show weak variation with respect to driving field strength.

When F exceeds a threshold value, here $F\approx0.04$ a.u., the harmonic spectra enter the ''plateau'' regime in which all are of comparable intensity. The phases of harmonics in the plateau regime seem to be flat except for isolated structures.

Several resonant structures are prominent in the perturbation regime. These resonances are associated with multiphoton transitions from the dressed atomic ground state to a field-shifted Rydberg state, and they are known to be the principal mediators of atomic ionization in strong laser fields [9]. To explore these resonances, we compare the power spectra and phases for different pulse durations. Figures 1 and 3 correspond to $D=32$ and 64 optical cycles, respectively. Although the longer pulse case reveals more resonance structures and complicated variations in plateau regime, the broad appearances of the spectra are very similar. It should be noticed that the phases of harmonics in the plateau regime attain the same average values, even though their fluctuations differ.

Association of the spectral fluctuation with atomic resonances is illustrated in Fig. 4, which shows the final-state atomic populations as a function of F. Population enhancements of the states E_2, E_4, and E_6 occur at the field

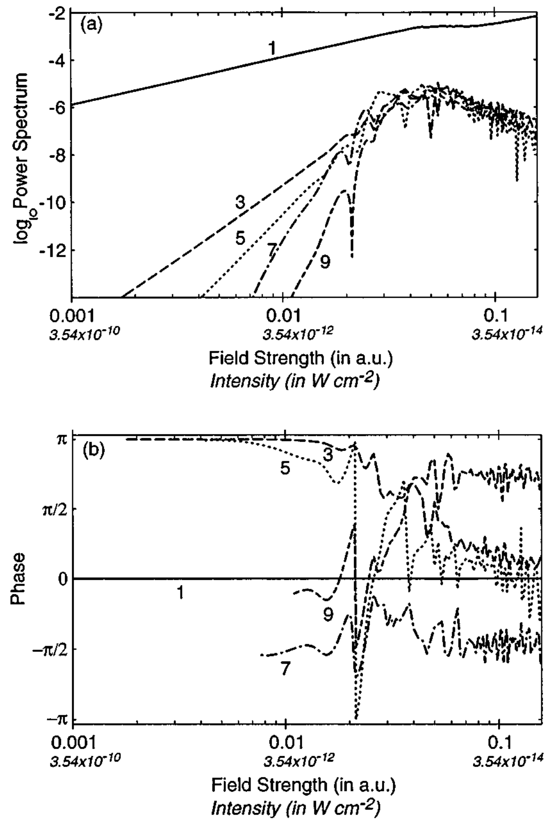

FIG. 1. Power spectrum and phase of harmonics of one-dimensional model atom. Power spectra are in arbitrary units. Laser frequency is $\omega=0.073\,489$ a.u. ($\lambda\approx620$ nm). The lines 1, 3, 5, 7, 9 are of the fundamental and the third, fifth, seventh, and ninth harmonic of ω, respectively. In (a) the power spectra are seen to follow the behavior predicted by perturbation theory at low fields, and to exhibit the plateau effect at $F>0.04$ a.u. The feature near $F\approx0.02$ a.u. is associated with a dressed-state atomic resonance identified in Ref. [9].

strengths associated with Floquet quasienergy crossings calculated by Edwards and Clark [9], which are shown in Table II. These population enhancements are accompanied by valleys in power spectra and sudden changes in phase, visible in Fig. 3. The most significant case is the change in the seventh and ninth harmonics near $F\approx0.022$ a.u., which corresponds to a six-photon transition between states E_0 and E_4. Since this is a six-photon process, it gives prominent effects on higher ($n>6$) harmonics by contributing directly to their lowest-order perturbation term, but has a lesser influence on lower harmonics: in the language of perturbation theory, this would derive from higher-order terms.

One question that motivated the present investigation concerned the possible influence of such resonances on harmonic generation, in particular whether they could be exploited in the multiphoton excitation process to enhance harmonic yields. We see that orders of magnitude changes of harmonic response are induced by these resonances, though these are most pronounced before the onset of the plateau. Nevertheless, this issue may be worth further exploration, as we can see that the harmonic yield exhibits a maximum with respect to driving field strength, located near the resonance regime. Furthermore, we believe that the sharp dips in har-

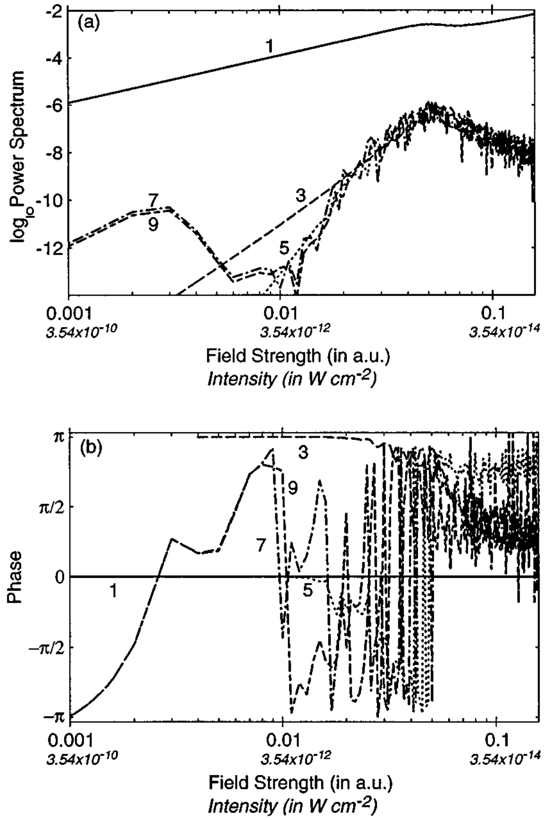

FIG. 2. (a) Power spectrum and (b) phase of harmonics of one-dimensional model atom. Laser frequency is $\omega = 0.045\,54$ a.u. ($\lambda \approx 1$ μm).

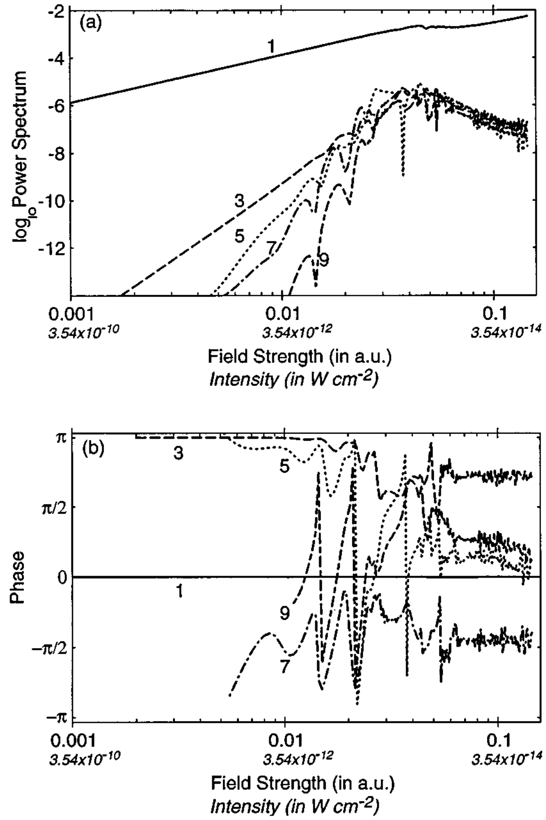

FIG. 3. (a) Power spectrum and (b) phase of harmonics of one-dimensional model atom in the same condition as in Fig. 1, except the pulse duration is doubled to 64 optical cycles.

monic yield apparent at higher intensities are also associated with atomic resonances.

For two-frequency driving fields, we set up laser fields with frequencies of $\omega = 0.045\,54$ a.u. and 3ω. The amplitude of the fields are identical. We have performed calculations with different relative phase between the two fields, and do find variations of ionization rates, intensities of harmonics, and resonance structures with phase. However, the general features summarized here are independent of phase, and the results shown in Fig. 5 for zero phase shift may be taken to be characteristic. We see that the sharp resonance features apparent in the one-color case of Fig. 2 are largely washed out, and that the phases in the plateau region exhibit smoother variation. This is probably due to the fact that a given harmonic can now be produced by a lower-order process than in the case of one frequency (e.g., the fifth harmonic can be produced by absorption of one photon of frequency 3ω and two photons of frequency ω, as well as five photons of frequency ω), and the lower-order processes are intrinsically stronger and less affected by resonances. On the other hand, the two-frequency system will be characterized by its own set of Floquet resonances, and we would expect to see their influence in the plateau region.

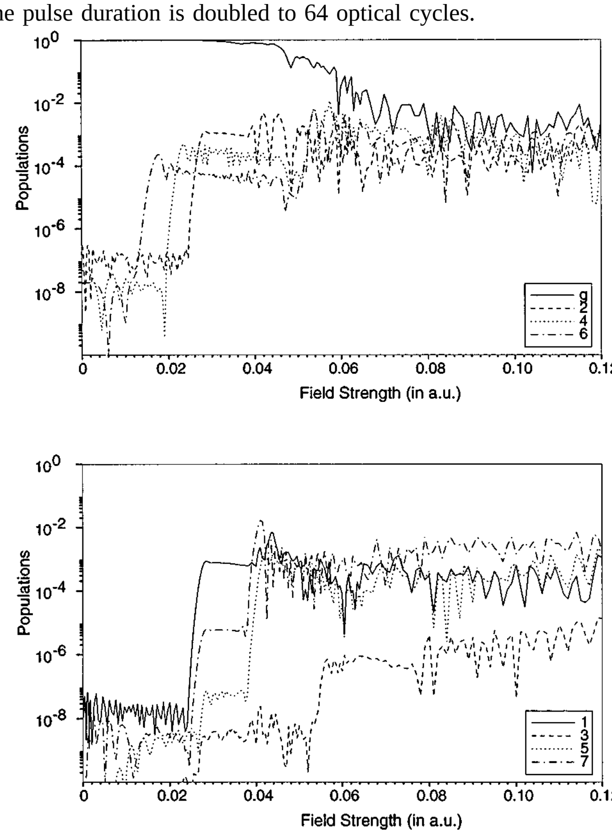

FIG. 4. Final-state population distribution of ground state and the first to the seventh excited states in pulse duration of 64 optical cycles. (a) is for even-parity states and (b) is for odd-parity states.

IV. DISCUSSION

In the one-dimensional model, the variations of intensity-dependent phases of high harmonics in the plateau regime are small, except for fluctuations near multiphoton reso-

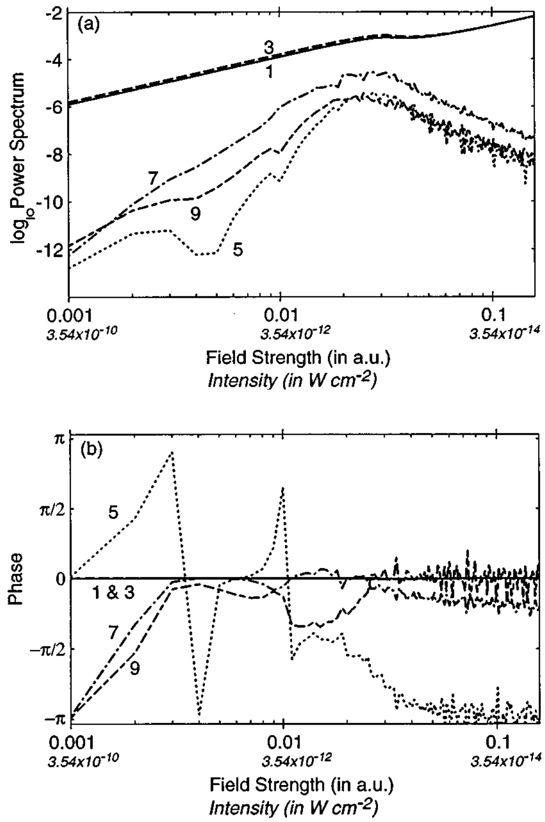

FIG. 5. (a) Power spectrum and (b) phase of harmonics of one-dimensional model atom. Laser frequency is $\omega = 0.045\,54$ a.u. ($\lambda \approx 1$ μm) and 3ω, with no phase difference.

nances. In the two-frequency case, these variations are reduced even further. The phases are not sensitive to pulse duration, though the complicated variations in the plateau regime exhibit minor changes with respect to pulse duration. The optimal phase-matching condition suggested by Shkolnikov *et al.* [3,4] should hold.

Most of the prominent structures in power spectra and phase of harmonics in the perturbation regime and in the beginning of the plateau regime can be identified with the dressed-state multiphoton resonances that are primarily responsible for ionization [10,11,9]. In the plateau regime we find a high density of such structures; although this makes it difficult to identify them uniquely with specific resonances, we believe they do originate in dressed-state multiphoton phenomena similar to those encountered below the plateau threshold.

A very different sort of behavior of the phase of high harmonics is observed in the low-frequency model proposed by Lewenstein *et al.* [15], which has been compared quite favorably to some experiments. In that model, the phase varies extremely rapidly with intensity, and there is also a good deal of structure apparent in the harmonic intensities. Those features cannot be due to atomic resonances, since there is no representation of atomic structure contained in that model. Since the model requires that the ionization potential $I \gg \omega$, it is not strictly applicable in the range of parameters we have

TABLE II. This table contains a list of the resonant intensities for model-atom levels E_2–E_8 with a laser photon energy of 0.073 489 a.u. ($\lambda \approx 620$ nm), as from the Floquet results of Ref. [10].

Level	Resonant intensity (W cm^{-2})	Resonant field strength (a.u.)
E_2	2.5×10^{13}	0.027
E_4	1.8×10^{13}	0.023
E_6	8.5×10^{12}	0.016
E_8	4.7×10^{12}	0.012

studied here. However, we felt it worthwhile to make some statements of comparison.

We have carried out calculations for the model of Ref. [15], with an additional aspect of including the effects of a finite pulse (the original model was developed for monochromatic excitation). We do this by applying the pulse summation technique in the adiabatic approximation. Under adiabatic conditions, the pulse shape envelope change is much slower than the atomic response, and we can get $x(t)$ and $d_n(t)$ in pulsed fields from $x(f)$ and $d_n(f)$ in continuous fields when the pulse strength is f at the time t. We then sum up $d_n(t)$ over the pulse duration to obtain the harmonic spectrum.

The phase under pulsed-field conditions shows much slower changes than are observed in the case of monochromatic excitation, since the pulsed case contains contributions from a range of driving intensities, and the process of summation tends to smooth the power spectrum and phase of harmonic. We have also calculated spectra of two-frequency fields with driving frequencies ω and 3ω. Under pulsed-field conditions, the phase change in two-frequency fields at the high-intensity regime is significantly slower than in one-frequency fields.

From these results, we conclude that the pulse summation technique reduces the rapidity of variation of phase changes in the low-frequency model, and that the addition of a second frequency has a similar effect. Small variations of power spectra and phases in plateau regime thus appear in both the model primarily discussed in this paper and the low-frequency model. The low-frequency model, however, does not produce any structure in the perturbation regime, since the model contains no representation of any bound states other than ground state. Thus it is not clear that this resemblance of the predictions of both models has a common origin.

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