Role of Autoionizing State in Resonant High-Order Harmonic Generation and Attosecond Pulse Production

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We suggest a high-order harmonic generation (HHG) model describing enhancement of the generation efficiency for the harmonic resonant with the transition between the ground and autoionizing state of the generating ion. The results of numerical and analytical calculations based on this model are in good quantitative agreement with the experiments showing HHG enhancement up to 2 orders of magnitude. Moreover, this model reproduces well the essential difference in HHG efficiency for different ions. We show that intense but relatively long attosecond pulses can be generated using the enhanced harmonics.

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High harmonic generation (HHG) via interaction of intense laser radiation with matter provides a unique source of coherent collimated xuv femto- and attosecond pulses. Simple, but very fruitful ''three-step model'' of the HHG in gaseous media [1,2] describes it as a result of tunneling ionization, free-electronic motion in the laser field, and recombination accompanied by the xuv emission upon the return to the parent ion. The generation efficiency is relatively low, but it can be enhanced using some resonances of the generating system, similar to resonant low order harmonic generation discovered in the very beginning of the nonlinear optics. The experimental evidence [3] of the resonant HHG was demonstrated ten years ago, but real success was achieved recently in experiments on HHG in plasma plumes [4–10] (see also recent review [11] and references therein). Generation efficiency up to 10^{-4} was achieved for the harmonic resonant with the transition from the bound to the autoionizing state of the generating ions; the enhancement (ratio of the resonant harmonic intensity to that of the neighbor harmonics) exceeded 2 orders of magnitude.

Theories describing resonant HHG involving *only* bound-bound transitions [12,13] can hardly explain the mentioned experiments. In particular, observed rapid decrease of the harmonic yield with the laser ellipticity [4] shows that free-electronic motion is essential for the resonant HHG in these experiments: when the electron accelerated by the elliptically polarized field misses the parent ion, the recombination probability rapidly decreases. Several resonant HHG theories contain the free-electronic motion [14–19]. The theories taking into account single- or multiphoton resonance of the laser field with bound-bound transitions [14–17] predict enhancement of multiple harmonics in contrast to the strong enhancement of a single harmonic (or very few ones) observed in the mentioned experiments. Another model [18] predicts enhancement of the harmonics resonant with the transitions of the *parent* ion. Finally, a theory explaining the resonant HHG between ground and autoionizing state of the generating atom (or ion) was suggested recently [19]. However, this approach assumes high initial population of the autoionizing state and its coherence with the bound state. For the HHG in plasma plumes, this assumption seems unrealistic. Thus, none of the existing resonant HHG models can adequately explain the huge generation enhancement observed in the mentioned experiments.

In this Letter, we suggest a modification of the three-step model for generation of the harmonic resonant with the ground-autoionizing state transition as illustrated in Fig. 1 ($\Omega \approx q\omega$, where Ω and ω are the transition and the laser frequencies, q is the resonant harmonic order). For this harmonic, the contribution of the usual three-step process is certainly relevant; however, that of the following modified process can be more essential. Instead of the last step (radiative recombination from continuum to the ground state), the free electron is trapped by the parent ion, so that the system (parent ion + electron) lands in the auto-

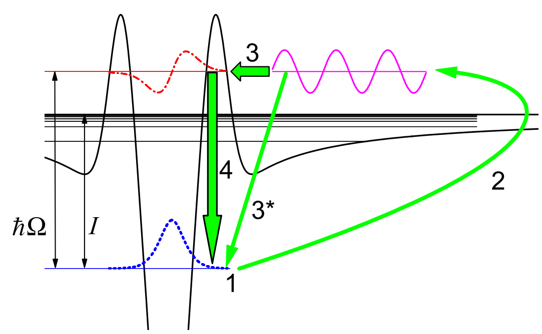

FIG. 1 (color online). The ''simple-man'' three-step model includes ionization (step 1), free-electronic motion in the field (2), and recombination (3*); in the suggested model, the latter stage is replaced by the radiationless transition to the autoionizing state (3), and relaxation with xuv emission (4). Solid black curve presents the model potential given by Eq. (1) in which the electron has the ground state (dotted blue line), multiply bound exited states and one quasistable excited state (dashed-dotted red line).

ionizing state (step 3 in Fig. 1), and then it relaxes to the ground state emitting xuv (step 4).

At first glance, the replacement of the single recombination step 3* by the two steps would decrease the probability of the process. However, large inelastic scattering cross section (controlling step 3) and high oscillator strengths of some ground-autoionizing state transitions (controlling step 4) result in higher probability of the 4-step process, as we show in this Letter with a numerical and analytical approaches. In a sense, the autoionizing state of the atom (or the ion) here is similar to the compound nucleus playing a key role in some nuclear reactions.

Describing autoionizing states assumes multielectron dynamics study; therefore, accurate numerical simulation of our model is problematic. So a single-active electron approximation is used in our numerical investigation, and the role of the other electrons is reproduced with a model potential of the *parent* ion. Since most experiments demonstrate resonant HHG with singly charged ions, we consider doubly charged parent ions. We are using the potential (atomic units are used throughout)

$$V_Z(\mathbf{r}) = -\frac{2}{\sqrt{a_0^2 + r^2}} + a_1 \exp\left[-\left(\frac{r - a_2}{a_3}\right)^2\right]. \quad (1)$$

The first term is the standard "soft Coulomb" potential, and the second one provides a barrier making possible bound quasistatic states with positive energy; these states reproduce the autoionizing states of the ion.

Below, we simulate HHG with the In^+ ion. The oscillator strength of the transition from the ground to the autoionizing state $4d^{10}5s^2\ ^1S_0 \rightarrow 4d^9 5s^2 5p(^2D)^1P_1$ essentially (by a factor greater than 12) exceeds the strengths of the other transitions [20], and the transition frequency is very close to the resonance with the 13th harmonic of the 800 nm radiation. We neglect other autoionizing states and choose the parameters of the potential (1) so that there is only one quasistatic state in this potential. The choice of the parameters allows reproducing the energy of the ground and the autoionizing state, as well as the decay time of the autoionizing state of the In^+ ion. Figure 1 presents the used potential, the ground, and quasistatic states. Note that they have different parity, so the transition is allowed, as it in fact is in the real ion. Localization of both states near the origin provides high oscillator strength of the transition between them.

We solve numerically axially symmetric three-dimensional Schrödinger equation (TDSE) for an electron in the described potential exposed to an external laser field. The numerical methods are described in [21]. The pulse intensity (10^{15} W/cm^2) and duration (40 fs) are close to those used in the experiments [4,9].

Figure 2 shows the calculated spectrum. The resonant harmonic is much more intense than all the others. The enhancement (ratio of the 13-th harmonic intensity to the average for the 11-th and the 15-th) is 83.

Similarly, the parameters of the model potential for Sn^+ were chosen to reproduce the transition $4d^{10}5s^2 5p\ ^2P_{3/2} \rightarrow 4d^9 5s^2 5p^2(^1D)^2D_{5/2}$, which is resonant with the 17-th harmonic and has a very high oscillator strength [22]. The TDSE calculation yields the enhancement value of 20. Both values (for In^+ and Sn^+) are close to the experimental ones, which we discuss in details in the final part of this Letter.

Attosecond pulse production using a group of several neighbor high harmonics is possible because the phases of the harmonics are linked to each other [23]. The harmonic phases are controlled mainly by the electronic motion in continuum [24]. This step is accounted for identically in the three-step model and in our one. Thus, the model we suggest provides the harmonic phase locking as well.

Production of an attosecond pulse train requires several harmonics with similar intensity. Thus, the enhancement of a single harmonic is not sufficient for a proportional increase in attosecond pulse intensity. In our calculations, a very pronounced enhancement of the resonant harmonic is accompanied by a much smaller, but still important intensity increase for a group of surrounding harmonics. The attopulse train resulting from the harmonic group is shown in the inset in Fig. 2. The calculated attosecond pulse duration of 240 as is similar to the one obtained experimentally using resonant HHG in chromium plasma plume [25]. The domination of a single harmonic in the calculated spectrum leads to relatively long attopulses and pronounced pedestal in the train. Strong enhancement of a *group* of harmonics is possible using an ion having several transitions with suitable frequencies and similar oscillator strengths, or a mixture of ions. HHG in such medium can provide relatively short attosecond pulses with energy enhanced commensurably with the increase in resonant harmonic intensity.

Now we will describe the analytical approach for calculation of resonant harmonic production enhancement. Namely, we provide an estimate for the resonant HHG

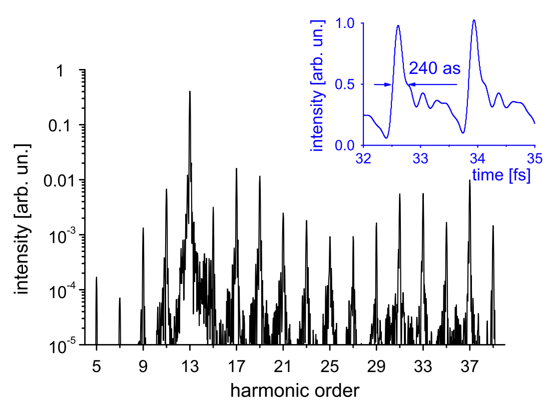

FIG. 2 (color online). Intensity spectrum found numerically for the model In^+ ion. The inset shows the attosecond pulse train obtained using harmonics from the 9-th to the 25-th.

enhancement as a ratio of the harmonic intensity generated via the four-step and three-step models. We assume that the process is periodic, so the free wave packet in the vicinity of the origin can be expanded in Fourier time series. Each component of this series corresponds to a wave which appeared in the continuum from the ground state after absorbing q field quanta (below we use the notation [26])

$$\chi(p) = c(p)\exp(ipz) \tag{2}$$

where the momentum is $p = \sqrt{2(q\omega - I)}$.

We shall see below that the amplitude of this wave $c(p)$ determines the intensities of the harmonic emission both due to the three-step and the four-step process. The amplitude cancels when calculating the *ratio* of the two intensities. This allows deriving relatively simple expressions describing the harmonic enhancement.

The harmonic amplitude due to the transition from the continuum to the ground state in the three-step process is calculated as the quantum-mechanical expectation value of the force acting on the electron. Using Eq. (2), the amplitude can be written as (see [26])

$$f_{c\text{-}g} = ac(p)pM(p)Z^* + \text{c.c.} \tag{3}$$

where a is the amplitude of the ground state and $Z^* = \sqrt{I/\mathcal{R}}$ is the effective charge of the parent ion, $\mathcal{R}$ is one rydberg, $M(p) = 4i\sqrt{\pi}[p - \arctan(p)]/p^3$.

Further using the four-step model, we derive the amplitude for the resonant harmonic. The autoionizing state is populated due to inelastic scattering of the continuum electronic wave χ and depopulated mainly via photoionization. Thus, this state's population $\tilde{s}$ is described by the equation $d\tilde{s}/dt = p\sigma|c|^2 - \tilde{s}\Gamma_{\text{ph}}$, where σ is the inelastic collision cross section and Γ_{ph} is the photoionization rate. Using this equation we find the equilibrium population s as

$$s = p\sigma|c|^2/\Gamma_{\text{ph}}. \tag{4}$$

Our additional numerical calculations show that for the considered laser intensities, the depopulation rate of the autoionizing state is close to the laser frequency: $\Gamma_{\text{ph}} \approx \omega$.

The second derivative of the dipole moment of the ion due to autoionizing ground state transition is written in terms of the states' population and the transition oscillator strength f_{osc} as (see [27])

$$|f_{ai\text{-}g}| = \Omega^2 a\sqrt{sf_{\text{osc}}/[2(2l+1)\Omega]} \tag{5}$$

where l is the azimuthal quantum number of the autoionizing state.

Finally, using the expression for the inelastic collision cross section σ from [28] and Eqs. (3)–(5), we find the enhancement as

$$\frac{|f_{ai\text{-}g}|^2}{|f_{c\text{-}g}|^2} = \frac{2\pi\Omega^3 f_{\text{osc}}}{\omega p^3[Z^* M(p)]^2}\left[\frac{\Gamma^2}{4(\Omega - q\omega)^2 + \Gamma^2}\right] \tag{6}$$

where Γ is the autoionizing resonance width.

Using the published data on the ionic autoionizing states [20,22,29–35], we calculate the resonant harmonic enhancement with Eq. (6), for singly charged ions and 800 or 400 nm generating field. Figure 3 summarizes our numerical and analytical results for various ions as well as published experimental data.

Analytical and numerical results agree rather well. The difference between them mainly originates from our numerical model's lacking precision in reproducing the oscillator strengths of the ground-autoionizing state transition. Namely, these values are 0.81 for In^+ and 0.79 for Sn^+ in our numerical model, whereas in real ions, they are 1.11 and 0.304, respectively [20,22]. This leads to an underestimation of the enhancement for In^+ and its overestimation for Sn^+ in our numerical calculation.

Fundamental or second harmonic of Ti:Sapp laser was used in the experiments. In the latter case, the points in the figure are marked as "400 nm," and the harmonic order is shown with respect to the 800 nm fundamental (15×2 for Cr^+ and 17×2 for Mn^+).

Figure 3 shows that while the enhancement values for different media differ almost 2 orders of magnitude, the theoretical results are close to the experimental ones. The difference between them is attributable to the medium effects (harmonic absorption and detuning from the HHG phase matching) that are not taken into account in this theory. The plasma parameters presented in paper [9] for the experiment with Mn^+ and the absolute cross section [35] of uv photoionization for Mn^+ allows concluding that the absorption should essentially reduce the enhancement. On the other hand, phase-matching effects can increase or decrease the enhancement depending on the ionic refrac-

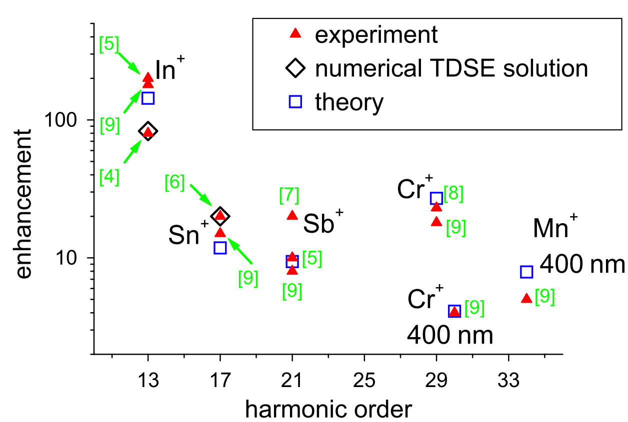

FIG. 3 (color online). Comparison of the published experimental data on the resonant harmonic enhancement with our analytical and numerical results. References to the experimental papers are presented in the figure.

tion index, which can be essential [36] for the xuv frequency near the resonance.

Thus, in this Letter, we suggest the four-step model of the resonant HHG due to the autoionizing state, and develop the numerical and analytical approaches to describing this process. In particular, we have found Eq. (6) describing microscopic resonant harmonic enhancement. Numerical and theoretical enhancement values are close to those measured experimentally. Our model predicts the phase locking of the resonant harmonics which leads to production of intense but relatively long attosecond pulses. This model can be used while searching for new media providing effective harmonic generation, as well as shortening of the attopulses generated via resonant HHG. Moreover, application of this model opens a wide field where numerous well-developed issues of the nonresonant HHG (phase-matching optimization, attopulse generation gating, etc.) can be generalized to the resonant case.

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