Simple model for harmonic generation from atomic clusters

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We model the response of an atomic cluster to an intense laser pulse by numerically solving the time-dependent Schrödinger equation for a simplified one-dimensional system. We address the questions of the generation of high-order harmonics of the laser frequency and of the ionization dynamics of the atoms within the cluster for very short laser pulses. [S1050-2947(99)09111-8]

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I. INTRODUCTION

Recently, medium-sized rare-gas clusters with a few thousands of atoms have been shown to provide harmonic yields significantly higher than those obtained from the monomer gas with the same average density [1,2]. The experimental findings can be summarized as follows: (i) for not-too-high-order harmonics ($N=3-9$), up to fivefold enhancements have been observed; (ii) a N_0^3 scaling law in terms of the average atomic density N_0 has been suggested [2]. This contrasts notably with the usual N_0^2 dependence observed in monomer gases.

We note that harmonic emission has been observed at rather moderate laser intensities, namely, around $I_L \approx 10^{14}$ W/cm^2. At significantly higher intensities, hard x rays and fast highly charged ions, as well as hot electrons, are produced. It is by now commonly accepted that these effects result from the response of the underdense plasma created after the atoms in the cluster have been stripped from their outer electrons [3–6]. Here we shall restrict ourselves to the moderate intensity regime and, hereafter, we will concentrate the discussion on harmonic generation and on the early stages of atomic ionization within the cluster.

The above-listed items will be addressed with the help of a simplified one-dimensional model which retains the essential physical properties of a small cluster in which atoms are loosely bound together via van der Waals–like interactions. As we shall show, the dominant features of the harmonic spectra can be deduced from the analysis of the time-dependent collective dipole, as obtained from the resolution of the time-dependent Schrödinger equation (TDSE) for such a system. The organization of the paper is as follows: In Sec. II we shall present the theoretical background. Selected numerical results will be presented and discussed in Sec. III and a brief conclusion will end the paper in Sec. IV.

II. MODEL AND THEORY

The cluster is modeled by a linear chain of one-electron atoms, located at regular intervals. Each atomic electron is submitted to the potential of its nucleus (or ionic core). More precisely, within the Jth ''atom'' of the row, the electron with coordinate x_j experiences the so-called ''soft-Coulomb'' potential [7],

$$V_J^{(0)}(x_j) = -\frac{1}{\sqrt{a^2+(x_j-X_J)^2}}, \tag{1}$$

where the parameter a is adjusted to reproduce the ionization energy of the atom considered and X_J is the location of the supposedly fixed nucleus. The row is made of equally spaced atoms with $X_{J+1}=X_J+d$, where d is the interatomic distance. Typical values for d are around $d=6$ a.u., which is representative of van der Waals clusters. Atomic units will be used throughout, unless otherwise mentioned.

In the initial state, at $t=0$, it is assumed that an atomic electron is submitted to this sole potential. Its wave function, denoted $\Phi_J(x_j,0)$, is determined numerically by solving the corresponding time-independent Schrödinger equation. This assumes that it does not ''see'' the neighboring atoms. The assumption is certainly correct when applied to atoms pertaining to van der Waals clusters. However, things can change dramatically in the presence of a strong external laser field, as explained below.

At $t>0$, i.e., after the laser is turned on, we assume that an atomic electron is submitted, in addition to the above potential of its parent nucleus, to two time-dependent potential terms representing, respectively, (i) the electron-laser interaction,

$$H_{Int}(x_j,t) = -x_j F(t)\sin(\omega_L t), \tag{2}$$

where $F(t)$ represents the envelope of the time-dependent laser field with frequency ω_L, and (ii) the additional potential resulting from the presence of the (time varying) electronic charge densities in neighboring atoms. For the jth electron, pertaining to the Jth atom, we have chosen to model it as follows:

$$V_J(x_j,t) = \sum_{(I,i)\neq(J,j)} [V_J^{(I)}(x_j,t) - V_J^{(I)}(x_j,0)]. \tag{3}$$

The structure of the latter potential can be understood as follows: First, it is assumed that the ith electron, when in the state $\Phi_I(x_i,t)$ located close to $x_i \approx X_I$, exerts on the jth electron an averaged (repulsive) potential of the form

$$V_J^{(I)}(x_j,t) = \int_{-\infty}^{+\infty} \frac{|\Phi_I(x_i,t)|^2}{\sqrt{b^2+(x_j-x_i)^2}}\, dx_i. \tag{4}$$

This corresponds to the contribution of the first term in Eq. (3). We mention that such a treatment is reminiscent of the model introduced recently to account for the electron correlations in the process of double ionization of helium [8].

On the other hand, the contribution of the second term in Eq. (3) is an indirect representation of the attractive potential created by the nucleus located at X_I. We have chosen to represent it as the counterpart (i.e., with reversed sign) of the repulsive potential exerted by the ith electron when it is in the ground state of the Ith atom, at $t=0$. This has the advantage of being consistent with our hypothesis that, in the initial state, atoms do not ''see'' each other, i.e., $V_J(x_j,0)=0$. As a further check of the validity of this approximation, we have verified that, for the set of parameters used here, the ground-state atomic wave function is not significantly affected by the presence of other atoms. One notes also that, in the presence of the laser, as the electron probability densities are strongly driven by the field and photoionization starts to take place, local minima, approximately located at the atomic positions X_I, begin to show up in $V_J(x_j,t)$, thus accounting for the appearence of positive ions within the cluster. This feature can be conveniently displayed by considering the effective potential $V_{J,\mathrm{eff}}(x_j,t)$, experienced by the electron in the Jth ''atom,'' which is obtained by combining Eqs. (1) and (3),

$$V_{J,\mathrm{eff}}(x_j,t)=V_J^{(0)}(x_j)+V_J(x_j,t). \tag{5}$$

In order to illustrate the evolution in time of $V_{J,\mathrm{eff}}(x_j,t)$, we have numerically solved the TDSE for a chain of five ''Neon'' atoms, with ionization potential $I_p\approx 0.79$ a.u. [i.e., $a=0.81$ in Eq. (1)]. In the electron-electron repulsion terms, Eq. (4), we have chosen the parameter $b=\sqrt{2}$. We will come back to this choice below. The laser frequency is $\omega_L=0.057$ a.u. ≈ 1.55 eV and the time dependence of the field amplitude is modeled by a linear turnon with a duration of one laser period T_L, followed by a plateau extending over more than $16T_L$. Then, the wave function for each atomic electron is computed self-consistently, taking into account the potentials, Eqs. (1)–(3).

We display first the effective potential $V_{3,\mathrm{eff}}(x_3,t)$, experienced by the electron in the atom at the center of the chain, at different times and for two representative laser intensities. In Figs. 1 it is shown for $I_L=2\times 10^{14}$ W/cm^2 at times around, respectively, $t=2\ T_L$, i.e., at the beginning of the pulse [Fig. 1(a)], and $t=16\ T_L$, i.e., when the field is well established and the influence of transients can be safely neglected [Fig. 1(b)]. As compared to the field-free atomic potential, one observes the appearence of local minima located approximately at the positions of the other four atoms (in fact, their positions vary periodically in time around the positions of the fixed nuclei, depending on the orientation and magnitude of the field). At such an intensity, ionization remains negligible for the relatively short time delay considered, the total ionization yield not exceeding 5%, at $t=16T_L$. In fact the effective potentials are very similar at $2T_L$ and at $16T_L$. This suggests that the observed modifications result from the polarization of the electronic clouds induced by the laser field which renders less effective the screening of the other nuclei by their own atomic electron. A consequence of this feature is that, within the cluster, an atomic electron can experience an additional time-dependent anharmonicity while being driven by the laser field, as compared to the single-atom case. As we shall show below this feature is reflected in the harmonic spectra generated by atoms located within the cluster.

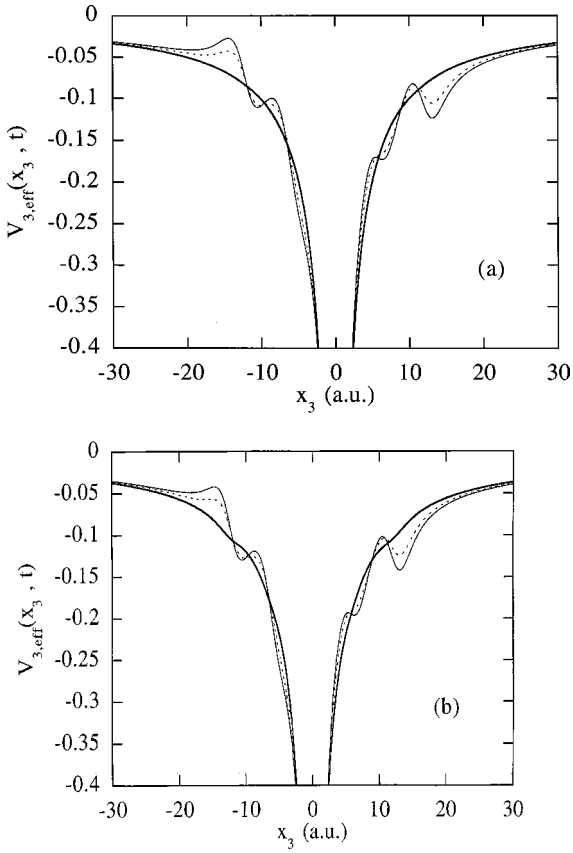

FIG. 1. Time-dependent effective potential $V_{3,\mathrm{eff}}(x_3,t)$ experienced by the electron in the third atom for a chain of 5 neon atoms; see Eq.(5). The intensity of the laser field is 2×10^{14} W/cm^2 and the frequency is $\omega_L=0.057$ a.u. (a) $t=2T_L$: thick line, $t=2T_L+T_L/8$: dotted line; $t=2T_L+T_L/4$: thin line; (b) $t=16T_L$: thick line, $t=16T_L+T_L/8$: dotted line, $t=16T_L+T_L/4$: thin line.

At higher laser intensities, i.e., when ionization becomes important, the effective potential experienced by an atomic electron at the center of the chain is even more strongly modified, as compared to the one of the isolated atom. As the ionization yields increase, the screening of the neighboring nuclei by their electron is less effective and the barrier between atoms is lowered, the situation becoming reminiscent of a metal cluster. This situation is illustrated in Fig. 2(a), where $V_{3,\mathrm{eff}}(x_3,t)$ is computed at times around $t=16\ T_L$, for an intensity $I_L=2.7\times 10^{14}$ W/cm^2. The shape of this effective potential clearly indicates that, when combined with the electron-laser interaction potential, Eq. (2), the ionization barrier is significantly lowered, see Fig. 2(b). This feature accounts for the fact that atoms within a cluster are more easily ionized than when isolated [9].

The situation for an electron belonging to an outer atom is illustrated in Fig. 3, where the effective potential for the electron within the fifth atom of the row is shown for $I_L=2\times 10^{14}$ W/cm^2, the same as in Fig. 1(a). Again, local minima appear, approximately located at the positions of the

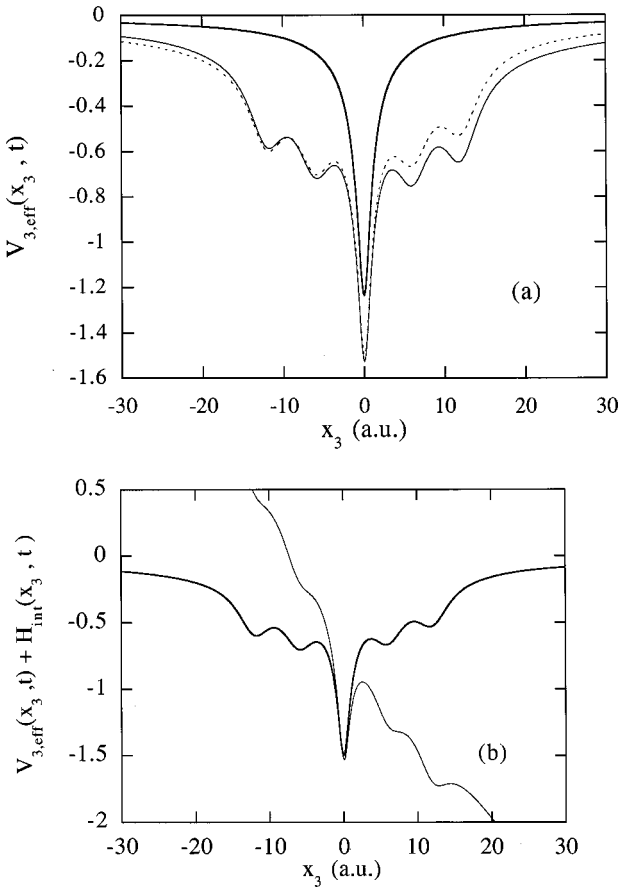

FIG. 2. (a) Time-dependent effective potential $V_{3,\mathrm{eff}}(x_3,t)$ experienced by the electron in the third atom for a chain of 5 neon atoms; see Eq.(5). The intensity of the laser field is 2.7×10^{14} W/cm^2 and the frequency is $\omega_L = 0.057$ a.u. $t=0$: thick line; $t=16T_L$: dotted line; $t=16T_L+T_L/4$: thin line; (b) effective potential combined with the electron-laser interaction for $t=16T_L$ (thick line) and $t=16T_L+T_L/4$: thin line.

other nuclei. Here, however, ionization will be favored in one direction, i.e. each half-cycle, and the anharmonicity is only one-sided, a fact which is likely to influence harmonic generation by these atoms. We turn now to the analysis of the harmonic spectra, as deduced from the Fourier analysis of the time-dependent dipole acceleration of the system.

III. NUMERICAL SIMULATIONS OF HARMONIC SPECTRA

By solving the TDSE for the above model, one can compute harmonic spectra generated either from individual atoms within the cluster or from the cluster itself. It is also an easy matter to determine approximate ionization yields from the norm of the corresponding wave functions. The results of such an analysis are presented next.

Broadly speaking, one expects that harmonic yields from a cluster of N atoms exhibit at least a standard N^2 increase with respect to the single-atom response. The question at stake here is whether or not one observes deviations from this simple law. This can be answered by comparing the harmonic yields from chains of N atoms with the ones obtained from an isolated atom. To this end we have defined, for a given harmonic with frequency ω and at a fixed laser

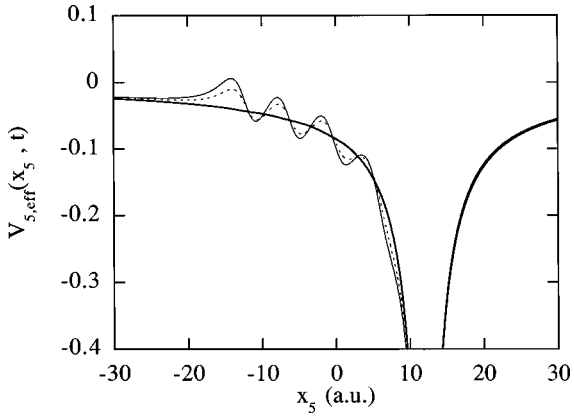

FIG. 3. Time-dependent effective potential $V_{5,\mathrm{eff}}(x_5,t)$ experienced by the electron in the fifth atom for a chain of 5 ''Neon'' atoms; see Eq.(5). The intensity of the laser field is 2×10^{14} W/cm^2. $t=2T_L$: thick line; $t=2T_L+T_L/8$: dotted line; $t=2T_L+T_L/4$: thin line.

intensity, the following scaled ratio:

$$R_N(\omega) = \frac{1}{N^2} \times \frac{I_N(\omega)}{I_1(\omega)}, \tag{6}$$

where $I_N(\omega)$ is the harmonic intensity computed for a chain of N atoms, while $I_1(\omega)$ corresponds to a single atom. In accordance with the definition, a value of $R_N(\omega)=1$ implies that there is no effect from the cluster structure of the system, as compared to separated atoms.

The dependence of such ratios in terms of the harmonic frequencies for chains of $N=3$, 5, and 7 atoms and at different field intensities, is shown in Figs. 4(a)–4(d). It appears first that for the lowest intensity considered, $I_L = 1 \times 10^{14}$ W/cm^2 [10], the ratios are consistently larger than unity and most often grow with the cluster size, see Fig. 4(a)–4(d). This trend is observed for almost all the harmonics pertaining to the plateau, which extends here up to around $\omega \approx 29\ \omega_L$. We note that the position of the cutoff agrees well with the ''$I_p + 3U_p$'' law which applies to harmonic spectra in single atoms. Here I_p is the ionization potential of the atom and $U_p = F_0/(4\omega_L^2)$ is the ponderomotive energy of a free electron within the field, [11]. On the other hand, we attribute the notable fluctuations observed for $R_N(\omega)$, when considering different harmonic orders, to the fact that emission yields can vary by several orders of magnitude for a given harmonic as compared to the average value observed in the plateau. An example of this atypical behavior is provided here where $R_N(7\omega_L) > 20$, for $N=\{3,5,7\}$ (not shown). This large value results from the fact that the single-atom yield for the seventh harmonic $H7$ is very small compared to other harmonics within the plateau, thus artificially increasing the ratio. Similar considerations hold for the fifteenth, $H15$, and twenty-fifth, $H25$, harmonics when generated by chains of five and seven atoms.

At $I_L = 2 \times 10^{14}$ W/cm^2, the efficiency of harmonic emission from clusters increases significantly beyond the N^2 law and larger clusters seem to be favored. This is shown in Fig. 4(b), where one observes that the ratios $R_N(\omega)$, from a chain of $N=7$ atoms, are most often above those from $N=5$ and

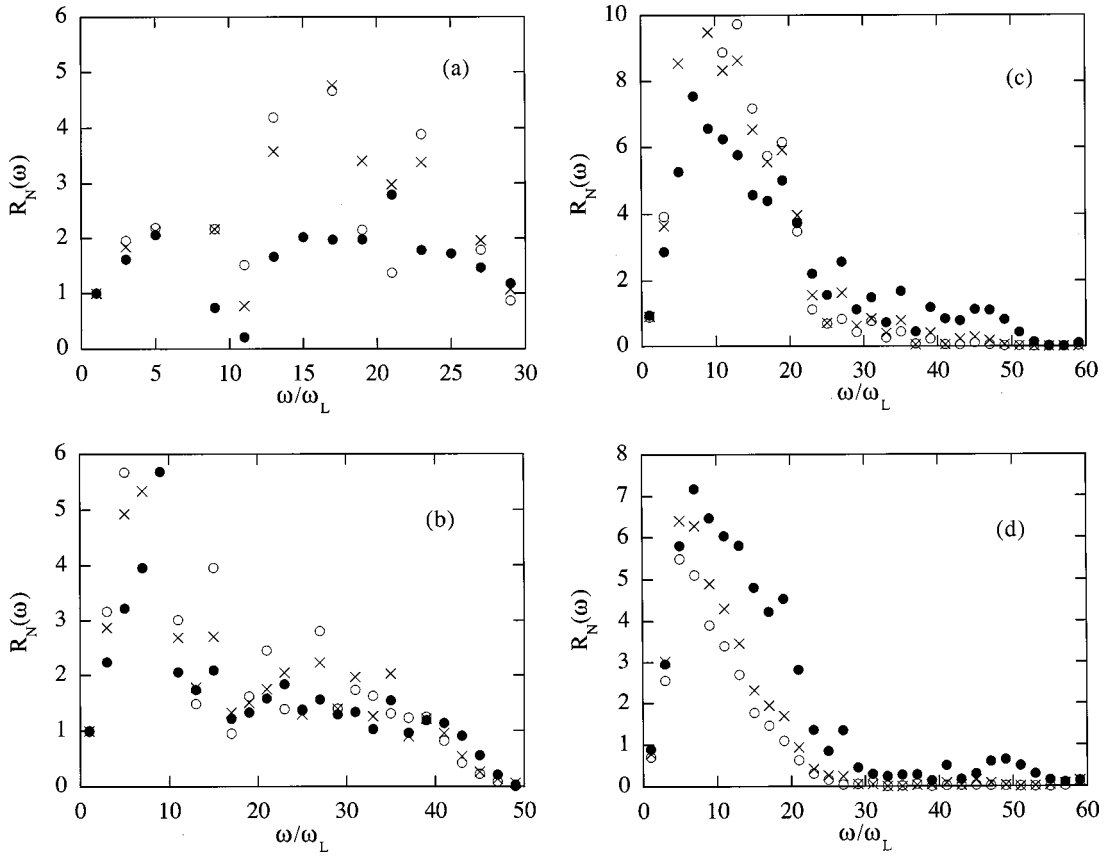

FIG. 4. Ratio R_N, as defined in Eq. (6), as a function of the harmonic order ω/ω_L for chains of $N=3$ (●), 5 (×) and 7 (○) neon atoms. The laser frequency is $\omega_L=0.057$ a.u. (a) $I_L=10^{14}$ W/cm^2; (b) $I_L=2\times10^{14}$ W/cm^2; (c) $I_L=2.5\times10^{14}$ W/cm^2; (d) $I_L=2.7\times10^{14}$ W/cm^2.

from $N=3$. Enhancement factors larger than four are not uncommon. However, a feature is to be noticed: this does not apply to the harmonics beyond the thirty-seventh, $H37$, the single-atom cutoff being located around $H43$ at the considered intensity. This result indicates that, for higher harmonics, clusters may be less efficient than a sample of isolated atoms with the same average density.

This observation is confirmed at higher intensities, as shown in Fig. 4(c) where the variations of the ratios $R_N(\omega)$ are displayed for $I_L=2.5\times10^{14}$ W/cm^2. At this intensity, the harmonic yields from a chain of $N=7$ atoms are always larger than those from chains of $N=5$ and $N=3$ atoms. Enhancement factors larger than eight are even observed for $H5$, $H7$, and $H9$. But the $N=7$ chain keeps being advantaged only for harmonics up to $H19$ and, beyond $H23$, the situation is completely reversed: then the three-atom chain is more efficient than larger ones. Moreover, beyond $H35$, there is no evidence that clusters could provide larger yields than single atoms. We note again that this result is not related to the extension of the plateau which goes up to $H47$ in the single-atom spectrum.

Most interestingly, it appears that at a slightly higher intensity, namely at $I_L=2.7\times10^{14}$ W/cm^2, the seven-atom chain becomes less efficient than smaller ones, see Fig. 4(d). For the three-atom chain, enhancement factors larger than five are still observed for the harmonics $H5-H13$, while for longer chains such enhancements are observed only for $H5$ and $H7$. Moreover, beyond $H25$, one observes that clusters are *less efficient* than single atoms. This suggests that large clusters are not a suitable medium for obtaining larger yields for higher harmonics in the plateau (here, it extends up to $H49$ in the single-atom spectrum). We have checked that this tendency is amplified at higher field strengths.

We attribute this property of clusters to the fact that, at a given laser intensity, ionization takes place more easily within a cluster than in an isolated atom. In other words, an isolated atom can withstand higher field intensities without being ionized, thus yielding higher harmonic emission rates in the high intensity regime. This interpretation is supported by two complementary arguments. First, the effective potentials, Eq. (5), experienced by the atomic electrons within the cluster, clearly show that the barrier against ionization is much lowered as compared to the single-atom case, see Figs. 2, and 3. As shown next, a second, more quantitative, argument is provided by the comparison between ionization yields from different chains of atoms.

Rough estimates of the ionization yields for a given atom in the row, can be obtained by computing the projection of the time-dependent wave function, solution of the TDSE, onto the bare ground-state function at times multiple of the laser period (i.e., when the laser field is zero). This provides an estimate of the electron population density remaining in the vicinity of the considered nucleus and, by difference to unity, an approximation of the ionization yield. By summing and averaging over the contributions of each atom, it is then possible to derive an approximate value of the ionization yield for the entire row. For the above set of parameters, the dependence of these averaged yields at times $t=16T_L$ and

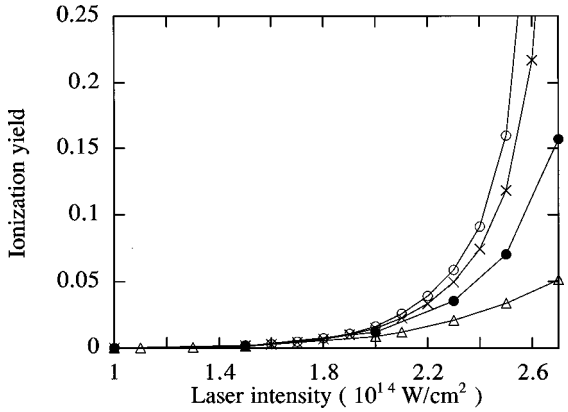

FIG. 5. Ionization yield as a function of the laser intensity for the isolated atom (△) and for chains of N = 3 (●), 5 (×), and 7 (○) neon atoms.

for different laser intensities are compared in Fig. 5, for a single atom and for rows from three to seven atoms. It is expected that averaged ionization yields should grow steadily with the laser intensity. Within the framework of the above model, it appears, however, that averaged ionization yields grow much faster when the number of atoms in the chain is increased. This confirms the qualitative discussion in Sec. II, based on the intensity dependence of the effective potential $V_{J,\mathrm{eff}}(x_j,t)$ experienced by an atomic electron. These variations account also for the fact that larger clusters are not so effective for higher-order harmonic generation.

Within the framework of our model, besides the value of the parameter a^2 which determines the ionization energies of individual ''atoms'' in the row, see Eq. (1), there are two other key quantities governing the dynamics of the ''cluster.'' One is the ''interatomic distance'' d and the other is the b^2 parameter entering the expression of the electron-electron repulsive potentials, Eqs. (3)–(5). In order to ascertain our conclusions, it is thus of interest to discuss to which extent our results depend on the chosen numerical values for these parameters.

We have first investigated the influence of the choice of the value of b. The function of the b^2 parameter is to ''regularize'' the Coulomb singularity which plagues 1D computations involving charged particles. At this point, it is worth mentioning that there is no absolute criterion on how to choose an optimal value for b. This is essentially because in a 1D model, it is impossible to reproduce realistically the spectrum of an actual three-dimensional (3D) atom, even in the simpler case of a two-electron system. We note that smaller values of b make more singular the repulsive terms entering the expression of the effective potential modeling the electron correlations, Eqs. (3)–(5). On the contrary, larger values of b tend to smooth the electron-electron interaction. The implications are twofold: first, choosing smaller values of b will increase the anharmonicity experienced by an atomic electron when being driven by the field. This is expected to increase the harmonic yields. However, another closely related consequence is to also increase the ionization probability. These two competing tendencies are confirmed

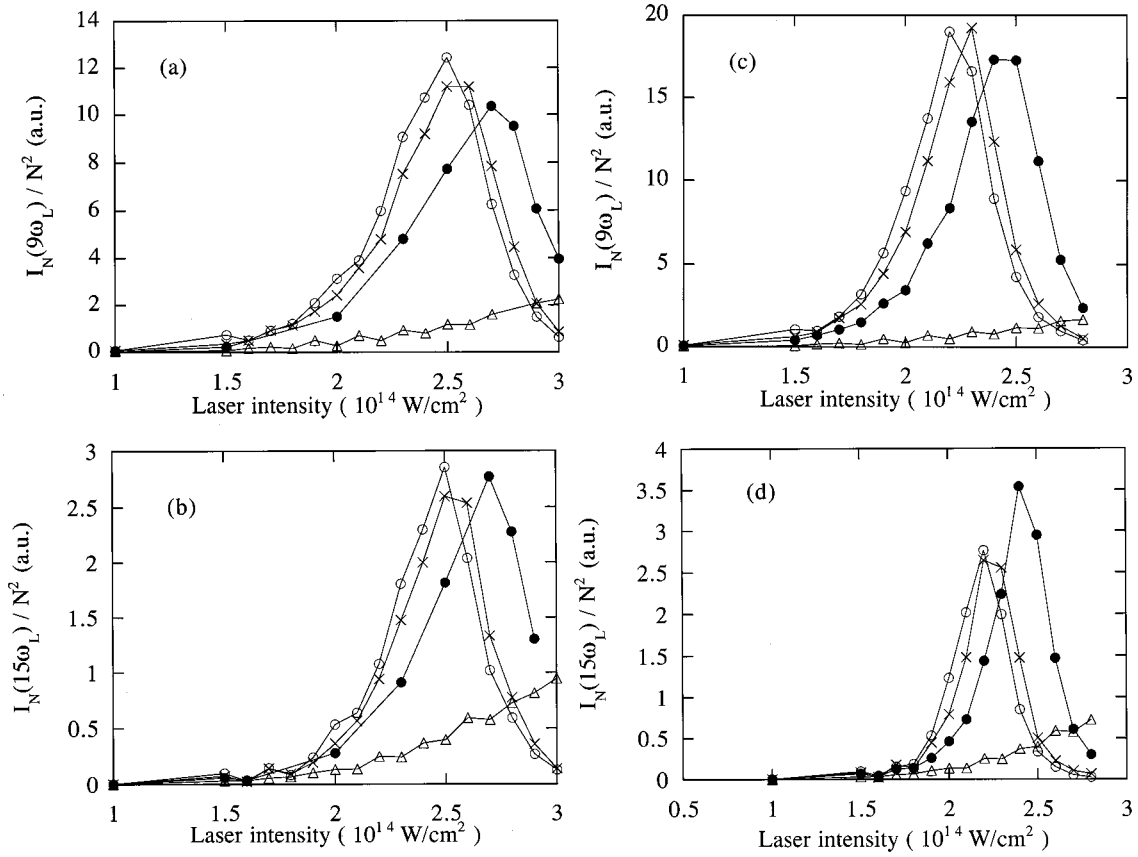

FIG. 6. Scaled harmonic yield $I_N(q\omega_L)/N^2$ as a function of the laser intensity for the isolated atom (△) and for chains of $N=3$ (●), 5 (×), and 7 (○) neon atoms. (a) H9 for $b=1.414$; (b) H15 for $b=1.414$. (c) H9 for $b=1$; (d) H15 for $b=1$.

in Figs. 6(a)–6(d), where the dependence of the emission yields in terms of the field intensity and of the chain length are shown for the harmonics $H9$ and $H15$, for two different values of b. We have chosen $b=\sqrt{2}$ (as above) and $b=1$. By comparing Figs 6(a)–6(c) and 6(b)–6(d), respectively, one observes first that to the smaller value of b correspond larger emission yields. This is particularly noticeable for $H9$ (note the change of scale between $H9$ and $H15$). One notes also that, again, when the intensity increases, the contributions of the larger clusters drop faster, the tendency being amplified when b is smaller. It is very likely that this is linked to the increase of the ionization probability which becomes higher when the repulsive interelectronic potential is more singular. The most important point, however, is that the general conclusions regarding the harmonic emission yields are globally unchanged when changing b. The main modifications affect principally the intensity ranges in which a cluster is more efficient than an isolated atom. In short, the intensity domain in which clusters are more efficient that isolated atoms, is reduced for smaller values of b. We turn now to a brief discussion of the role of the interatomic distance d, which has been shown to be of crucial importance when modeling the response of molecular systems to intense radiation pulses, see [12].

In the above analysis, we had chosen values for both the a parameter, see Eq. (1), and the ''interatomic distance'' d, which were supposed to approximately represent a row of ''neon'' atoms (namely $a=0.81$ a.u. and $d=6$ a.u.). The question is to determine if whether or not our general conclusions would be affected for another set of values, i.e., for other types of clusters. As an example, we have considered the case of a row of ''argon'' atoms with ionization potential $I_p=0.58$ a.u. ($a=1.175$) and ''interatomic distance'': $d_{Ar}=7$ a.u.

Before we discuss the case of clusters, let us review the main features of the single-atom response. We note first that, given the smaller values of the ionization energy and of the energy-level spacings, argon atoms have a significantly larger dynamic polarizability than neon for infrared frequencies. A direct consequence is that sizable harmonic emission can take place in argon, at lower field intensities than in neon. Moreover, at a given laser intensity, the emission yields are significantly higher from argon than from neon for relatively low harmonics ($H5-H9$). This can also be ascribed to the larger polarizability of argon. However, for higher harmonics the situation is reversed and emission yields are larger in neon. This comes from the fact that it can withstand higher intensities without being ionized.

These differences in the single-atom response strongly influence the dynamics of harmonic emission from rows of such atoms. This is illustrated in Fig 7, where we have compared the emission yields from single atoms with those from a row of seven neon and argon atoms. First, a general remark is that harmonic emission in argon clusters takes place at intensities below $I_L=1\times10^{14}$ W/cm^2, while in neon one has to go beyond $I_L=1.5\times10^{14}$ W/cm^2 to get comparable yields. This is specially striking for the harmonics $H5$ and $H9$, see Figs. 7(a) and 7(b). It appears also that the range of field intensity in which argon clusters are more efficient than single atoms is much reduced as compared to the case of neon. This results certainly from the fact that Ar atoms are more easily ionized, harmonic yields drop faster when the field intensity increases. As expected, this tendency is amplified in a row of atoms and the emission yields drop dramatically, after $I_L=7\times10^{13}$ W/cm^2 for $N=7$ atoms, see Figs. 7(a) and 7(b).

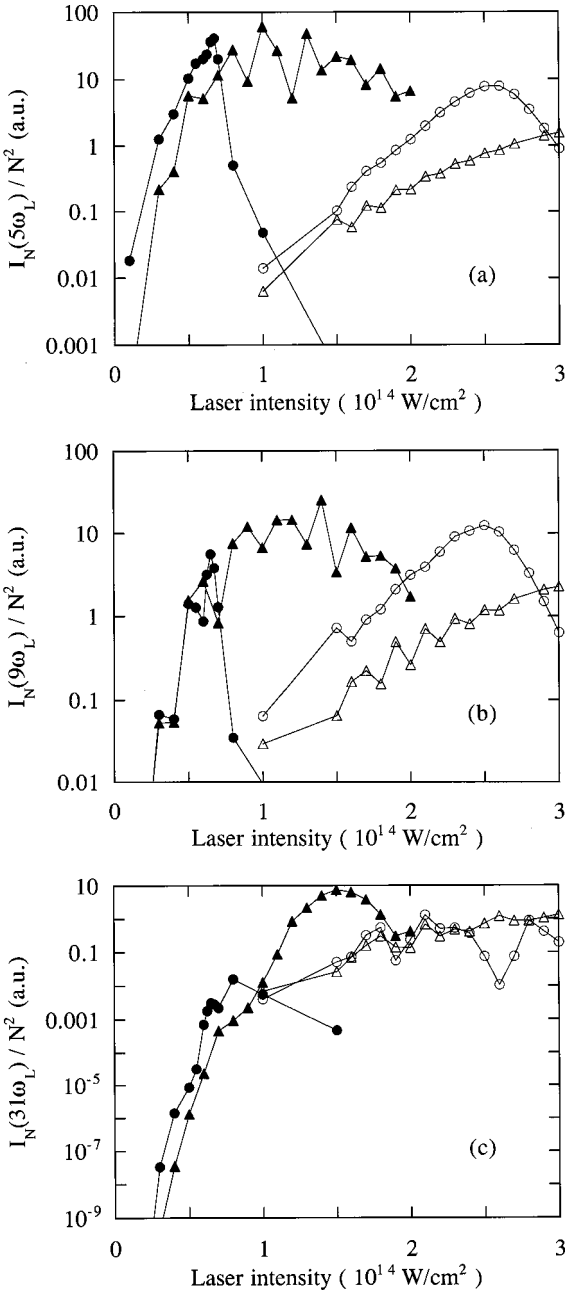

FIG. 7. Scaled harmonic yield $I_N(q\omega_L)/N^2$ as a function of the laser intensity for the isolated atom ($\triangle$) and for chains of $N=7$ atoms ($\bigcirc$). Open symbols correspond to the case of a neon atom ($a_{Ne}=0.81$ and $d_{Ne}=6$ a.u.) while filled symbols correspond to the case of a argon atom ($a_{Ar}=1.175$ and $d_{Ar}=7$ a.u.) (a) H5; (b) H9; (c) H31.

Another interesting point is that argon clusters can be almost ten times more efficient than those from neon for the generation of $H5$, see Fig. 7(a). This happens in spite of the fact that the maximum efficiency is reached at a much lower intensity in argon (here $I_L=7\times10^{13}$ W/cm^2), than in neon where it is when $I_L\approx2.5\times10^{14}$ W/cm^2. For $H9$, argon and

neon clusters provide comparable yields, although in different intensity ranges.

The situation is very different for higher harmonics. In Fig. 7(c), we have reported the dependence of the emission yields for $H31$ in terms of the field intensity from single atoms as well as from rows of neon and argon. There, neon is globally advantaged, clusters not being significantly more efficient than single atoms. The main reason is that, as already noted, neon can withstand higher intensities without being ionized. Further, in the comparatively rather low intensity range where argon's response is optimum, $H31$ is still in the cutoff domain, with relative emission yields much lower than for harmonics in the plateau.

IV. CONCLUSIONS

In the present paper we have presented a simplified theoretical model designed to address the problem of the response of a rare-gas cluster to an intense laser pulse. More specifically, we have discussed the question of high-order harmonic generation by such systems. The cluster has been modeled as a row of atoms aligned along the laser polarization direction. The nuclei are equally spaced at fixed positions and the one-electron atoms are modeled with the help of one-dimensional ''soft-Coulomb'' potential. We stress that, as a consequence of the limitations of the model, our discussion is only valid for the early stages of the interaction of a laser pulse with a real rare-gas cluster, i.e., for times short enough so that the motion of the nuclei can be disregarded and while multiple ionization is still negligible.

With these restrictions in mind, after solving the TDSE for such rows of $N=3$, 5, and 7 atoms in the presence of a laser pulse, we have simulated the harmonic spectra generated by small van der Waals clusters. The dynamics of the response of a chain of atoms depend significantly on a few key parameters characterizing the ''cluster,'' the most important ones being the ionization potential of the ''atoms,'' the interatomic distance and the modeling of the repulsive interaction between electrons from different atoms within the cluster. In order to disentangle the respective influences of these parameters, we have performed a set of numerical simulations for a fixed laser frequency ($\omega_L=0.057$ a.u. ≈ 1.55 eV, Ti:sapphire laser), at representative field intensities comprised between $I_L=1\times10^{13}$ W/cm^2 and $I_L=3\times10^{14}$ W/cm^2. Then, in order to address the question of the relative efficiency of clusters as compared to samples of isolated atoms with the same average density, we have compared the harmonic yields to those derived from the single-atom response.

The main result of our analysis is to confirm that clusters can be a medium more efficient for harmonic generation than a sample of isolated atoms. Enhancement factors of up to ten in the emission yields are not uncommon, for not too high harmonics ($H9-H13$) and at relatively moderate field intensities. However, our simulations do not permit us to conclude regarding the N^3 scaling law in terms of the number N of atoms in the cluster, which was proposed some time ago in order to interpret the experimental results [2]. Our simulations show also that the advantage of clusters does not extend to higher harmonics. On the contrary, scaled emission yields are *lower* than those from isolated atoms for harmonics close to the end of the plateau and, *a fortiori*, beyond the cutoff.

Another general result is that the intensity range in which harmonic yields are significant is much narrower in clusters than in isolated atoms. At lower intensities, the scaled response of a cluster does not differ from the one from a single atom. This results from the fact that the atoms are too loosely bound to enable a collective response to enable build up. On the contrary, at higher intensities, the harmonic yields from a cluster drop considerably faster than those from a single atom. The drop is correlated with the growth of the ionization yields in clusters. The lowering of the barrier against ionization in clusters is clearly evidenced when considering the time-dependent effective potential experienced by atomic electrons, see Figs. 1–3. This accounts for the fact that clusters are less efficient for higher harmonics than isolated atoms which can withstand higher intensities without being ionized. Another related finding is that clusters made from atoms with smaller ionization potential (and larger dynamical polarizability) generate harmonics in a narrower window located at lower laser intensity than those made from less polarizable atoms.

To conclude, although no quantitative data can be extracted from our simulations, they indicate that clusters are more efficient than isolated atoms for harmonic generation in restricted ranges of laser intensities. This is linked to the fact that the variations of the emission yield for a given harmonic in terms of the field intensity always follow a typical bellshape pattern. The main difference between clusters and single-atom responses is that the distribution for clusters is much narrower and is shifted towards lower intensities. However, it should be noted that isolated atoms can provide comparable or even larger yields when submitted to higher field intensities. This is particularly true for higher harmonics.

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