High Harmonic Generation by anions and atoms: Effect of initial/final state wave functions

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Abstract.

When High Harmonic Generation (HHG) is considered theoretically within the ideas of the rescattering mechanism, the specifics of initial/final state of the active electron usually attracts little attention. We explore such a specifics related to initial/final bound p-state of the active electron (existing studies concentrated on s-states). The halogen anions and noble gas atoms (except He) provide important examples of such targets. The present theory with realistic wave functions gives HHG rates by one or two orders of magnitude lower than previous calculations with asymptotic (single-exponential) wave functions. This indicates that the HHG process is much more sensitive to the description of the bound state wave functions in the inner domain than the Above Threshold Ionization where the asymptotic (large-r) approach is sufficient and self-consistent. The rates of HHG processes with and without change of electron magnetic quantum number m_ℓ are compared for B(2p) and H(2p) targets.

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1. Introduction

When High Harmonic Generation (HHG) by atoms and ions in a laser field is considered theoretically, the specifics of initial state of an active electron usually attracts little attention. In most cases an s-state was considered and simple exponential expressions were employed for the wave function, see references below. The specifics of states with higher orbital momentum were discussed in the recent paper by Ostrovsky and Greenwood [1]. The present work continues that study.

The important specifics of harmonic generation by atoms or ions with an active electron in higher-ℓ states ($\ell \geq 1$) is that Degenerate Combinational Harmonic Generation (DCHG) is possible. In such a process initial and final electron states are substates differing only in magnetic quantum number m_ℓ. In case of a filled shell (like $n\mathrm{p}^6$ shells in inert gases) such processes are forbidden by Pauli Exclusion Principle, but for other species with non-closed shells they are operative. These reservations apply for any harmonic generation process (i.e. harmonic order N is not necessarily high).

Some additional features appear when generation of high harmonics is under investigation. The appropriate physical framework in this case is provided within the ideas of the rescattering (or three-step) mechanism. It is worthy to remind here that the idea that HHG by an atom in a laser field (as well as some other processes, such as the double ionization) proceeds via a rescattering mechanism was initially put forward by Kuchiev [2, 3], Corkum [4] and Krause *et al.* [5, 6]. This idea was implemented quantitatively via different schemes (see, for instance, references [7, 8, 9, 10, 11]; and [12, 13] for reviews). The three steps involved are the above threshold ionization (ATI) of an atom‡, the propagation of electron in a laser field and the laser assisted recombination (LAR) that is accompanied by an additional exchange by energy with the laser field. In order to ensure that an active electron in the laser-dressed continuum returns to the residual core in course of its wiggling motion in linearly polarized laser wave, the electron has to be emitted along the direction of the field electric vector. Clearly, such emission is efficient for substates with magnetic quantum number $m_\ell = 0$ (with the quantization axis directed along the field vector), but quite non-efficient for $m_\ell = \pm 1$, since the related wave functions turn zero along the quantization axis§. This amounts to an *effective selection rule for HHG*. On the last step (LAR) the active electron might end up in a substate with an arbitrary value of m_ℓ, since no strict selection rules apply for laser-dressed states. The situation when final m_ℓ differs from initial $m_\ell = 0$ mean DCHG processes. Note that in the case of m_ℓ-changing transition the emitted light has different polarization characteristics.

The specifics discussed above is related to the *angular* wave function of the active

‡ Below we often refer to ATI in a broad sense, including above threshold detachment (ATD) processes for negative ions.

§ It is worthwhile to note that due to the same general reason the ATI rates are strongly suppressed with increase of the initial-states quantum number m_ℓ, see, for instance, review [14] and recent calculations [15, 16].

electron. Another issue is the choice of its *radial* wave function and the gauge for the electromagnetic field. The problems of this type pertain all the theory of interaction between the matter and the electromagnetic field (except the special 'exactly solvable' case of the hydrogen atom). For the process of HHG they practically were not explored (an attempt in this direction was carried out by Kuchiev and Ostrovsky [10] for the case of an active s-electron).

Within the framework of the rescattering mechanism the problem of gauge and wave function choice is split in two parts: these for ATI and LAR. On the ATI stage the length gauge stresses large separations of the active electron from the core in the initial-state wave function. This allows one to use the large-r asymptotes of wave functions and also favors the single active electron approximation. The combination of the length gauge with the asymptotic wave functions makes the approach self consistent, physically justified and easy to apply. This features were stressed by Gribakin and Kuchiev [17, 18] ‖ (see also Refs. [20, 21, 22]). The results for ATD obtained in this way agree well with the recent experiments [23, 24, 25] in the fairly demanding test of angle-differential rates for individual ATD channels. The results are also in agreement with the exactly solvable zero-range potential model [26, 27]. Recently Kjeldsen and Madsen [28] extended and supported this approach by calculating ATI on molecules.

For the last (LAR) step in the HHG process the situation is much less transparent. The necessary matrix element is contributed by entire range of electron-core separation r. This circumstance was noticed for LAR process in Ref. [29] and for HHG in Ref. [9]. Moreover, it becomes particularly important in case of HHG with an active p-electron where for DCHG the matrix element might diverge if an asymptotic wave functions is substituted [1]. In these circumstances there seems to be no other choice than to try to improve quality of the wave function in entire space.

The objective of the present work is to study the influence of the choice of the bound state wave function on the calculated HHG rates. To the best of our knowledge the investigations of this issue were not carried out before. The targets considered are the halogen anions and noble gas atoms (Section 2). The principle conclusion is that the matter is not about some small corrections: the use of non-asymptotic realistic wave functions generally reduces HHG rates by one or even two orders of magnitude, although reverse situation also might occur. We also present the comparative study of DCHG and conventional HG processes, correcting our previous results for excited hydrogen H(2p) atom and providing results for B(2p) atom with a single p-electron above a filled shell (Section 3). Section 4 contains some concluding remarks.

‖ Gribakin and Kuchiev provided a valuable modification of Keldysh [19] approach to ATI; for the recent review of Keldysh theory see paper by Popov [14].

2. HHG rates and choice of bound state wave functions

2.1. Theory

The present calculations of HHG are carried out within the general theoretical framework developed earlier [9, 10] and subsequently adjusted [1] to calculations for the case of an active p electron. The only difference is in the choice of the bound state wave function. According to discussion in the Introduction we retain the physically justified asymptotic (large-r) wave functions for the ATI step and use for the LAR step the realistic wave functions that provide a good approximation for the entire range of r. This is conveniently done within the approach of Ref. [9, 10] where ATI and LAR steps are explicitely singled out in the formulae for the HHG amplitude.

The realistic bound state wave functions for an active electron were used in the form of superpositions

$$\phi_{n_i\ell m_\ell}(\vec{r}) = \sum_i C_i \chi_{n_i\ell m_\ell}(\vec{r}) \ , \tag{1}$$

$$\chi_{n_i\ell m_\ell}(\vec{r}) = R_{n_i,\ell}(r)\, Y_{\ell m_\ell}(\hat{\vec{r}}) \ , \qquad \hat{\vec{r}} = \vec{r}/r \ , \tag{2}$$

$$R_{n_i,\ell}(r) \ = N_i r^{n_i-1} \exp(-\zeta_i r) \ , \tag{3}$$

where the parameters ζ_i, C_i and N_i are tabulated [31, 32, 30]. The number of terms in the sum (1) varied from 4 to 5.

The asymptotic wave functions have form

$$\phi_{n_i\ell m_\ell}(\vec{r}) \approx A_a r^{\nu-1} \exp(-\kappa r)\, Y_{\ell m_\ell}(\hat{\vec{r}}) \ , \tag{4}$$

where $\nu = Z/\kappa$ and κ is related to the binding energy of the active electron: $E_b = \frac{1}{2}\kappa^2$. The asymptotic parameters A_a are listed in the book [30].

2.2. HHG by halogen anions

Our calculations are carried out for the laser wavelength $\lambda = 800$nm, corresponding to a photon energy of $\omega = 0.057$a.u. The m_ℓ-changing transitions are not considered for halogen anions and noble gas atoms since they are forbidden by the Pauli Exclusion Principle as indicated in Introduction. Relatively weak laser intensities I were chosen in order to show formation of the plateau as I increases. Fig. 1 displays results of calculations for the lightest (F^-, circles) and the heaviest (I^-, triangles) halogen anions. The open symbols show our present results while the closed symbols refer to the previous calculations [1] where the asymptotic wave functions were used both for LAR and ATI steps of HHG. The general trend is that the present results are between one and two orders of magnitude lower. The reason for this is clear from the fact that the asymptotic approximation (4) is similar to retaining a single term in the sum (1), with the appropriate parameters chosen to match the large-r behavior¶. It does not reproduce wave function oscillations in the inner (small-r) region and thus generally overestimates

¶ Note, however, that the preexponential factor $r^{\nu-1}$ in (4) has non-integer power of r related to the energy parameter κ, while in formula (1) all n_i are assumed to be integer.

the electron density in this domain. The inner domain does not essentially contribute to the ATD amplitudes but it is probed on the LAR step of HHG processes.

In the plateau domain the HHG rates are known to oscillate with harmonic order N (see, for instance, Ref. [7, 8, 9, 10, 11, 33]). The positions of maxima and minima generally depend on the bound-state wave function employed, so that the relation between the previous and present refined results for rates is reversed for some N.

2.3. HHG by noble gas atoms

Fig. 2 shows the results of calculations for the lightest (Ne, circles) and the heaviest (Xe, triangles) noble gas atoms with outer p electrons. The same frequency of laser field is used as above. Again, the open symbols show the present results while the closed symbols refer to the results of previous calculations [1]. The refined rates tend to be lower, although the difference is less pronounced than in the case of halogen anions. The reversed relation occurs in the plateau domain for some N while beyond the plateau the new and old results tend to merge.

For noble gas atoms with large ionization potentials the characteristic number of photons involved in the processes is large that favours applicability of the present theory [10]. The calculated HHG rates apparently become more sensitive for the wave function details when the number of photons is smaller, as for anions.

3. Degenerate Combinational HHG

In this section we compare rates for the conventional (m_ℓ-conserving) HHG process and the m_ℓ changing (DCHG) process taking B($1s^2 2s^2 2p$) atom as a target. The choice is explained by the fact that this is the simplest atom with a single p-electron above closed shell in the ground state; this is the first comparative study of the two processes for a ground state atom. As above, we use the realistic wave functions for the LAR step of HHG and the asymptotic functions for the initial ATI step (the asymptotic parameters of the wave functions are $\kappa_{\rm B} = 0.781$, $A_a = 0.88$ [30]). Our interest to such calculations stems from the fact that there is no obvious general grounds to conclude which HHG process is more efficient, so that the direct calculation seems to be the only way to resolve the issue. Fig. 3 shows that for the conventional process the rate is higher almost by an order of magnitude. This situation is reversed for some values of harmonic order N within the oscillatory patterns in the plateau domain.

In Fig. 4 the similar results for the excited hydrogen atom H(2p) are presented (they replace our previous results [1] that were found to be in error). In this case the difference between the conventional HG process and DCHG is much more significant than that for B atom. The main distinction seems to be that in the case of B atom an active electron has larger binding energy, $\frac{1}{2}\kappa_{\rm B}^2 = 0.305$ a.u., compared to 0.125 a.u. for H(2p). Note that in the both cases an active electron has nodeless radial wave function, although the details of behavior in the small-r domain are, of course, different. For

the linearly polarized laser field it would be probably more appropriate to consider an excited hydrogen atom in various parabolic states. However, we limit the present study to the spherical states since the main interest is to compare H(2p) and B(2p) atoms with an active electron in the states of the similar type.

4. Conclusion

In the present paper the HHG process is considered in the aspect of an initial/final wave functions choice. Both radial and angular parts of wavefunction are substantial. We show that the refined choice of the radial wave functions leads to decrease of the calculated HHG rates by one or two orders of magnitude, albeit the trend might be reversed in the plateau domain where the rates depend on the harmonic order in an oscillatory fashion. The reason for this is that the HHG process generally probes the inner domain of configurational space for the final electron state. For the same laser frequency ω the sensitivity of HHG rates to the final state wave function is higher for the halogen anions than for the noble gas atoms.

The non-spherical angular part of wave function ($\ell \neq 0$) implies that the magnetic quantum number m_ℓ might change leading to emission of energetic photons with the same energy as in the conventional HHG. The m_ℓ-changing processes generally have lower rates than conventional HHG, although the difference might be less than order of magnitude. Again, the relation might be reversed in the plateau domain. The m_ℓ-changing transitions are of interest since the emitted harmonic differs in polarization from that of the original laser wave; the harmonics produced by the conventional and DCHG processes are not coherent, although have the same frequency.

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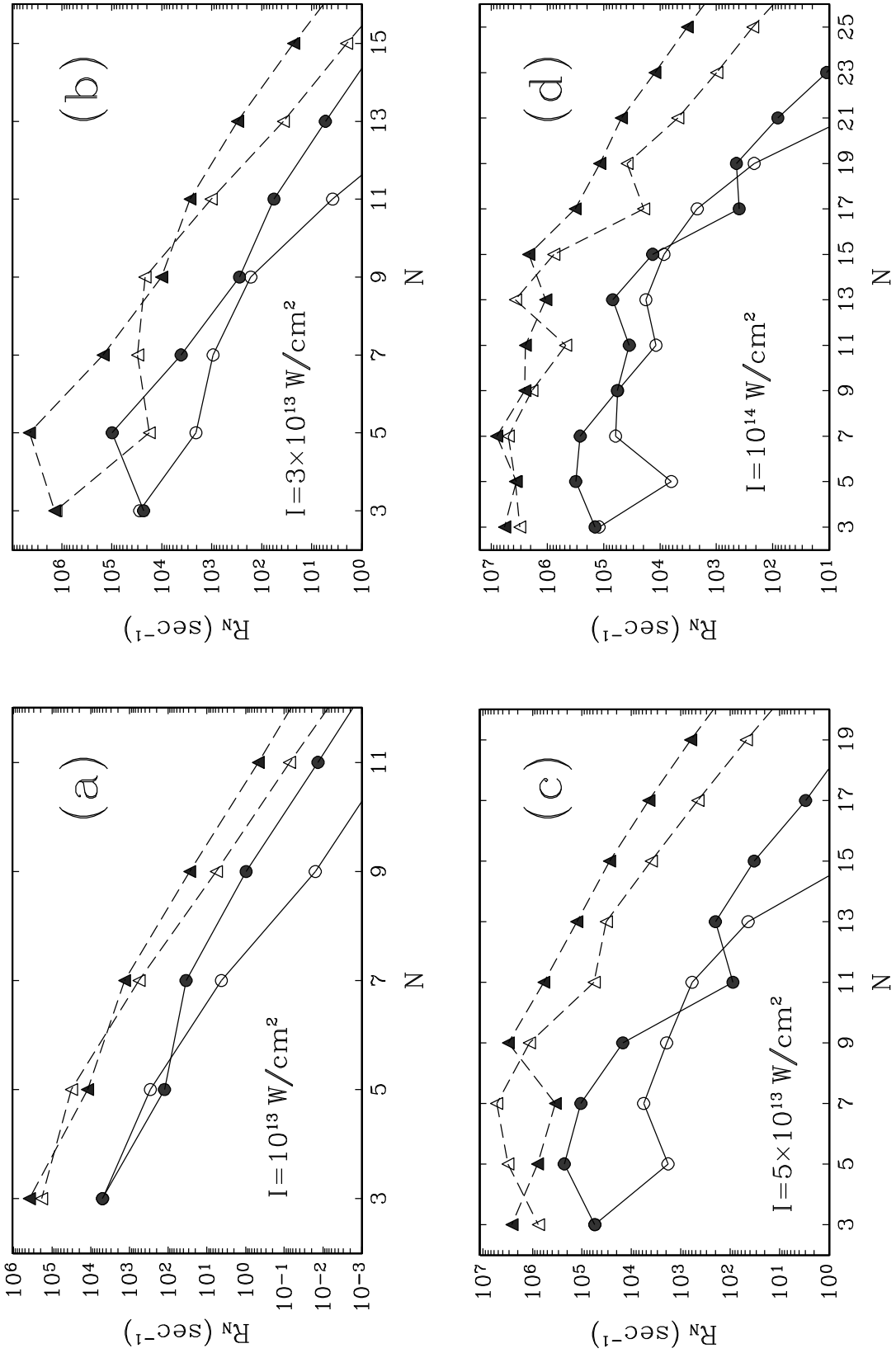
Fig. 1



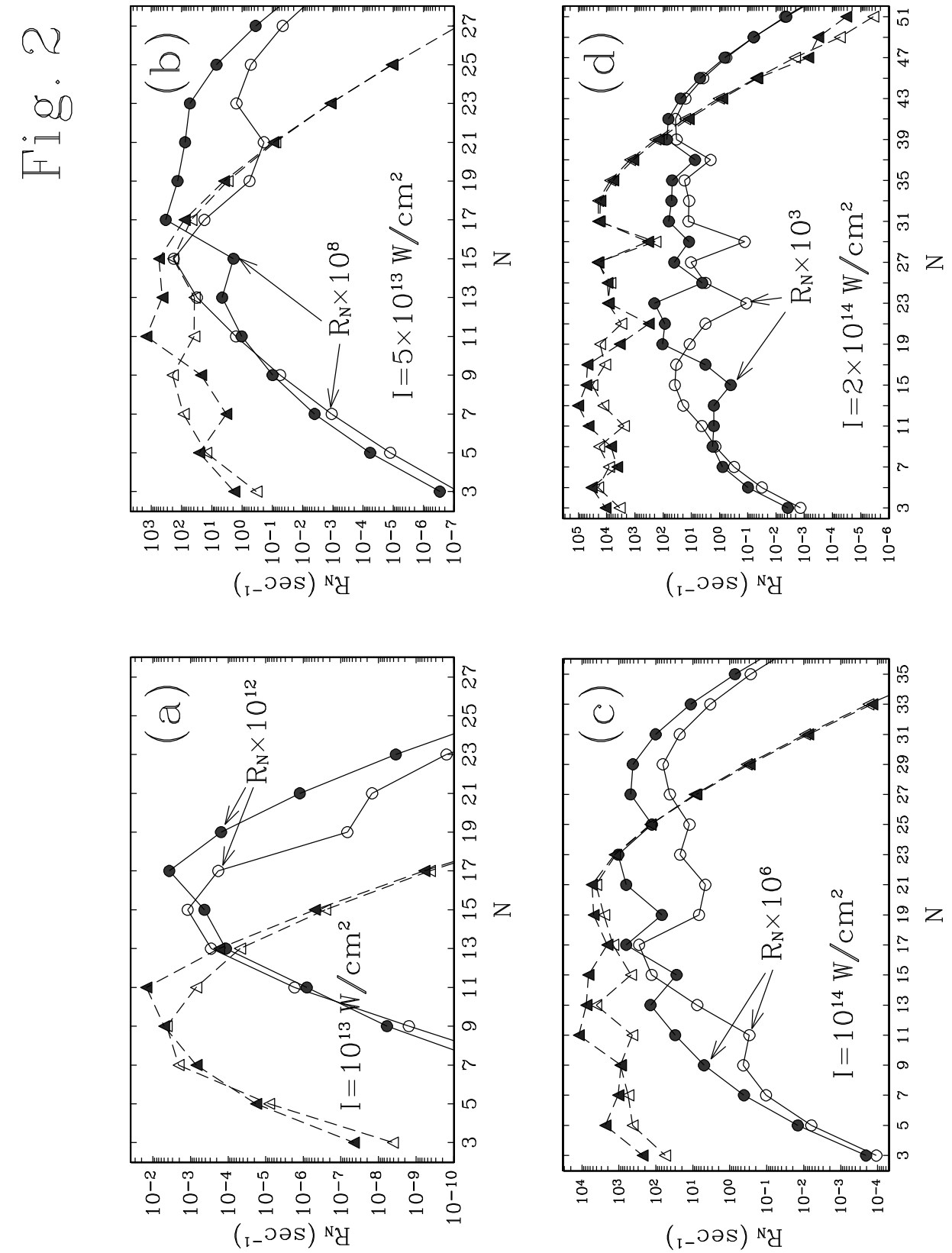

(a)
$I = 10^{13}$ W/cm^2
$R_N \times 10^{12}$
(b)
$I = 5 \times 10^{13}$ W/cm^2
$R_N \times 10^{8}$
(c)
$I = 10^{14}$ W/cm^2
$R_N \times 10^{6}$
(d)
$I = 2 \times 10^{14}$ W/cm^2
$R_N \times 10^{3}$
R_N (sec^{-1})
N

Fig. 2

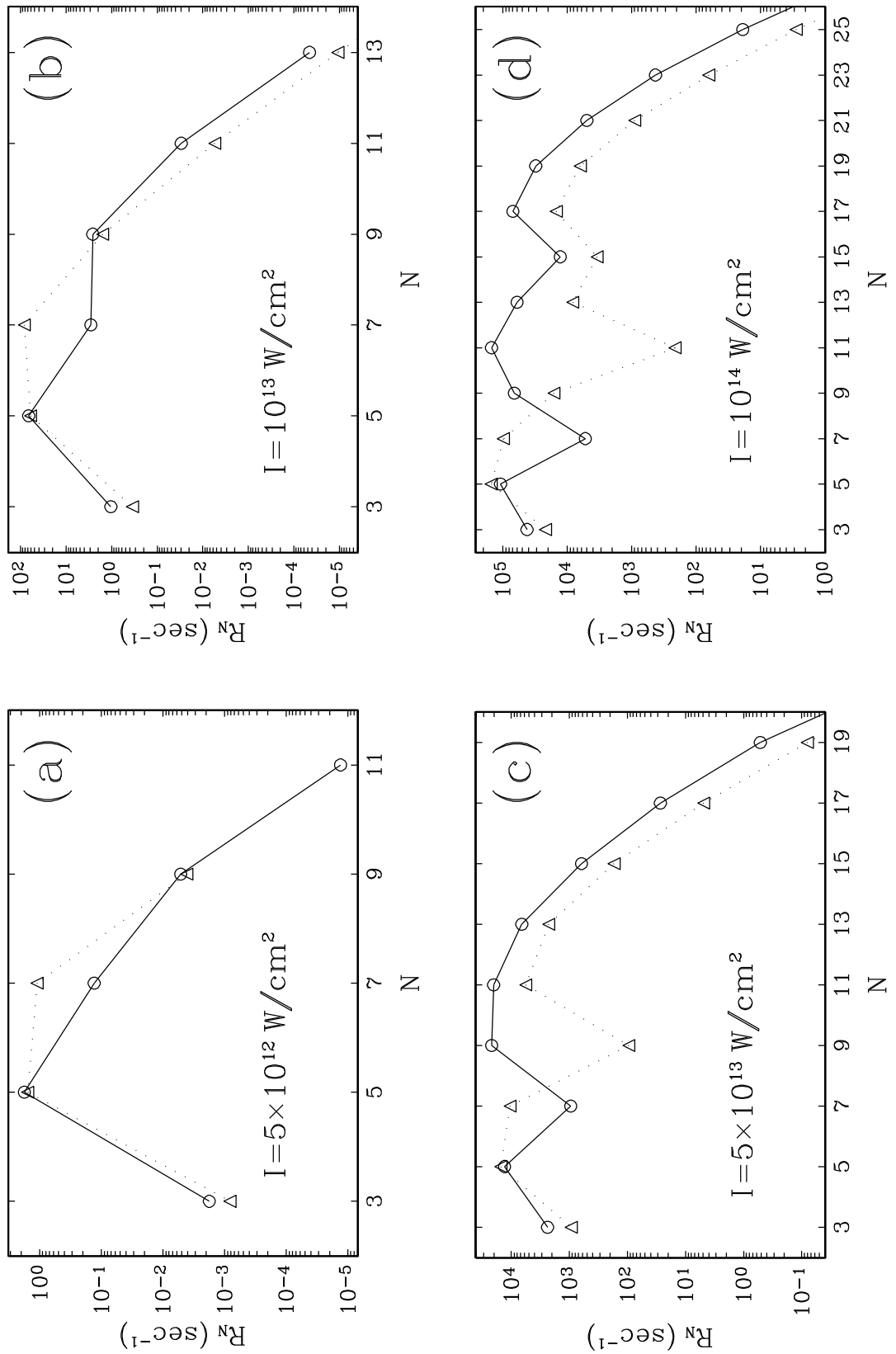


Fig. 3

Fig. 4

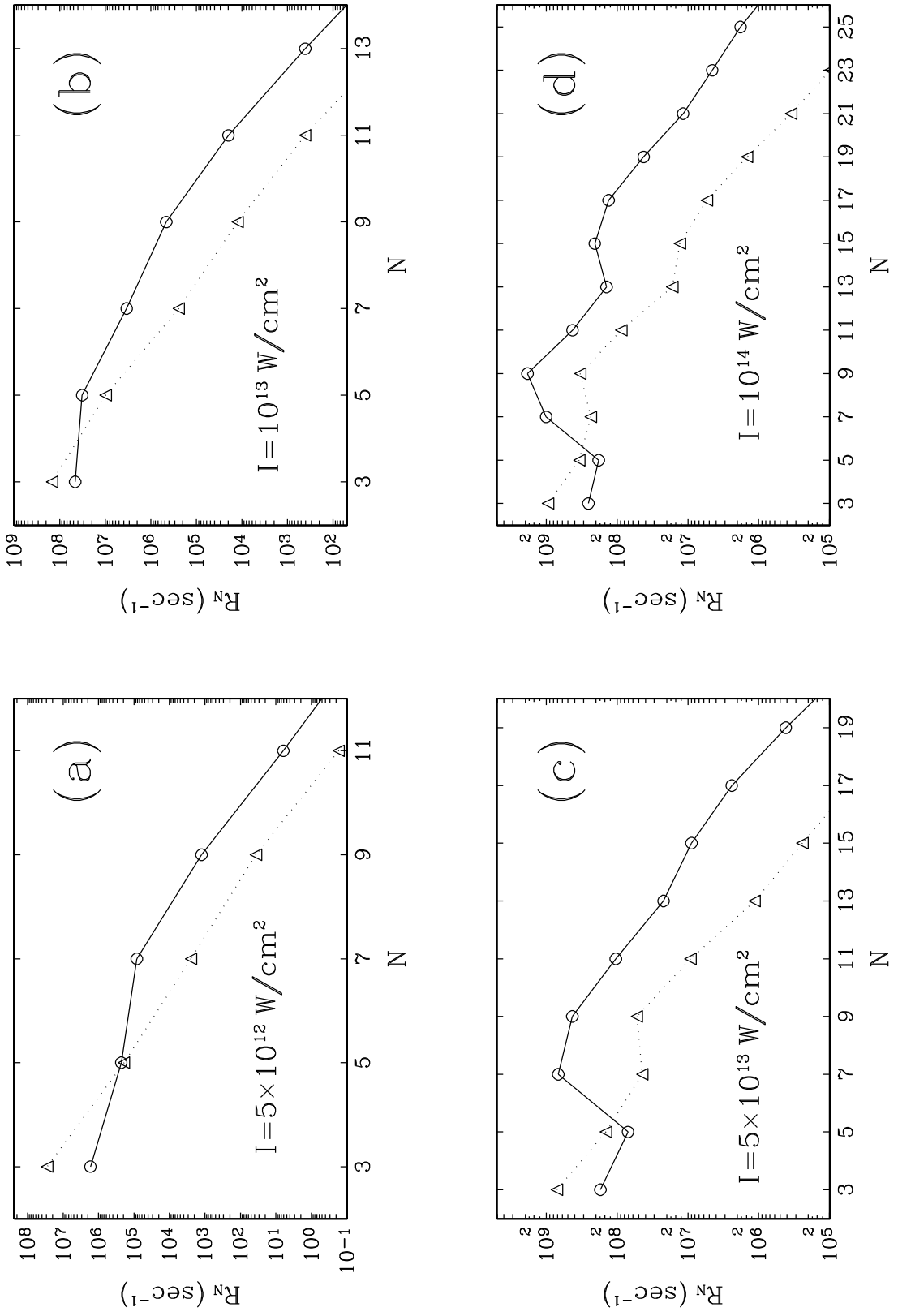


Figure 1. Rates for HG by halogen anions depending on the harmonic order N for various targets: circles – F^-; triangles – I^-; open symbols – the present calculations with realistic wave functions; closed symbols – calculations with asymptotic wave functions [1]. The symbols are joined by lines to help the eye. The laser field intensities I are shown in the plots.

Figure 2. Rates for HG by noble gas atoms with an active p-electrons depending on the harmonic order N for various targets: circles – Ne; triangles – Xe; open symbols – the present calculations with realistic wave functions; closed symbols – calculations with asymptotic wave functions [1]. The laser field intensities I are indicated in the plots.

Figure 3. Rates for HG by ground state B(2p) atom depending on the harmonic order N. The rates for conventional HHG process (circles) are compared with m_ℓ-changing (or DCHG) process rates (triangles) for various laser field intensities I indicated in the plot.

Figure 4. Same as in Fig. 3, but for excited hydrogen atom H(2p).