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FAST TRACK COMMUNICATION

Strongly enhanced high-harmonic generation via antisymmetric ionic states

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Abstract

In this work a mechanism to enhance the electron recollision dynamics after laser-induced ionization of antisymmetric states of atomic ions is pointed out. This is accomplished by combining the directed ionization characteristics of antisymmetric states with the laser-induced magnetic field drift. As an important application, we focus on the improved efficiency of high-harmonic generation.

(Some figures in this article are in colour only in the electronic version)

Since its discovery in 1987 [1] high-harmonic generation (HHG) has been in the focus of numerous both theoretical and experimental research activities in the field of laser–matter interaction [2–4]—until nowadays. These efforts are justified by the fact that HHG may serve as a source of coherent light, in which an up-converting of the irradiated laser radiation to shorter wavelength coherent light takes place. At present, in terms of experimental applications, high-harmonic radiation represents the basis for the generation of attosecond pulses. This renders the possibility of investigating atomic and molecular processes on the attosecond time scale by means of intriguing pump-probe experiments [5–7]. From the theoretical point of view the three-step model of HHG [8] has brought basic understanding to this phenomenon. With this, one can account for general important characteristics of HHG spectra, for instance, the occurrence of a maximum cut-off frequency. The first step of this model consists in the ejection of an electron out of a bound atomic state due to the interaction of an atom with a strong laser field which initiates a tunnelling process of the electron into the continuum by tilting the core potential. Subsequently, in the second step, the electron oscillates in a quasi-free motion along the polarization direction of a linearly polarized laser field. When the laser field changes its sign, the electron is accelerated back towards the mother ion. In the final third step, the actual recollision process takes place. In this process the recombination of the recolliding electron into the initial state occurs under the emission of high-harmonic radiation. This radiation contains multiples of the irradiated laser frequency—up to a cut-off frequency of $\omega_{\max} = I_p + 3.17U_p$, where I_p represents the ionization potential

of the specific atomic species and U_p denotes the ponderomotive potential of the laser field. Here, we would like to emphasize that in the wake of an efficient recollision dynamics not only HHG but also other alternative processes such as high-order above-threshold ionization (ATI) [9] and non-sequential double ionization [10] would be efficiently enhanced. In the latter, the recolliding electron promotes another bound electron into the continuum, whereas in the high-order ATI-process the recolliding electron is elastically scattered from the nucleus. Moreover, it is thinkable that nuclear processes may be initiated by the recollision process [11, 12].

In the case of the recombination process the term $3.17U_p$ describes the maximum kinetic energy of the recolliding electron which gives rise to the highest harmonic frequency. In order to achieve higher harmonic frequencies, it appears appropriate, at the first glance, to increase the intensity of the applied laser field (this leads to a larger U_p). The generation of extreme short-wavelength harmonic radiation is desirable, for the prospects that HHG represents a suitable light source for the investigation of bio-chemical processes with unprecedented resolution are promising [13, 14]. In view of the cut-off formula above it is meaningful to employ highly charged atomic ions for HHG so that the atomic ions possess a large I_p and are thus able to withstand very intense laser fields. A too rapid depletion of the initial state already during the turn-on phase of the laser field would result in the fact that the maximum cut-off frequency according to the formula above could not be achieved. One would also face a strong reduction of the harmonic signal since the corresponding matrix element would turn out to be small. Moreover, applying very intense laser fields is prone to the limitations caused by the drift in the laser propagation direction, which is induced by the laser magnetic field. The displacement of the recolliding electron in laser propagation direction reduces the efficiency of the recollision process. As a result the magnetic-field drift impedes not only HHG but also other processes such as high-order ATI, non-sequential double ionization and the possibility of employing the recollision dynamics for nuclear processes are affected. Therefore, in view of the fundamental role of the recollision dynamics for the processes mentioned above a solution of this problem would be of broad interest. The solutions up to now suggest to employ pre-accelerated particles [11, 15], to tailor the applied laser pulse [16] or to use crossed laser fields [12, 17]. A further concept is based on choosing particular systems in laser fields such as positronium [18, 19]. Very recently, another solution has been pointed out which consists in the application of specific properties of antisymmetric molecular orbitals in order to achieve, by means of the magnetically induced drift, an increase in recollisions and with this an enhancement of HHG [20]. The interplay of antisymmetry and the Lorentz drift leads to increased recollisions and an enhanced harmonic emission. As for HHG, the detrimental impact of the laser magnetic field can be removed by this measure, but the harmonic signal is still somewhat reduced due to the ionization of the fragile molecule.

In this fast track communication we demonstrate how the concept mentioned above can be successfully transferred to highly charged atomic ions. A too rapid depletion of the initial state is suppressed by the high binding energies of the ions. Therefore, in comparison with molecules, atomic ions exhibit a higher harmonic signal strength and an easier realization of the 3.17-formula. We investigate orbitals that are antisymmetric with respect to a nodal plane. In order to study the influence of the laser magnetic field we perform our simulations with and without employing the dipole approximation for the laser field. Our simulations consist in the direct numerical integration of the time-dependent Schrödinger equation in two dimensions. The antisymmetric atomic orbitals under consideration are the 2D-equivalents of real 2p-orbitals of hydrogen-like ions.

First we illustrate the method to exploit mirror antisymmetry of atomic wavefunctions in order to compensate for the drift induced by the laser magnetic field, so that an enhanced

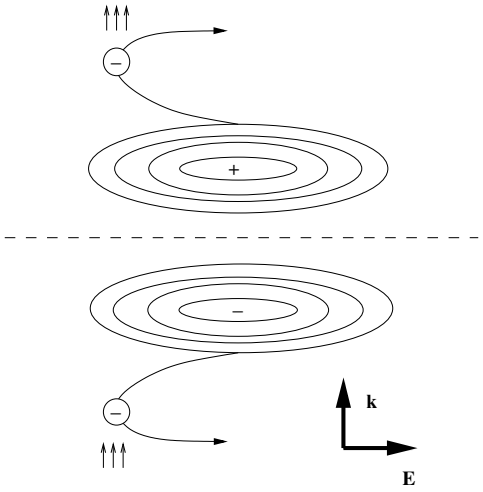

Figure 1. Schematic diagram of an antisymmetric atomic orbital subjected to a linearly polarized laser field with polarization vector **E** and propagation direction **k**. The dashed line symbolizes the nodal line. Within the dipole approximation, the ejected electrons cannot travel parallel to the nodal line. The magnetically induced electron drift helps to transfer the electron towards the nucleus, as indicated by the small arrows in the lower part of the figure.

harmonic emission can take place. We consider these antisymmetric wavefunctions subject to a short, strong linearly polarized laser pulse restricting ourselves to the plane spanned by the laser polarization direction and the laser propagation direction in our simulations. This is the plane in which the crucial physics occurs. Figure 1 depicts such an antisymmetric excited atomic state with a nodal line (dashed line). Considering this initial state as a reservoir of electrons, the electrons are ejected out of this state via tunnel ionization as a result of the interaction of this state with the laser field. The continuum electrons oscillate in a quasi-free manner along the laser polarization direction. In addition to this oscillation, there is a drift motion of the electrons in the laser propagation direction caused by the laser magnetic field. This drift is indicated by the small arrows in the figure. As a result, the electrons which are accelerated towards the nucleus after the laser field has changed its sign cannot efficiently recollide. Hence, an efficient recombination process under emission of harmonic radiation is suppressed.

Here we make use of the following crucial property of antisymmetric wavefunctions: the density of the corresponding momentum wavefunction which belongs to the antisymmetric initial wavefunction in coordinate space possesses also a nodal line which is parallel to the nodal line in coordinate space[3]. This means that the tunnelled electrons show an additional lateral drift perpendicular to the nodal line. There are no electrons ejected with momentum vectors strictly parallel to the nodal line. For the particular orientation of the wavefunction as in figure 1 with the nodal line parallel to the laser polarization direction, this drift is directed either along the laser propagation direction (upper part of the figure) or opposite to this direction (lower part of the figure). In the latter case the drift due to antisymmetry is employed to compensate for the drift induced by the laser magnetic field. In the upper part of

[3] The atomic antisymmetric 2D-eigenfunctions possess in coordinate space a quite similar shape to the 2D-eigenfunctions of H_2^+. Similarly, the atomic momentum-space wave functions also exhibit the symmetry properties of the molecular ones. For details see [21].

the figure, the electrons are steered even further away in the laser propagation direction. As a net effect, however, there is compensation of the magnetically induced drift resulting in an increased harmonic emission as our simulations show. The orientation chosen in the figure with the nodal line of the antisymmetric wavefunction perpendicular to the laser propagation direction represents an optimal combination of both drifts. If the nodal line is parallel to the laser propagation direction, the electrons are preferably ejected in the laser polarization direction due to antisymmetry. But in the laser polarization direction the electron dynamics is governed by the laser electric field. Hence, the drift in the laser propagation direction induced by the laser magnetic field remains uncompensated and it therefore hampers recollisions and HHG. Thus, the orientation of the antisymmetric atomic wavefunctions with regard to the laser polarization direction is vital for the success of our method.

Here, we would like to note that the preparation of the real 3D 2p-orbitals can be accomplished by a modestly intense preparation pulse of appropriate frequency and polarization according to the common dipole selection rules ($\Delta l = \pm 1, \Delta m = 0, \pm 1$), neglecting the fine structure. Starting from the groundstate of a hydrogen-like system with quantum numbers ($n = 1, l = 0, m = 0$) one can, for instance, excite the ($n = 2, l = 1$, $m = 0$)-state by means of light which is polarized along the chosen z-axis. The nodal plane of this state is perpendicular to this axis. By choosing the polarization direction of this preceding pulse it is possible to obtain a desired orientation of the wavefunction with regard to the polarization direction of the following strong laser pulse. In order to establish the 3D scenario corresponding to the one depicted in figure 1, one would have to irradiate the strong laser pulse perpendicular to the nodal plane of the ($n = 2, l = 1, m = 0$)-state.

In order to investigate our method in detail, we have solved the time-dependent Schrödinger equation numerically for the antisymmetric 2D-states subject to a short, strong linearly polarized laser pulse. The interaction between the laser field and the antisymmetric wavefunction is described by the following Hamiltonian:

$$H(x, y, t) = \frac{1}{2}\mathbf{p}^2 + \frac{1}{c}A_x(y, t)p_x + \frac{1}{2c^2}A_x(y, t)^2 + V_{\text{ion}}(x, y). \tag{1}$$

Here t denotes the time and c the speed of light. The x-axis denotes the laser polarization direction, whereas the y-axis represents the laser propagation axis. The operator $\mathbf{p} = (p_x, p_y, 0)$ is the canonical momentum. $A_x(y, t)$ is the non-vanishing component of the vector potential $\mathbf{A} = \big(A_x(y, t), 0, 0\big)$ describing the laser pulse. Retaining the spatial dependence of $A_x(y, t)$ means that the laser magnetic field is taken into account in the calculation. In the dipole approximation this dependence is neglected, which results in the neglect of the laser magnetic field. Both variants have been performed in order to study the influence of the laser magnetic field. The envelope of the sine laser pulse possesses a trapezoidal shape. The term $V_{\text{ion}}(x, y)$ describes the interaction of the nucleus of the hydrogen-like ion N^{6+} with the electron. It is modelled by a soft-core potential [22]:

$$V_{\text{ion}}(x, y) = -\frac{10.96}{\sqrt{1 + x^2 + y^2}}. \tag{2}$$

The soft-core parameter in the numerator of the term has been adapted so that the first excited state of this potential, which represents the antisymmetric state under consideration in this work, possesses an eigenenergy of -6.1 au, which corresponds to the value of the real (3D) first excited state of the hydrogen-like system N^{6+}. The initial wavefunction has been obtained by a spectral method [23]. By selecting an appropriate initial testing function, which is antisymmetric with regard to a nodal line of desired orientation to the laser polarization direction, it is possible to achieve a certain alignment of the atomic antisymmetric eigenfunction. In order to solve the time-dependent Schrödinger equation with the

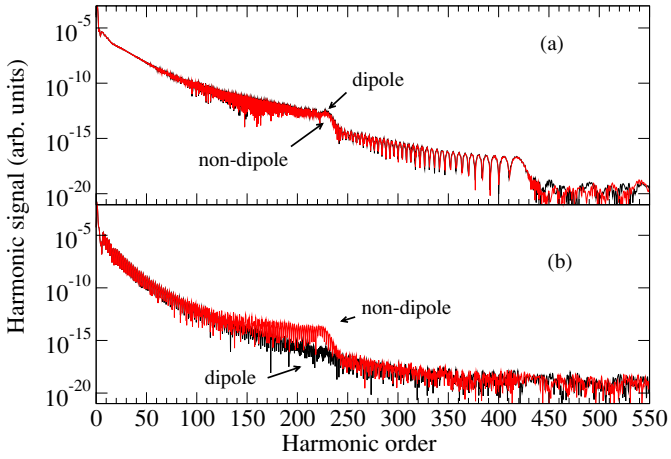

Figure 2. Harmonic spectra of the first excited antisymmetric state of N^{6+} subject to a linearly polarized sine laser pulse of 1.0×10^{17} W cm^{-2}. The trapezoidal shape of the pulse comprises one cycle linear turn-on, two cycles of constant intensity and one cycle linear turn-off. The results depicted in black have been obtained in the dipole approximation, whereas the red (grey) lines represent results beyond this approximation. In part (a) the nodal line of the initial state is perpendicular to the laser polarization direction. The drift due to antisymmetry does not counteract the magnetically induced drift giving rise to a reduction of the harmonic signal. In contrast to this, in part (b) the nodal line is parallel to the laser polarization direction. As a result, both drifts mutually counteract leading to a significant enhancement of harmonic emission.

Hamiltonian (1) we employ the established split-operator method [24]. The harmonic spectrum is computed as the modulus square of the Fourier transform of the dipole acceleration [25].

Figure 2 shows harmonic spectra of N^{6+} in the prepared antisymmetric excited state which interacts with a trapezoidal short laser pulse of 1.0×10^{17} W cm^{-2}. The laser wavelength is 248 nm. In part (a) the nodal line is orthogonal to the laser polarization direction, while in part (b) it is parallel to this direction. The black lines represent results obtained in the dipole approximation. The red ones (grey in print) depict results beyond this approximation. The figure confirms the proposed mechanism outlined above. In part (a) the drift induced by the laser magnetic field is directed in the laser propagation direction, whereas the drift due to antisymmetry of the orbital is parallel to the polarization direction. These two drifts do not mutually compensate. Therefore, the laser magnetic field drift accounts for the reduction of the harmonic signal. The electron in the antisymmetric state is still sufficiently bound ($I_p = 6.1$ au). No distinct depletion of this state takes place in the turn-on phase of the pulse in spite of the very high intensity. Thus, the cut-off predicted by the 3.17-formula of a maximum harmonic order of 380 could be realized. In part (b) of the figure both drifts mutually counteract, leading to increased recollisions and enhanced harmonic generation in the non-dipole case. Here, the enhancement of the harmonic signal is up to two orders of magnitude. In this case the drift due to antisymmetry prevails, which reduces recollisions over-compensating the magnetically induced drift. Thus, the maximum harmonic cut-off is not attained.

In the following, we point out the advantage of employing antisymmetric states of hydrogen-like atomic ions with large ionization potential (I_p) over the usage of molecules with naturally low I_p. Figure 3 compares harmonic spectra of N^{6+} and H_2^+; both subject to the same laser pulse as in figure 2. The results have been achieved without the dipole

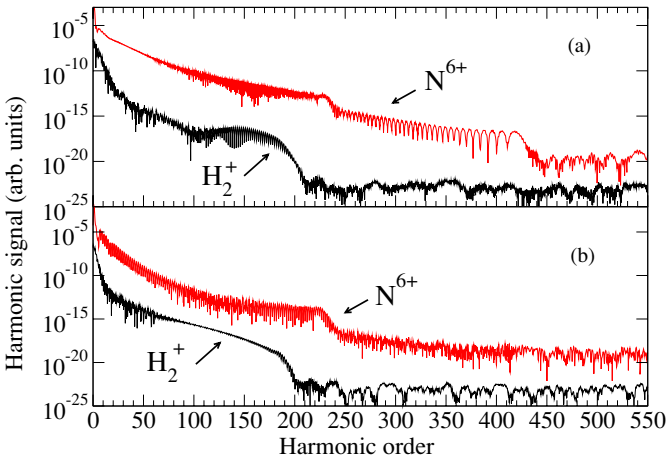

Figure 3. Comparison of harmonic spectra of N^{6+} with H_2^+. Both substances are found in the first excited antisymmetric state prior to the interaction with the same laser pulse as in figure 2. The red (grey) lines depict N^{6+}-spectra, while the black ones represent H_2^+-spectra. In part (a) the nodal line of the corresponding wavefunction is perpendicular to the laser polarization direction; in part (b) it is parallel to it. All spectra have been obtained without employing the dipole approximation. The figure demonstrates the enhanced harmonic emission in the case of the highly charged ion N^{6+} owing to the higher binding energy (see text).

approximation. As in figure 2, the first excited antisymmetric state with $I_p = 6.1$ au is taken into consideration for N^{6+}, while in the case of H_2^+ the first electronically excited state with $I_p = 0.67$ au has been investigated in the Born-Oppenheimer approximation. For details of the molecule calculation see [20]. In part (a) of the figure the nodal lines of both the H_2^+ state and the N^{6+} state are orthogonal to the laser polarization direction, whereas in part (b) the corresponding nodal lines are parallel to the laser polarization direction. The red lines (grey in print) represent the N^{6+}-spectra and the black ones illustrate the H_2^+-spectra.

Note that the N^{6+} and the H_2^+ results have been obtained with the same code, merely in the case of H_2^+ we have a double-well soft-core potential [20]. A thorough analysis of the H_2^+-computation yields that there is substantial ionization of H_2^+ already in the turn-on phase of the pulse [20]. Therefore, the cut-off of the 373th harmonic order according to the 3.17-formula is not attained. In contrast to the H_2^+-spectra, the N^{6+}-spectra exhibit a considerably enhanced harmonic signal by many orders of magnitude. This makes the investigation of antisymmetric states of highly charged ions particularly interesting in view of experiments. The enormous difference in the signal strengths can be attributed to the higher binding energies of the highly charged *atomic* ions, since they do not suffer from rapid depletion despite the very high laser intensity. This gives rise to the circumstance that the matrix element $\langle\phi_{\rm init}(t)| - \frac{\partial V_{\rm ion}}{\partial x}|\phi_{\rm rec}(t)\rangle$ ($|\phi_{\rm init}(t)\rangle$ denotes the initial wavefunction and $|\phi_{\rm rec}(t)\rangle$ represents the recolliding wave packet), which is decisive for the recombination process, turns out to be considerably larger, since the occupation of $|\phi_{\rm init}(t)\rangle$ at the time t of recollision is substantially higher for highly charged atomic ions. This is responsible for the enhanced harmonic emission observed.

In conclusion, we have outlined a concept for antisymmetric 2p atomic orbitals of highly charged ions, which combines the properties of antisymmetry with the drift induced by the laser magnetic field in order to achieve an enhancement of harmonic emission in spite of a very high laser intensity. In particular, we have pointed out the advantage of strongly bound

atomic ions over weakly bound molecular systems which consists in the fact that *atomic* ions yield a considerably more intense harmonic signal. This circumstance makes antisymmetric states of highly charged atomic ions particularly attractive for applications requiring efficient harmonic yields in the x-ray regime.

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