Interaction of Superintense Laser Pulses with Relativistic Ions

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At high intensities, three-step recollision processes driven by low frequency laser pulses, such as high-order harmonic generation and high-order above-threshold ionization, are normally severely suppressed by the magnetic-field component of the laser field. It is shown that this suppression is not severe, even for ponderomotive energies well above 10 keV, for multicharged ions moving at a sufficiently high relativistic velocity against a counterpropagating infrared laser pulse. Numerical results are presented for high-order harmonic emission by a single Ne^{9+} ion moving with a Lorentz factor $\gamma = 15$ against a Nd:glass laser beam. The calculations are done within a Coulomb-corrected nondipole strong field approximation. The approximation is tested by comparing with accurate results.

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This work is motivated by the development, in the near future, of a new accelerator complex at the GSI laboratory (Darmstadt, Germany) where relatively dense bunches of positive ions, in arbitrary charge states, will be accelerated to Lorentz factors up to about 25. It will be possible to irradiate these ions with ultraintense laser pulses produced by the PHELIX laser facility, currently developed on the same site, which has a working wavelength of 1053.7 nm and a peak intensity above 10^{21} W cm^{-2}. In what follows, we shall assume that the laser beam is oriented so that it counterpropagates with respect to the direction of motion of the beam of ions, which will be feasible at GSI. Because of the relativistic Doppler effect, the ions are then exposed in their frame of reference (moving with respect to the laboratory frame with a speed $v \approx c$) to a laser field of angular frequency $\omega_I = (1 + v/c)\gamma\omega_L$ and electric field amplitude $F_I = (1 + v/c)\gamma F_L$, where $\gamma = (1 - v^2/c^2)^{-1/2}$ is the Lorentz factor and ω_L and F_L are the angular frequency and electric field amplitude of the laser pulse in the laboratory frame.

An important question is the efficacy, in these conditions, of multiphoton processes [1] such as high-order above-threshold ionization, multiple ionization, and high-order harmonic generation. In this Letter, we examine the process of high-order harmonic generation which, at the single-ion level, is similar to the other two in arising from the same three-step mechanism [2,3]. In the first step, the active electron is detached from the core through tunneling ionization or "over the barrier" ionization. In the second step, it propagates in the continuum like a "quasifree" electron accelerated by the laser field. In the third step, the electron interacts with the core if it comes back to its vicinity. At this point, the electron may (i) be simply scattered, which leads to high-order above-threshold ionization, or (ii) knock out other atomic electrons, which leads to multiple ionization, or (iii) recombine radiatively, which leads to the emission of an energetic photon, corresponding to high-order harmonic generation. It has been established recently [4–7] that these three-step processes are inhibited at high intensities by the Lorentz force exerted by the magnetic-field component of the laser pulse, which tends to push the electron in the direction of propagation of the pulse, away from the core. This inhibition is particularly severe for electrons returning towards the core with a high relative velocity. In the conditions that can be achieved at GSI, however, and as we prove in this Letter, the inhibition is much attenuated, for the same relative velocity, by the Doppler boost in frequency arising from the relativistic motion of the ions [8].

Our calculations are carried out using the nonrelativistic, nondipole strong field approximation (SFA) we proposed recently [5,9]. Within this approach, the instantaneous dipole moment of the ion interacting with the laser field is given, in the rest frame of the ion, by the expression [9,10]

$$\mathbf{d}(t) \simeq -2\mathrm{Im}\sum_{t_d} \mathbf{a}^*_{\mathrm{rec}}(t, t_d) a_{\mathrm{pr}}(t, t_d) a_{\mathrm{ion}}(t, t_d). \qquad (1)$$

The sum runs over detachment times, t_d, for which the detached electron comes back to the core at time t. The ionization amplitude $a_{\mathrm{ion}}(t, t_d)$, the propagation amplitude $a_{\mathrm{pr}}(t, t_d)$, and the recombination amplitude $\mathbf{a}^*_{\mathrm{rec}}(t, t_d)$ are obtained in the way described in Refs. [9,10]. However, in the present work we correct the SFA for the Coulomb potential of the core. We assume that the electron interacts with the core through a pure Coulomb potential, $-Z/r$, with $Z = (2I_p)^{1/2}$, I_p being the ionization potential of the initial state. (Atomic units are assumed throughout this Letter, unless specified otherwise.) The ionization amplitude is calculated using the correction proposed by Krainov [11] for taking into account the effect of the Coulomb barrier on the tunneling electron. Accordingly, our $a_{\rm ion}(t, t_d)$ is a factor $(8I_p)^{3/4}/(\pi F_I)^{1/2}$ larger than the ionization amplitude given in Ref. [9]. The recombination amplitude is obtained by using the impulse approximation [12]. Namely, the amplitude is calculated assuming a Coulomb wave with incoming wave behavior rather than a plane wave for the spatial part of the wave function describing the returning continuum electron. The Coulomb interaction is taken into account in different ways in the ionization amplitude and the recombination amplitude because the corresponding steps are physically different: at the ionization stage, the electron escapes from the core with essentially zero velocity, while at the recombination stage it moves at high velocity. We have tested this procedure by comparing the Coulomb-corrected SFA photon emission spectrum with the result of a numerically exact integration of the time-dependent Schrödinger equation [13], for the case of an He^+ ion at rest in the laboratory and irradiated by an ultrashort Ti-sapphire pulse ($v = 0$, $\omega_I = \omega_L = 0.057$ a.u., $F_I = F_L = 0.534$ a.u.). Illustrative results are presented in Fig. 1, where the squared modulus of the Fourier transform of the dipole moment is plotted against the harmonic order q for q up to 400 ($q = \Omega_I/\omega_{\rm I}$, where Ω_I is the angular frequency of the emitted photon). The low-energy part of the spectrum, for $q < 40$, is dominated by bound-bound transitions. The rest of the spectrum arises from the three-step mechanism outlined above and can be readily understood by analyzing the classical trajectories of the detached electrons [14]. Except in the troughs of the oscillation, the Coulomb correction significantly improves the agreement of the SFA results with the exact results. (The same can also be said for the high energy part of the spectrum, not shown in Fig. 1, although in this case differences by up to a factor of 3 exist between the Coulomb-corrected SFA and the *ab initio* results.)

Let us now return to the case of relativistic ions. In order to examine how the relativistic Doppler boost affects three-step recollisional processes, we compare photon emission by two ions exposed to the same laboratory laser field. We assume that the first ion is at rest in the laboratory, while the second is moving relativistically in the direction opposite to the laser pulse. The latter is modeled as a monochromatic electromagnetic field of wavelength $\lambda_L = 1053.7$ nm and constant intensity $I_L = 10^{17}$ W cm^{-2} in the laboratory frame. This intensity is high enough for the magnetic-field induced suppression of recollision to be significant, so that nondipole effects must be taken into account, but not so high as to invalidate the nonrelativistic character of our nondipole SFA [9]. The ponderomotive energy, $U_p = F_L^2/(4\omega_L^2) = F_I^2/(4\omega_I^2)$, is 10.4 keV. The maximum energy of the photons emitted by the ion, in the ion's rest frame, is $I_p + 3.17U_p$ [3]. (Neither the ponderomotive energy, nor the Keldysh adiabaticity parameter $\gamma_K = \omega_L Z/F_L = \omega_{\rm I} Z/F_I$, depend on the speed of the ions.) The first ion is a sodiumlike Ar^{7+} ion ($Z = 3.247$, $I_p = 143$ eV), which is assumed to be at rest in the laboratory frame ($\gamma = 1$). For the laser parameters chosen here, this ion experiences an ionization loss of about 2% per optical cycle. The laser intensity is thus close to saturation. The corresponding Keldysh parameter is $\gamma_K = 0.08$. The second ion is a hydrogenlike Ne^{9+} ion ($Z = 10$, $I_p = 1.36$ keV), assumed to move in the laboratory frame with a Lorentz factor $\gamma = 15$. The ionization loss per cycle, in this case, is about 1%. (This species was chosen because at $\gamma = 15$ it ionizes in the field at about the same rate as Ar^{7+} at $\gamma = 1$, which facilitates the comparison.) The Keldysh parameter is 0.26, sufficiently small that the SFA can still be expected to be adequate. Owing to the Doppler boost in intensity, the Ar^{7+} ion would be promptly ionized if moving at $\gamma = 15$ against the laser pulse. Indeed, in the ion's frame of reference, the intensity is almost 9×10^{19} W cm^{-2} when $\gamma = 15$.

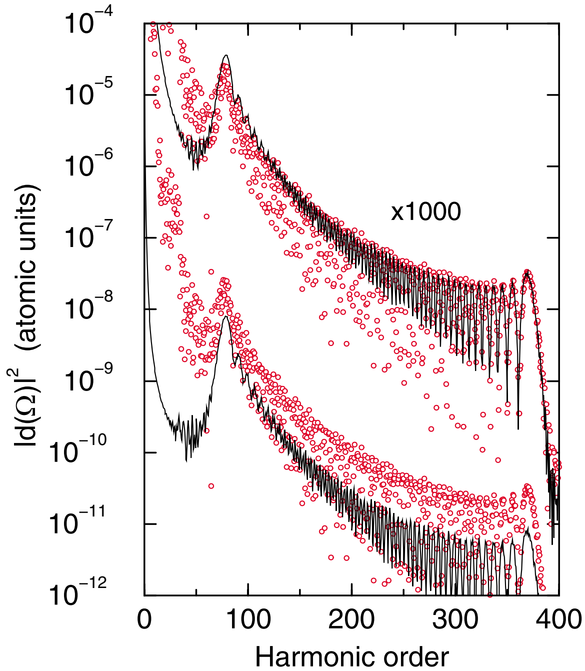

FIG. 1 (color online). The magnitude squared of the Fourier transform of the dipole moment as a function of the harmonic order, for a He^+ ion exposed to a 2-cycle laser pulse of 800 nm wavelength and 10^{16} W cm^{-2} peak intensity. Nondipole effects are negligible here. The vector potential is $\mathbf{A}(t) = \hat{\epsilon}(F_L/\omega_L)\sin^2(\omega_L t/4)\sin(\omega_L t + \varphi)$, where $\hat{\epsilon}$ is the polarization vector and the carrier phase $\varphi = 0$. The adiabatic approximation is not made. Solid lines: the predictions of the SFA, with (upper curves) and without (lower curves) the Coulomb corrections in the ionization and recombination amplitudes. For clarity, the results with Coulomb corrections are shifted upwards by a factor 1000. Circles: the spectrum resulting from a fully numerical solution of the time-dependent Schrödinger equation, and a copy of this spectrum shifted upwards by a factor 1000.

The spectra of the photons emitted by these two ions through the three-step mechanism are presented in Fig. 2 [15]. Two sets of results are shown for each ion, namely, the results obtained within the dipole approximation (where the effect of the magnetic-field component of the laser field is neglected), and the results obtained with the magnetic field taken into account within our nondipole SFA. In the case of Ar^{7+}, the magnetic field reduces the intensity of emission by about 3 orders of magnitude in the high energy part of the spectrum. This dramatic reduction, and also the bending of the plateau, disappearance of intermediate cutoffs and of the oscillations marking the dipole spectrum, are well-known nondipole effects and are readily explained within the semiclassical theory of three-step processes [7,9]. The difference between dipole and nondipole spectra is much smaller for Ne^{9+}. In particular, and despite the higher intensity, there is no significant decrease in the efficiency of the harmonic emission as compared to the dipole calculation. Also striking is the much larger intensity of emission.

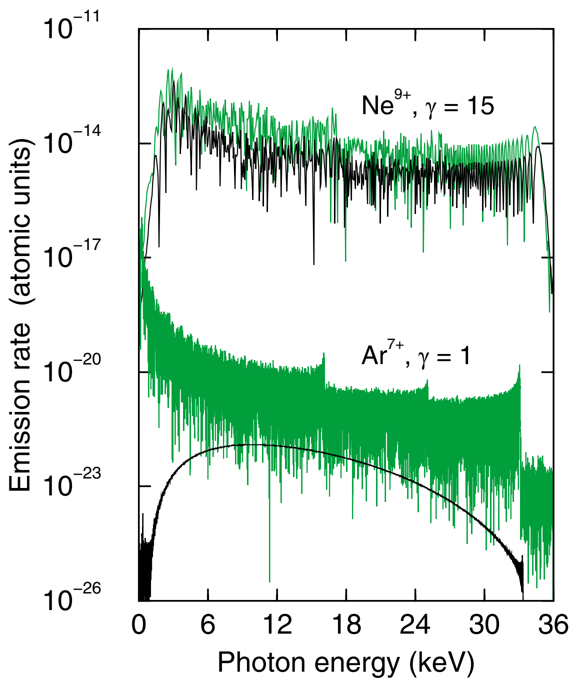

FIG. 2 (color online). The rate of emission of harmonic photons in the direction of propagation of the driving laser pulse, $\Omega^3|d_\epsilon(\Omega)|^2/(2\pi c^3)$, as a function of the photon energy for (a) a Ne^{9+} ion moving at $\gamma = 15$ in the laboratory frame, and (b) an Ar^{7+} ion at rest, as obtained within the Coulomb-corrected SFA. The rate and the photon energy are given in the ion's rest frame. For each ion, the upper curve shows the results obtained within the dipole approximation and the lower curve the results obtained without making this approximation. The laser field is monochromatic and linearly polarized. In the laboratory frame, the intensity is 10^{17} W cm^{-2} and the wavelength is 1053.7 nm.

The higher emission efficiency of Ne^{9+} has a double origin: It is due both to the larger recombination amplitude, which is proportional to $Z^{5/2}$, and to the lesser spreading of the electronic wave packet between the time of detachment and the time of recombination. (In the frame of reference of the ion, the difference between these 2 times is smaller by a factor 30 for $\gamma = 15$.) We expect that the adverse effect of wave packet spreading on the efficiency of high-order above-threshold ionization (ATI) and nonsequential double ionization would be similarly reduced by going to relativistic velocities.

The relative weakness of the nondipole effects in the case of Ne^{9+} originates from the dependence of the ionization amplitude $a_{\text{ion}}(t, t_d)$ on the magnetic field and on the ionization potential of the initial state. The largest part of the difference between the value of $a_{\text{ion}}(t, t_d)$ in the dipole approximation and its nondipole value can be traced to an exponential factor this amplitude is proportional to. In the ion's rest frame, one has [4,9]

$$|a_{\text{ion}}(t, t_d)|^2 \propto w(p_\parallel) = \exp\left[-\frac{2}{3}\frac{(2I_p + p^2)^{3/2}}{|F_I(t_d)|}\right], \quad (2)$$

where $F_I(t_d)$ is the electric field at the time of detachment and $p_\parallel$ is the momentum in the laser propagation direction that an electron must have at time t_d to return to the core at time t. Within the dipole approximation, $p_\parallel = 0$. In the nondipole SFA, which takes into account the Lorentz force acting on the electron due to the laser's magnetic-field component, $p_\parallel = \pi_k$, where

$$\pi_k = -\frac{1}{2c(t - t_d)}\int_{t_d}^{t} dt'|\mathbf{A}(t') - \mathbf{A}(t_d)|^2, \quad (3)$$

with $\mathbf{A}$ denoting the vector potential. For the above laser parameters, $\pi_k \approx 2$ a.u. for electrons returning with the highest possible kinetic energy ($3.17U_p$), and is the same for the two ions (π_k is a Lorentz invariant). In the dipole approximation, the probability for the active electron to be ionized and to return to the core at high velocity is similar for the two ions: taking $F_I(t_d)$ equal to the electric field amplitude, we obtain $w(p_\parallel = 0) = 1.4 \times 10^{-6}$ for Ar^{7+} at $\gamma = 1$ and $w(p_\parallel = 0) = 1.9 \times 10^{-6}$ for Ne^{9+} at $\gamma = 15$. However, beyond the dipole approximation we have $w(p_\parallel = 2) = 3.1 \times 10^{-10}$ for Ar^{7+} at $\gamma = 1$ whereas $w(p_\parallel = 2) = 8.7 \times 10^{-7}$ for Ne^{9+} at $\gamma = 15$. While the probability of returning to the ionic core is reduced for both ions when the laser's magnetic field is taken into account, it is much less reduced for Ne^{9+} than for Ar^{7+}. We note, with the authors of Ref. [4], that $w(\pi_k) \approx w(0) \times \exp[-\pi_k^2(2I_p)^{1/2}/F_I(t_d)]$. Since π_k is the same for $\gamma = 1$ and $\gamma = 15$, the smaller difference between dipole and nondipole results for Ne^{9+} follows from the smaller magnitude of $(2I_p)^{1/2}/F_I(t_d)$ for this ion. From a physical point of view, the width of the potential barrier is essentially $(I_p + p^2/2)/|F_I(t_d)|$, and is therefore larger by

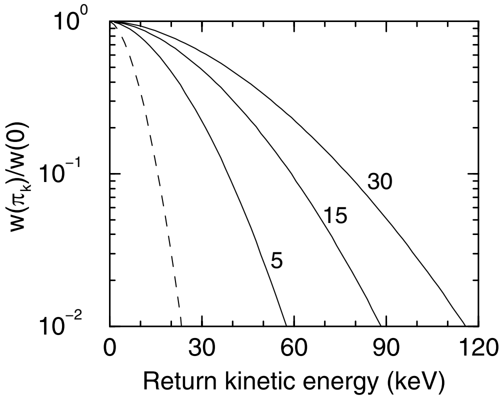

FIG. 3. The ratio $w(\pi_k)/w(0)$, indicative of the importance of the magnetic-drift induced suppression of recollision, for ions exposed to an intense 1053.7 nm laser pulse. Dashed line: ions at rest. Solid lines: ions moving at a Lorentz factor of 5, 15, or 30.

about $\pi_k^2/[2|F_I(t_d)|]$ in the nondipole SFA. Hence, the larger $|F_I(t_d)|$, for a constant π_k, the smaller the correction. It is thus more likely that the electron is ejected with the necessary longitudinal momentum.

The ratio $w(\pi_k)/w(0)$ therefore indicates how much the process is suppressed by the magnetic-field component of the laser pulse: the smaller this ratio, the larger the suppression [4,16]. The probability of a high energy recollision is insignificant unless $\pi_k^2(2I_p)^{1/2}/F_I(t_d)$ is small enough, and in the same time I_p is large enough for the ion to sustain the high intensities required but not that large that no ionization occurs. Assuming a driving field in the infrared, the former condition can be fulfilled, in principle, by taking γ large enough. This last point is illustrated by Fig. 3, which gives the ratio $w(\pi_k)/w(0)$ for returning electrons with maximal kinetic energy (i.e., 3.17 U_p). As above, the wavelength of the driving field is 1053.7 nm in the laboratory. Results are presented for ions either at rest or moving at $\gamma = 5$, 15, or 30. The ion species, and hence the value of I_p, is chosen in such a way that the ionization loss per cycle is 2% and is therefore different for different values of γ and U_p. The diagram shows that the range of return energies for which recollision is not severely suppressed can be extended significantly by using relativistic ions.

We note, to conclude, that the smaller importance of the nondipole effects at large Lorentz factors illustrated here for high-order harmonic generation can also be expected for high-order ATI or nonsequential double ionization, as it originates from the ionization stage of the process and this stage is virtually identical for all three-step processes. We stress that this smaller importance is *not* a relativistic effect *per se*. The speed of the ions intervenes only through the Doppler effect, which makes it possible to detach electrons from multicharged ions at high intensity without imposing a large magnetic drift to these electrons. Clearly, identical results would be obtained by irradiating ions at rest by a laser field of sufficiently high intensity and sufficiently high frequency.

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[15] For $\gamma \gg 1$, and depending on the angle of observation, the energy of the emitted photon may be larger by a factor almost 2γ in the laboratory frame.
[16] Eq. (7) of Ref. [4], which gives an estimate of the importance of nondipole effects, predicts that the ratio of rates of emission of harmonic photons between the dipole calculation and the nondipole calculations should be about 3×10^6 Ar^{7+} and 4.6 for Ne^{9+}, at the high energy end of the plateau. We find, respectively, 1×10^5 and 3.3.