Phase-matched coherent hard X-rays from relativistic high-order harmonic generation

M. C. Kohler, M. Klaiber, K. Z. Hatsagortsyan\(^{(a)}\) and C. H. Keitel

Max-Planck-Institut für Kernphysik - Saupfercheckweg 1, D-69117 Heidelberg, Germany, EU

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Abstract – High-order harmonic generation (HHG) with relativistically strong laser pulses is considered from multiply charged ions in counterpropagating, linearly polarized attosecond pulse trains. The propagation of the harmonics through the medium and the scaling of HHG into the multi-kilo-electronvolt regime are investigated. We show that the phase-mismatch caused by the free electron background can be compensated by an additional phase of the emitted harmonics specific to the considered setup which depends on the delay time between the pulse trains. This renders feasible the phase-matched emission of harmonics with photon energies of several tens of kilo-electronvolt from an underdense plasma.

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In the last decades, atomic high-order harmonic generation (HHG) in the non-relativistic regime \([1,2]\) has been developed to a reliable source of coherent extreme ultraviolet (XUV) radiation and of attosecond pulses \([3]\) opening the door for attosecond time-resolved spectroscopy \([4]\). The further advancement of this technique into the hard X-ray domain would, in particular, allow for ultrafast coherent diffraction imaging of single particles, clusters and biomolecules with sub-ångström resolution, tracking the electron motion in atoms and even for the investigation of time-resolved dynamics of nuclear excitations. The large-scale X-ray free electron lasers are likely to fulfill this task partly but are limited to energies around 10 keV \([5]\).

Is it possible to extend the table-top HHG sources into the hard X-ray domain? In principle, the harmonic photon energy can be increased by using stronger driving laser fields. The state-of-the-art technique allows to generate coherent X-ray photons up to the keV energy range \([6]\) and to produce short XUV pulses of less than 100 as \([7]\) from non-relativistic HHG in an atomic gas medium. The most favorable conversion efficiency for non-relativistic keV harmonics is anticipated with mid-infrared driving laser fields \([8]\). However, progress in this field appears to have reached a limit. Most importantly, the further increase of the driving field intensity transfers the interaction regime into the relativistic domain where the drift motion of the ionized electron in the laser field propagation direction prohibits the recollision and, consequently, suppresses HHG \([9]\). This happens when the drift distance becomes larger than the electron wave packet size at the recollision moment which is above laser intensities of \(4 \times 10^{16} \text{ W/cm}^2\) at infrared wavelengths \([10]\), corresponding to the HHG cutoff frequency of \(\omega_c \approx 10 \text{ keV}\). This indicates the limit of non-relativistic HHG. The second point hindering HHG at high intensities is the phase-matching problem. In strong laser fields, outer-shell electrons are rapidly ionized and produce a large free electron background causing phase-mismatch between the driving laser wave and the emitted X-rays. While in the non-relativistic regime macroscopic coherent emission can be achieved by optimizing the gas pressure in a setup with mid-infrared driving fields \([8]\), by employing non-adiabatic self-phase matching in very short driving laser pulses \([11]\) or by using quasi-phase-matching schemes, \(e.g.\) employing a weak counterpropagating infrared laser field \([12]\), in the relativistic regime the application of usual methods of phase-matching is not efficient because of the extreme shortness of the HHG coherence length, the large free electron background and the peculiarities of the specific relativistic HHG setup. Generating relativistic harmonics means mastering both challenges: circumventing the drift and coping with the phase-mismatch.

Various methods for counteracting the relativistic drift have been proposed. However, no universal solution has been found, each method has its drawbacks. To suppress

\(^{(a)}\)E-mail: k.hatsagortsyan@mpi-hd.mpg.de
the drift, highly charged ions moving relativistically against the laser propagation direction [13] or a gas of positronium atoms [14] can be used. Different combinations of laser fields have also been proposed for this purpose such as a tightly focused laser beam [15], counterpropagating crossed laser beams with linear polarization [16] or with equal-handed circular polarization [17]. In the latter field configuration, the relativistic drift is eliminated, however, in this scheme the phase-matching is especially problematic to realize [18]. In the weakly relativistic regime, the Lorentz force can also be compensated by a second weak laser beam being polarized in the strong beam propagation direction [19]. Two consecutive laser pulses [20] or the laser field assisted by a strong magnetic field [21] have been proposed as well but this requires large magnetic fields and dilute samples.

We have shown in [22–24] that strong attosecond pulse trains (APTs) employed as a driving field for HHG can be very useful to suppress the relativistic drift. However, all these efforts have only addressed the emission probability of a single atom rather than the coherent emission from a macroscopic gas target.

In this letter, we investigate the feasibility of phase-matched harmonic emission from an underdense plasma of multiply charged ions for a relativistic HHG setup employing two counterpropagating APTs. We show that HHG driven by counterpropagating APTs has an additional intrinsic phase depending on the time delay between the pulses as well as on the pulse intensity. This phase avoids to compensate the phase-mismatch between the driving laser field and the emitted harmonics due to the free electron background. The latter can be achieved by modulating the driving field intensity with a slowly decreasing envelope. We have performed a complete, quantitative analysis of the macroscopic yield of the relativistic HHG evidencing a small but detectable signal.

The applied setup for relativistic HHG is shown in fig. 1. First, we concentrate on describing the process in case of a single atom, see fig. 1(a). The driving fields are two counterpropagating APTs consisting of 100 as pulses with a peak intensity of the order of $10^{16}$ W/cm$^2$ and a spectral range of about 20 eV. Such pulses could be generated in the future by employing the relativistic oscillating mirror of an overdense plasma surface in a strong laser field [25] which can have a HHG efficiency approaching a few percent for harmonics less than 100 eV, as particle-in-cell simulations show [26]. The electron is liberated by the first pulse, driven by it in the continuum and undergoes the relativistic drift. This part of the trajectory is indicated by the light blue coloring. Thereafter, the electron propagates freely (gray dashed) in the continuum. A moment later, the second pulse reaches the electron, it reverts the drift and realizes rescattering (dark blue). The drift compensation is very efficient as one can deduce from fig. 2(a) where the single-atom spectral emission rate in the present setup (black) is compared with that in the dipole approximation (DA) (blue) where the drift is neglected. The heights of the plateaus coincide meaning that the setup exhibits no significant drift any more. Note that in the DA calculation the relativistic mass shift is included via the leading kinetic energy part of the Hamiltonian $H_{\text{kin}} = \sqrt{p^2c^2 + \ell^2} - \ell^2 \approx p^2/2 - p^2/(8c^2)$. The rate for the applied setup (the black curve in fig. 2(a)) is much larger than the one for a conventional laser field (the black curve in fig. 2(b)) with the same cutoff. In the latter case, due to the drift, the rate would drop rapidly with increasing laser intensity. When comparing both plots in DA (blue curves), we observe a small suppression of the discussed setup compared to the conventional field because the classical trajectories start in a shorter time window compared to a conventional field. The pulse shape offers an increased ionization probability in this time window but the first effect is stronger. The laser field strength of the discussed setup ($E_0 = 21$ a.u.) has to be higher than for a conventional setup ($E_0 = 2.7$ a.u.) to reach the same cutoff because of the larger center frequency of the APT.

We continue by describing the macroscopic properties of the setup. The contributions of different parts of the medium to the harmonic emission are shown in fig. 1(b). As in common HHG scenarios, the medium mainly emits along the driving field propagation axis ($z$-axis), into
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Fig. 2: (Color online) Single-atom HHG rates via [24]: (a) represents the discussed setup either fully relativistically (black) or in DA including the relativistic mass shift (blue); the delay between the pulses is 1.5 fs, the laser field strength \( E_0 = 21 \text{ a.u.} \), and \( I_p = 27.18 \text{ a.u.} \) \((\text{O}^{6+})\); (b) displays the scenario for a conventional propagating laser field (black) with \( E_0 = 2.7 \text{ a.u.} \), \( I_p = 7.35 \text{ a.u.} \) and (blue) the latter within the DA including the relativistic mass shift. The indicated parameters are chosen such that both the cutoffs and the average ADK-tunneling rates are the same for the two fully relativistic curves (black curves in (a) and (b)).

The left as well as into the right direction for symmetry reasons. As the emission mechanism is symmetric, we only consider emission into the right direction. In this case, necessarily, recombination has to be arranged by a pulse also propagating to the right because then the emitted harmonics and the pulse triggering recollision propagate in the same direction. Opposite directions would result in a strong phase-mismatch. In the figure we show the setup at the time when the counterpropagating APTs overlap. All ions in the shaded zones (A, C, E) have previously experienced a pulse \((e.g., \text{pulse} 1 \text{ or } 3)\) propagating to the left and have potentially been ionized by such a pulse. Then these parts of the medium will experience a pulse propagating to the right and emission in this direction will be possible until other pairs of pulses meet \((e.g., 2 \text{ and } 3)\). Thereafter, the white zones (B, D) fulfill the requirement of emission into the right. Note that contributions of atoms experiencing two pulses simultaneously (light shaded areas) are frustrated due to the chaotic trajectories of the ionized electrons in this region [27]. This limits the volume in longitudinal direction to about 1/3 and the possible pulse delays are between 1–2 fs.

In the following, we specify our model in more detail. As HHG medium, an underdense plasma of O\(^{6+}\) ions (ionization potential \( I_p = 27.18 \text{ a.u.} \)) is used which is immediately formed when the first laser pulse of relativistic intensity is applied to a neutral atomic gas. This is because the outer-shell electrons of an oxygen atom are almost instantaneously ionized due to a much smaller binding potential \((0.5 \text{ a.u.}–5.1 \text{ a.u.})\) other than the two remaining electrons in the closed 1s-shell. HHG is produced only by the tightly bound inner electron having an ionization potential corresponding to the tunneling condition at relativistic intensities. The O\(^{6+}\) emission is slightly reduced by the depletion to O\(^{7+}\) whereas the O\(^{7+}\) emission is not phase-matched in the proposed phase-matching scheme. The driving laser pulses are plane waves numerically propagated in the relativistic free electron background using a Crank-Nicolson algorithm. The density of the free electrons is assumed to be constant because the outer-shell ionization time is small compared to the laser period. Absorption of the high-frequency HHG photons can be neglected because their energy is much higher than the largest atomic transition energy.

In order to find the overall HHG yield from the macroscopic gas target, the photon spectral density \( dN/\omega_H \) in the far field is calculated (in atomic units) [28]:

\[
\frac{dN}{d\omega_H} = \frac{c}{4\pi^2\omega_H} R^2 \int d\Omega' |\tilde{E}(n',\omega_H)|^2,
\]

where \( n' \) is the emission direction, \( R \) the radius at an observation point, \( \omega_H \) the positive harmonic frequency (see fig. 1). The medium emits mainly along the z-directions, however, with a tiny divergence angle determined by the phase-matching geometry (see fig. 3). In (1) we calculate the overall emitted spectral photon number.
within the divergence angle which would be measured by a detector.

The spectral component of the electric field at the detector reads
\[
\tilde{E}(\mathbf{n}', \omega_H) = i \frac{\omega_H \rho e^{-i \omega_H R/c}}{Rc^2} \int d^3 r_a \times d^3 t \int j_a(x, t, r_a) e^{i \omega_H (t - r/c \cdot n')} \]
\[
= i \frac{\omega_H \rho e^{-i \omega_H R/c}}{Rc^2} \int d^3 x a \tilde{j}_a(x_a, \omega_H, \mathbf{n}'),
\]
where \(\rho\) is the density of the uniformly distributed ions, \(j_a(x, t, x_a)\) the current density at a single multiply charged ion in the fields defined in [24]. We included a tunneling correction for the free electron evolution in the counterpropagating laser pulses successively. Accordingly, in each stage of the excitation, we approximate the Green function by the Volkov Green function in a field of the appropriate single laser pulse (see [24]):
\[
G(x, x') \approx i \int d^3 x R G^V_2(x, x^R) \frac{\partial}{\partial \nu} G^V_2(x^R, x'),
\]
where \(G^V_2(x, x')\) are the Volkov Green functions [30] for the electron motion in the first and second pulses, respectively. The time-space coordinate \(x^R = (ct^R, x^R)\) indicates the intermediate moment between the two pulses when the first laser pulse has already left the wave packet of the active electron and the second laser pulse has not acted yet. Equation (3) is evaluated in the saddle point approximation.

To understand how phase-matching arises in this setup, let us have a closer look at the phase difference of the harmonics emitted from different ions separated by a distance \(\Delta z\) in the propagation direction:
\[
\Delta \varphi = \Delta \arg \tilde{j} \approx \Delta z \left( \frac{\omega_H}{c} \Delta \nu - \frac{\partial \nu_i}{\partial z} \right).
\]
The first term describes the phase-mismatch due to the free electron dispersion with \(\Delta \nu = \omega_H^2 / 2 \omega_H^2\), \(\nu\) being the electron plasma frequency and \(\nu_i\) the group velocity of the driving laser pulse, whereas the last term is the single-atom emission phase \((\nu_i)\) depending on the laser field conditions [32]. This intrinsic phase \(\nu_i\) is determined by the classical action of the electron trajectory recolliding with the specific harmonic energy and is an integral over the electron’s energy in the laser field plus the binding energy. To illustrate our numerical results, the integral can be estimated as \(\nu_i \approx (\alpha U_p(r_a) + f) \tau(r_a)\), with the local ponderomotive potential \(U_p(r_a) = (E_0 \rho(r_a))/2\omega^2\), the electron excursion time \(\tau(r_a)\), a numerical constant \(\alpha\), the local laser field amplitude \(E_0 \rho(r_a)\) and a central frequency of \(\omega_0\). Here we take into account that during the relativistic motion in the laser field the electron energy is proportional to \((E_0 \omega)^2\), see, e.g., ref. [33]. \(\alpha U_p(r_a)\) estimates the average electron energy in the short laser pulse. In fact, the harmonic single-atom cutoff of this field configuration can be estimated in good approximation with the non-relativistic cutoff law \(3.2 U_p + I_p\). Thus, \(\nu_i\) depends on the laser intensity as well as on the delay between the two pulses. The latter, being unique for this laser setup, mainly affects the electron excursion time \(\tau(r_a)\) and varies along the propagation direction. In order to achieve phase-matching, one can vary the laser intensity along the propagation direction in each zone to balance the intrinsic phase with the phase slip due to dispersion. The required intensity variation to have a constant complex phase \(\arg \tilde{j}_a\) in the entire medium is calculated numerically and shown in fig. 4(a) for the first interaction zone (A in fig. 1(b)). Due to the dispersion of the APTs in the plasma, the required peak electric-field variation slightly differs for different zones. Exact phase-matching can only be achieved for a single energy. We chose the
long trajectory of 50 keV energy but in principle phase-matching could be optimized for any energy value below the single-atom cutoffs. Note that only one of the short and long trajectories can be phase-matched since their classical actions are different. For the analytical description of the spatial variations of the laser field in the expression for $G(x, x')$, the eikonal-Volkov approximation is applied [34]. This is justified because the additional driving field causing the modulation perturbs the electron energy only slightly. Consequently, the second derivatives of the additional phase of the electron wave function as well as the square of the additional phase are neglected in the Klein-Gordon equation. The experimental realization of the phase-matched scheme could be achieved, e.g., with a modulated hollow core waveguide.

We employ a medium length as short as the spatial extent of the APT to minimize dispersion. In our simulation, each APT consists of 15 pulses with an APT duration of 40 fs. Our calculations show that in the case of longer APTs, the pulses in the train strongly spread due to dispersion and overlap, thus, violating the condition for the drift compensation. All pairs of pulses have almost the same coherent contribution to the overall yield. Since the pulses in different zones have experienced a different propagation length through the plasma, their shapes differ slightly. However, phase-matching still can be maintained by slightly adjusting the modulation profile, as long as the pulse shape still supports the recollision scheme. The phase-matching scheme imposes a strong demand on the jitter of the laser field $\Delta E/E$: $\Delta U_p \tau \ll 1$ yields $\Delta E/E \ll (U_p \tau)^{-1} \sim 10^{-4}$. We choose a gas density of $\rho = 10^{19}$ cm$^{-3}$ (ionized by the laser as described before), a diameter of 1 mm and a length of 12.5 $\mu$m for the interaction volume. The geometry involves a divergence angle on the order of $10^{-8}$ at 50 keV and the emitted spectral photon number being integrated over the solid angle is shown in fig. 4. An integral over the spectrum yields an emitted photon number of $10^{-7}$ at 50 keV per one collision of APTs which corresponds to a signal of about 10 photons per day at an assumed 1 kHz repetition rate. Note that the choice of the atomic species is rather flexible. Multi-electron highly charged ions offer an enhanced recombination probability due to core polarization [35] but produce a larger electron background that can be balanced by a lower gas density. The overall efficiency is maintained or could even be enhanced. The bandwidth of phase-matched HHG in this scheme is about 150 eV near the cutoff and pulses with a duration of about 35 as can be produced.

The small magnitude of the harmonic signal compared to current XUV HHG experiments [7] can be explained by investigating the spectral HHG photon rate $N_n$ of the harmonic order $n = \omega_H/\omega$ for a single atom [22,31]

$$N_n \sim w_i(t_i) |\langle 0| \mathbf{V}_H|\mathbf{p}\rangle|^2 (v_\perp^2 \tau^2 \partial \omega_H/\partial t_i)^{-1}. \tag{6}$$

Here $w_i(t_i)$ is the ionization rate with the ionization time $t_i$, $|\langle 0| \mathbf{V}_H|\mathbf{p}\rangle|$ the recombination amplitude and the last factor accounts for the dynamical properties of the wave packet. $v_\perp^2 \tau^2$ expresses the transversal electron spreading and transversal spreading velocity $v_\perp$, $\tau$ the excursion time of the electron and $\partial \omega_H/\partial t_i$ is the so-called electron wave packet chirping factor discussed below.

We proceed by analyzing the scaling of $N_n$ with increasing laser intensity at a harmonic energy near the respective cutoff provided that $w_i(t_i)$ is kept constant by an appropriate choice of $I_p$. The recombination amplitude decreases with increased electron energy favoring scattering rather than recombination. Its scaling depends on the shape of the ionic potential: $|\langle 0| \mathbf{V}_H|\mathbf{p}\rangle| \sim I_p^{5/4}/\omega_H^2$ for a hydrogen-like ion and $|\langle 0| \mathbf{V}_H|\mathbf{p}\rangle| \sim (\sqrt{I_p}/\omega_H)^{2}$ for a zero-range potential with $I_p \ll \omega_H$ and $p^2 \sim \omega_H$. Regarding the last term of eq. (6) which is derived from the functional determinant and is attributed to the wave packet nature of the electron, we follow [31] to find $v_\perp = \sqrt{E/I_p^{3/4}} \sim (\omega_H/I_p)^{1/4}$ and illustrate the chirping factor in fig. 5. It describes that the bandwidth of the harmonics emitted from a fixed ionization time window rises with increasing laser intensity (i.e., the ionization probability per harmonic decreases) and can be estimated as $\partial \omega_H/\partial t_i \sim \omega_H/\Delta t_i$. Thus, the photon emission rate in a constant bandwidth for a zero-range potential scales as $N_n \sim I_p/\omega_H^{5/2}$. A rough estimate for the scaling of $I_p$ at a constant ionization rate can be derived fixing the common tunneling exponent yielding $I_p \sim E^{2/3} \sim \omega_H^{1/3}$ and, consequently, $N_n \sim 1/\omega_H^{3.17}$. The decrease for a hydrogen-like potential is even more
an increase of the chirping factor. The ionization probability per harmonic frequency expressed by harmonics increases from (a) to (b), consequently, decreasing the increase of intensity but the bandwidth of the contained particular ionization time. ∆t remains unchanged under the increase of intensity but the bandwidth of the contained harmonics increases from (a) to (b), consequently, decreasing the ionization probability per harmonic frequency expressed by an increase of the chirping factor.

In conclusion, we have shown that the drift and phase-matching do not restrict HHG to the non-relativistic regime. The proposed setup renders the relativistic regime of HHG in a multi-atom ensemble accessible.

REFERENCES

[1] Corkum P. B., Phys. Rev. Lett., 71 (1993) 1994.
[2] Lewenstein M. et al., Phys. Rev. A, 49 (1994) 2117.
[3] Agostini P. and DiMauro L. F., Rep. Prog. Phys., 67 (2004) 813.
[4] Krausz F. and Ivanov M., Rev. Mod. Phys., 81 (2009) 163.
[5] A small fraction of the FEL spectrum can also extend to few tens of keV being low-order harmonics of the main FEL emission line, see, e.g., Altarelli M. et al., XFEL The European X-Ray Free-Electron Laser Technical Design Report, DESY 2006-097, DESY, Hamburg, July 2006.
[6] Seres J. et al., Nature (London), 433 (2005) 596; Seres E. et al., Appl. Phys. Lett., 89 (2006) 181919; Sansone G. et al., Science, 314 (2006) 445.
[7] Goulielmakis E. et al., Science, 320 (2008) 1614.
[8] Popmintchev T. et al., Proc. Natl. Acad. Sci. U.S.A., 106 (2009) 10516; Chen M.-C. et al., Phys. Rev. Lett., 105 (2010) 173901.
[9] Salamin Y. I. et al., Phys. Rep., 427 (2006) 41.
[10] Palaniyappan S. et al., Phys. Rev. A, 74 (2006) 033403.
[11] Tempea G. et al., Phys. Rev. Lett., 84 (2000) 4329.
[12] Peatross J. et al., Opt. Express, 1 (1997) 114; Zhang X. H. et al., Nat. Phys., 3 (2007) 270.
[13] Mocken G. and Keitel C. H., J. Phys. B, 37 (2004) L275; Chirilă C. C. et al., Phys. Rev. Lett., 93 (2004) 243603.
[14] Henrich B., Hatsagortsyan K. Z. and Keitel C. H., Phys. Rev. Lett., 93 (2004) 013601; Hatsagortsyan K. Z., Müller C. and Keitel C. H., EPL, 76 (2006) 29.
[15] Lin Q., Li S. and Becker W., Opt. Lett., 31 (2006) 2163.
[16] Kylstra N. J. et al., Phys. Rev. Lett., 85 (2000) 1835; Taranukhin V. D., Laser Phys., 10 (2000) 330; Verschil M. and Keitel C. H., Phys. Rev. ST Accel. Beams, 10 (2007) 024001.
[17] Milesević N., Corkum P. B. and Brabec T., Phys. Rev. Lett., 92 (2004) 013002.
[18] Liu C. et al., New J. Phys., 11 (2009) 105045.
[19] Chirilă C. C. et al., Phys. Rev. A, 66 (2002) 063411.
[20] Verschil M. and Keitel C. H., J. Phys. B, 40 (2007) F69.
[21] Verschil M. and Keitel C. H., EPL, 77 (2007) 64004.
[22] Klaiber M., Hatsagortsyan K. Z. and Keitel C. H., Phys. Rev. A, 74 (2006) 051803(R); 75 (2007) 063413.
[23] Klaiber M. et al., Opt. Lett., 33 (2008) 411.
[24] Hatsagortsyan K. Z. et al., J. Opt. Soc. Am. B, 25 (2008) 93.
[25] Nomura Y. et al., Nat. Phys., 5 (2009) 124; Dromey B. et al., Nat. Phys., 5 (2009) 146.
[26] Tsakiris G. D. et al., New J. Phys., 8 (2006) 19.
[27] Bauer D., Mulsner P. and Steebe W.-H., Phys. Rev. Lett., 75 (1995) 4622.
[28] Landau L. D. and Lifshitz E. M., The Classical Theory of Field (Pergamon, London) 1975.
[29] Reiss H. R., Phys. Rev. A, 642 (1990) 1476; J. Opt. Soc. Am., 7 (1990) 574.
[30] Milošević D. B., Hu S. and Becker W., Phys. Rev. A, 63 (2001) 011403(R); Laser Phys., 12 (2002) 389.
[31] Ivanov M. Y. et al., Phys. Rev. A, 54 (1996) 742.
[32] Salières P. et al., Science, 292 (2001) 902.
[33] Sarachik E. S. and Schappert G. T., Phys. Rev. D, 1 (1970) 2738.
[34] Avetissian H. K. et al., Phys. Rev. A, 59 (549) 1999; Smirnova O. et al., Phys. Rev. A, 77 (2008) 033407.
[35] Gordon A. et al., Phys. Rev. Lett., 96 (2006) 232902.