Quasi-Phase-Matching High-Harmonic Radiation Using Chirped THz Pulses

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High-order harmonic generation in the presence of a chirped THz pulse is investigated numerically with a complete 3D nonadiabatic model. The assisting THz pulse illuminates the high-order harmonic generation gas cell laterally inducing quasi-phase-matching. We demonstrate that it is possible to compensate the phase mismatch during propagation and extend the macroscopic cutoff of a propagated strong IR pulse to the single-dipole cutoff. We obtain 2 orders of magnitude increase in the harmonic efficiency of cutoff harmonics (≈170 eV) using a THz pulse of constant wavelength, and a further factor of 3 enhancement when a chirped THz pulse is used.

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High-order harmonic generation (HHG) in a noble gas medium is currently the widely used method to produce coherent radiation in the XUV and soft-x-ray regime. The elementary laser-atom interaction leading to HHG is understood via the three-step model [1], but the complete description of the HHG process requires considering the elementary laser-atom interaction together with the macroscopic aspects of laser and harmonic field propagation in the ionized gaseous medium. The temporal, spectral, and spatial distortions of the fundamental pulse during propagation result in a varying intensity and phase which strongly influence the produced harmonic radiation on both the single-atom and the macroscopic level [2]. The ultimate goal in an experiment is to obtain intense, coherent XUV or soft-x-ray radiation which in turn are useful in the designed applications like molecular imaging or pump-probe experiments [3].

In Fig. 1 we show a typical case of phase mismatch (PMM) of the harmonic radiation along the HHG cell manifesting in a quasiperiodic oscillation of the harmonic intensity named Maker fringes [4]. In order to improve the phase-matching (PM) condition, a number of quasi-phase-matching (QPM) techniques have been developed to at least partially compensate for the PMM arising during propagation between the source and generated harmonics. There are currently several typical QPM techniques relying either on periodic modulation of the generating field strength: propagation in diameter-modulated capillaries [5]; HHG assisted by a periodic static electric field (or other periodic waveform, e.g., sawtooth) [6,7]; QPM induced using two counter-propagating pulses [8,9]; QPM through multimode excitation in a waveguide [10]. An alternative QPM arrangement relies on periodic off switching of generation in a multijet configuration [11,12].

We present here a new scheme, inspired by the idea described in [6] where the HHG gas cell was placed in a static electric field periodically modulated along the propagation direction; a pulsed CO2 laser and an amplitude mask were proposed to create ≈7 MV/cm static electric field periodically distributed along the HHG cell. In our opinion, this configuration was unrealistic because: (1) the CO2 laser pulse with ≈10 μm wavelength cannot be considered as a static electric field when compared to the ≈100 μm mask periodicity (2 mm cell length), and (2) producing a 7 MV/cm static electric field is not feasible experimentally. Yet, it is possible to overcome the shortcomings of the QPM configuration presented in [6]: we use a THz field imposing a proper time evolution to create QPM conditions. The advantages of this new scheme are multiple: strong THz pulses can be generated by optical rectification of femtosecond laser pulses, and as such the resulted THz pulse is inherently synchronized in time with the laser pulse generating the harmonics; fine control in space and time is available by tuning the wavelength, peak amplitude, chirp rate, and initial phase, thus the shape of the THz pulse can be adapted in order to optimize QPM and harmonic yield, and there is no need to

![FIG. 1 (color online). On-axis variation of the H111 along the propagation direction generated by the IR pulse. The width of several Maker fringes = 2Lcoh is indicated in μm, other parameters are in the text.](image-url)
apply any amplitude mask. We show a schematic representation of a possible geometric configuration in Fig. 2.

The THz field modifies the electron trajectories and hence the single-atom dipole phase (depending on return time and action). As the generating IR pulse propagates along the gas cell, the temporal variation of the THz pulse, which is simultaneously present with the IR, translates to a spatial modulation of the HHG conditions along z. According to our results presented below, assisting pulses of a few (or few tens) THz frequency and a few MV/cm peak electric field are required for the enhancement of HHG. In this wavelength range, the highest electric field already presented is 1.2 MV/cm [13] achieved with THz pulses having 1 μJ energy obtained using a tilted pulse front setup [14]. Recently THz pulses with 125 μJ energy were generated and production of THz pulses with more than 10 mJ were predicted [15]. These were single-cycle THz pulses, but it is expected that using quasisinusoidal intensity modulated pump laser pulses [16] multicycle THz pulses with similar energy can be generated resulting up to 10 MV/cm field strength in 10 ps long THz pulse on minimum 3 mm diameter spot.

The numerical model in which the THz-assisted HHG generation is implemented is based on a complete 3D nonadiabatic model [17], further developed to two-color HHG using arbitrary wavelength copropagating pulses [18]. The THz-assisted HHG has also been studied in the configuration where the two pulses propagate in the same direction [19]. For the current work the model has been extended to describe pulses propagating perpendicularly.

(i) The propagation equation for the IR pulse is solved accounting for diffraction, dispersion (on neutrals and plasma), absorption and nonlinear Kerr effect; (ii) nonlinear dipole response is calculated in the strong-field approximation [20]; (iii) the propagation equation for the generated harmonics is solved. In order to gain a more intuitive insight into the physics behind the effect of the modulating THz pulse, saddle-point calculations were also carried out providing the phases of the single-atom dipole and the total harmonic field.

We present a case study to demonstrate the new QPM scheme with THz-assisted HHG. We start from a standard HHG system by an IR pulse while looking for the optimal configuration of the THz pulse which induces the best possible QPM in a selected harmonic range. The particular conditions for the HHG are the following: the IR pulse has 800 nm central wavelength, 20 fs pulse duration, 0.2 mJ pulse energy, focused with a 20 cm focal length mirror resulting in 8 × 10^14 W/cm^2 peak intensity and 25 μm beam waist. The HHG cell is 2 mm long filled with 25 Torr Ne and begins 1 mm before the laser focus. All distances throughout this paper are measured from the laser focus. The single-dipole cutoff order is q = 111 (based on the relation I_p + 3.17U_p) corresponding to 172 eV and QPM is optimized for this spectral range.

Figure 1 illustrates the on-axis intensity variation of the 111th harmonic (H111) along the propagation axis as generated with the 800 nm pulse alone. We can see the experimentally observable high-contrast Maker fringes, a typical result of PMM in nonlinear phenomena. On the figure we also observe that PM conditions vary along the cell, the main causes being the intensity and phase changes of the generating pulse as it propagates. In particular, the width of the fringes increases from 90 to 290 μm during 2 mm of propagation. The actual period of the high-harmonic intensity modulation and its variation is determined by the focusing conditions of the IR pulse. We use the THz field to modulate HHG conditions along the cell and compensate for PMM. We note here that obviously, looser focusing provides longer coherence lengths, but the here proposed arrangement can be equivalently applied for longer cells with longer THz wavelengths.

From the results shown in Fig. 1 it is straightforward that for optimal QPM the wavelength of the modulation needs to be varied, a requirement that has been observed in [21]. We therefore aim at producing a quasiperiodic spatial modulation of the generating conditions via the temporal modulation of the wavelength (chirp) of our THz pulse. A QPM effect of the perpendicularly propagating THz field is expected when the modulation caused by the field matches the Maker fringes observed in the IR-only case [6]. To verify this concept, we studied the effect of a THz field of fixed wavelength (120 μm, corresponding to 2.5 THz) on the HHG yield shown in Fig. 3(b) with the blue dashed curve. The black solid curve in the top part of the figure illustrates the oscillation of harmonic intensity for the IR-only case replotted from Fig. 1 in the same arbitrary units. We note here, that the THz amplitude and delay have been chosen appropriately.

For more efficient QPM we use a chirped THz pulse such that the wavelength variation of the THz pulse along the cell best matches the fringe structure of the chosen harmonic: \( \lambda_{THz}(z) \approx 2L_{coh}(z) \). In this case study, we designed a linearly chirped THz pulse that follows nicely the field oscillations, as illustrated in Fig. 3(a) with the green dashed curve. The parameters of the perpendicular THz field are: Gaussian pulse profile with 110 μm initial
the electric field. The total harmonic field along
are responsible for HHG, thus both amplitude and phase of
enhancement.

or moving the focus position) shows a
THz field (realizable experimentally by shortening the cell
intensities along the cell obtained with and without the
pattern leading to loss of QPM. Comparing the highest
THz field walks off the H111 IR-only intensity modulation
divergence and plasma defocusing of the beam, and the
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solid line in Fig. 3(b) indicates very efficient QPM for
ration, and 4 mm beam waist. For these parameters the red

/wavelength, $-9 \times 10^{-7}$ fs$^{-2}$ chirp rate resulting 180 $\mu$m
average wavelength, 5 MV/cm peak amplitude, 8 ps du-
ration, and 4 mm beam waist. For these parameters the red
solid line in Fig. 3(b) indicates very efficient QPM for
H111 up to $\approx 1.2$ mm in the cell. We note here that at
the end of the cell H111 emission is reduced due to
divergence and plasma defocusing of the beam, and the
THz field walks off the H111 IR-only intensity modulation
pattern leading to loss of QPM. Comparing the highest
intensities along the cell obtained with and without the
THz field (realizable experimentally by shortening the cell
or moving the focus position) shows a $\approx 300$ times yield
enhancement.

Optically ionized electrons traveling in the laser field
are responsible for HHG, thus both amplitude and phase of
the high harmonics are very sensitive to the exact shape of
the electric field. The total harmonic field along $z$ is the
sum of successive dipole emissions:

$$E_q(z)e^{i\varphi_q(z)} = \int_0^z d_q(z')dz' = \int_0^z a_q(z')e^{-i\varphi_q(z')}dz', \tag{1}$$

where $E_q(z)$ and $\varphi_q(z)$ are amplitude and phase of the qth
order HH field accumulated till $z$, $d$ is the qth Fourier
component of the dipole moment vector, and the dipole
phase $\varphi_q(z) = q\varphi_{IR}(z) + \Phi_{at}(z)$ is described in a traveling
frame moving with $c$. The applied THz pulse amplitude is
weak compared to the generating IR ($<1\%$), thus its main
effect is the modulation of the accumulated phase of the
electron while traveling in the field $|\Phi_{at}(z)|$. Efficient QPM
is explained at the harmonic dipole level. In particular, for
H111 the dipole amplitude variation $a_q(z)$ did not show any
correlation with $E_q(z)$, instead we found strong correlation
with the phase. Using saddle-point calculations for short-
to-cutoff trajectories in the central half-cycle of the IR
pulse we studied the $\varphi_q(z)$ and $\Phi_q(z)$ variations, and
compared the phase difference $\Phi_q(z) - \varphi_q(z)$ with the
total harmonic intensity $|E_q(z)|^2$. The results are shown
in Fig. 4 for the central part of the cell when the HH field is
built with IR pulse only (a), (c) and when assisted
with chirped THz pulse (b), (d). As expected, while
$|\Phi_q(z) - \varphi_q(z)| < \pi/2$ the successive dipole emissions
add up constructively and result in an overall increase of
the total harmonic field. The effect of the THz field is that it
modulates the dipole phase such to keep $|\Phi_q(z) - \varphi_q(z)|$
within $\pi/2$ over longer propagation distances and pass
through destructive zones in much shorter regions.

In Fig. 5 we compare harmonic spectra, on logarithmic
scale, obtained in different conditions. The black line
indicates the spectrum produced in a very thin ($20 \mu$m)
target with the IR pulse alone, which gives us practically
the single-atom spectrum, with the expected cutoff located
at H111. The conversion efficiency is very low due to
the short cell. The red line indicates the spectrum produced
by the IR pulse recorded at the position in cell where
we obtained the highest H111 intensity, i.e., $-40 \mu$m
[cf. Figure 3(a)]. We observe that harmonics in the lower
plateau (below 110 eV) experience an increase ($\approx 10^2$
times) with cell length, as more atoms contribute to the
HHG process, and apparently phase matching is favorable.
For the higher plateau and cutoff harmonics we observe
no yield enhancement with increasing cell length, which
is explained by the PPM discussed in detail for H111.

FIG. 3 (color online). (a) Black solid line: THz-free variation
of H111 intensity. Green dotted line: $E_{THz}(z)$ visualizes the
correspondence with the $2L_{coh}(z)$. (b) Red solid line: On-axis
variation of the H111 intensity along the propagation direction
when a chirped THz pulse is applied. Blue dashed line: Same
when the THz pulse has constant $\lambda = 120 \mu$m. Vertical lines
indicate the positions of maximum harmonic yield.

FIG. 4 (color online). Variation of the H111 dipole phase and
total harmonic phase without (a) and with THz field present (b).
Red-green dots: $\varphi_q(z) - \varphi_{IR}(z)$ phase difference with scale on
the left. Black line: H111 intensity $|E_q(z)|^2$, scale on the right.
(c) IR-only case; (d) THz-assisted case.

193903-3
Further investigation is going on to demonstrate that the presented QPM configuration is flexible and that by tuning THz pulse parameters one can select the spectral range in which harmonic radiation can be amplified. Our results demonstrate that by tuning the THz field one can finely shift the central frequency of the amplified spectral range toward lower harmonics. Another key is the IR pulse energy which shifts the HH cutoff. For our case, a ±25% IR pulse energy variation combined with appropriate THz field allowed amplification in the 140–220 eV spectral domain. This might be of central importance in seeding free electron lasers in this range of photon energies. Recently mid-IR laser pulses are being also used for HHG in order to achieve higher cutoff energy. As the harmonic yield scales with $A^{-5.5}$ [23], an efficient QPM is even more important for this domain. Applying the THz-assisted QPM scheme could significantly increase the harmonic yield at selected spectral ranges approaching the water-window.

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