On-the-Fly Control of High-Harmonic Generation Using a Structured Pump Beam

Liran Hareli, Lilya Lobachinsky, Georgiy Shoulga, Yaniv Eliezer, Linor Michaeli, and Alon Bahabad

Department of Physical Electronics, School of Electrical Engineering, Fleischman Faculty of Engineering, Tel-Aviv University, Tel-Aviv 69978, Israel

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We demonstrate experimentally a relatively simple yet powerful all-optical enhancement and control technique for high harmonic generation. This is achieved by using as a pump beam two different spatial optical modes interfering together to realize tunable periodic quasi-phase matching of the interaction. With this technique, we demonstrate on-the-fly quasi-phase matching of harmonic orders 29–41 at ambient gas pressure levels of 50 and 100 Torr, where an up to 100-fold enhancement of the emission is observed. The technique is scalable to different harmonic orders and ambient pressure conditions.

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Introduction.—High harmonic generation (HHG) is an extreme nonlinear optical up-conversion process driven with an intense ultrashort laser pulse, usually interacting with noble gas atoms [1–3]. The up-converted light can easily reach the extreme UV portion of the spectrum and in certain cases can extend up to keV x-ray photon energies [4]. At the single atom level, the process of HHG is most simply described by the three-step model [5,6] consisting of electron ionization, laser-driven acceleration, and recombination upon which excess kinetic energy is released in the form of a high-energy photon.

Macroscopically, due to dispersion and geometric effects such as waveguiding and focusing, the efficient buildup of each harmonic order is limited to its coherence length \( L_c = \pi/\Delta k_q \), which is the distance at which the accumulated phase difference between the pump beam and the harmonic emission is equal to \( \pi \). Here, \( \Delta k_q \) is the momentum phase mismatch—the difference between the wave vectors of the pump beam and the \( q \)th order harmonic signal in the direction of propagation.

Perfect phase matching conditions, \( \Delta k_q = 0 \), can be achieved at low intensities (low ionization rates) and loose focusing [7,8]. However, when working with a strong pump beam, plasma dispersion becomes dominant and phase matching is not possible [9]. Thus, a different approach is required for achieving an efficient buildup of the emitted radiation. The solution comes in the form of an ordered modulation of a parameter relevant to the interaction, which allows the restrictive momentum conservation condition associated with perfect phase matching to be replaced with a less restrictive condition [10] known as quasi-phase matching (QPM) [11,12]. In particular, using a periodic spatial modulation allows quasi-phase matching when \( \Delta k_q = 2\pi/\Lambda \), where \( \Lambda \) is the period of the modulation.

To date, most of the works applying QPM on HHG did this by modulating the medium properties [13–18], while others showed all-optical QPM theoretically [19–22] and experimentally [23,24]. While some of these works demonstrated tunability, they were associated with mechanically moving components. In addition, although a relatively simple method for a copropagating all-optical QPM was suggested [25], the all-optical methods that were demonstrated experimentally involved counterpropagating geometries which led to relatively complicated front ends [2,23,24].

Here we present experimentally an all-optical amplitude-beating scheme for QPM of HHG where the pump modes are copropagating and nonguided within a semi-infinite gas cell. As the modes are unguided, their wave vectors can be chosen from a continuum of available values, leading to an unprecedented degree of tunability of the applied modulation parameters. The copropagating scheme greatly reduces the complexity of the optical setup. Overall, this scheme allows for nonmechanical, on-the-fly, and complete control of the parameters of the perturbing amplitude modulation. In particular, the period of the modulation, its overall length, and the depth are easily controlled. With this scheme, we experimentally demonstrate tunable QPM of harmonic orders 29–41 in argon at ambient gas pressures of 50 and 100 Torr, with an up to 100-fold enhancement of the emission. This method is scalable to address different harmonic orders and ambient pressure conditions.

Theory.—HHG in a so-called semi-infinite gas cell [26] is driven with a pump beam which is focused to a point at the vicinity of a small exit hole in a thin foil terminating a gas cell. Vacuum conditions are established immediately after the cell. The phase mismatch for the generation of the \( q \)th harmonic is given by [27–29]

\[
\Delta k_q = qk_{\text{io}} - k_{q\omega} = qP \left( \frac{2\pi}{\Lambda} \Delta n_q - \eta N_{\text{atm}} r_e \lambda \right) + q \partial_z \phi_{\text{geometric}} + \partial_z \phi_q,
\]

where \( \Delta n_q \) is the linear index of refraction change of the gas, \( N_{\text{atm}} \) is the number density of the gas, \( r_e \) is the electron radius, and \( \lambda \) is the wavelength of the incident light. The term \( \phi_{\text{geometric}} \) is the geometric phase shift due to the spatial modulation, and \( \phi_q \) is the phase shift associated with the \( q \)th harmonic.

\( \Delta k_q \) is the distance at which the accumulated phase difference between the pump beam and the harmonic emission is equal to \( \pi \). This is achieved by modulating the medium properties with a spatial modulation allowing quasi-phase matching when \( \Delta k_q = 2\pi/\Lambda \), where \( \Lambda \) is the period of the modulation.
where \( q \) is the harmonic order, \( k_{p} \) is the pump wave vector, \( k_{q0} \) is the \( q \)th harmonic wave vector, \( P \) is the gas pressure, \( \eta \) is the ionization rate at the time of the emission of the harmonic radiation, \( \lambda \) is the pump wavelength, \( \Delta n_{q} \) is the difference between the refractive indices of neutral gas atoms at the pump and the \( q \)th harmonic frequencies, \( N_{\text{atm}} \) is the number density of atoms at atmospheric pressure, \( r_{c} \) is the classical electron radius, \( \phi_{\text{geometric}} \) is the geometric phase, and, finally, \( \phi_{q} \) is the intrinsic phase of the atomic dipole which is proportional to the pump intensity \[^{29,30}\]. The two pressure-dependent terms in Eq. (1) are due to gas and plasma dispersion, respectively. The third term is the geometric wave-vector mismatch, which is usually approximated to depend only on the geometric variation of the pump phase and is mostly associated with either waveguiding effects or with focusing (contributing a Gouy phase term).

We consider now the perturbation of the up-conversion process, in which \( q \) photons of an intense linearly polarized Gaussian beam are converted to a \( q \)th harmonic order, by the addition of a linearly polarized weak Bessel beam. In order to produce a superposition of a Gaussian and a Bessel beam at the interaction region, we split a Gaussian beam into two and modulate the spatial distribution of one part using a spatial light modulator (SLM). The modulated beam obtains the shape of a ring which is the Fourier transform of a finite Bessel beam \[^{31,32}\]. Focusing the ring in a \( 2f \) configuration creates a Bessel beam which is nondiffracting along a distance \( L_{\text{ND}} \) around the focus of the lens [see Fig. 1(a)]. Using simple geometrical optics, it can be shown that \( L_{\text{ND}} \) is dependent on the ring thickness \( W \), radius \( R \), and the lens focal length \( f \):

\[
L_{\text{ND}} = \frac{2f^{2}}{WR}.
\]

The two spatial modes comprising the beam (Gaussian and Bessel) possess different axial wave-vector components whose difference is denoted by \( \Delta k_{0} \) [see Fig. 1(b)]. As such, the two modes accumulate phase at different spatial rates. When the two beams are superposed coaxially to produce a structured pump beam, a periodic intensity and phase modulation, with an underlying period of \( 2\pi/\Delta k_{0} \), emerges on axis [see Fig. 1(c)].

The on-axis intensity and geometric phase of the structured pump beam can be approximated by

\[
I_{\text{total}}(z) = I_{G}(z) \{ 1 + 2\beta \cos[\phi_{\text{Gouy}}(z) + \Delta k_{0}(z - Z_{0})] \},
\]

\[
\phi(z)_{\text{geo}} = \int_{0}^{z} k_{\text{geo}}(z) dz
\]

\[
= \arg \left( \sin[\phi_{\text{Gouy}}(z)] - \beta \sin[\Delta k_{0}(z - Z_{0})] \right) + i \left( \cos[\phi_{\text{Gouy}}(z)] + \beta \cos[\Delta k_{0}(z - Z_{0})] \right),
\]

where \( \beta = \beta_{0} \sin[\pi(z - Z_{0})/L_{\text{ND}}] \) is the amplitude modulation depth (amplitude ratio between the Bessel and Gaussian fields at the focus of the Bessel beam), \( \phi_{\text{Gouy}}(z) = \arctan(z/z) \) is the on-axis Gouy phase of the Gaussian beam, where \( z_{R} \) is the Gaussian beam Rayleigh range, \( I_{G}(z) \) is the intensity of the Gaussian beam, and \( L_{\text{ND}} \) is the nondiffracting distance which is defined in Eq. (2). \( Z_{0} \) is the difference in the focus position between the two beams. This approximation is valid for a weak perturbation (\( \beta \ll 1 \)) and shallow focusing (\( L_{\text{ND}} \ll z_{R} \)). The intensity modulation acts on both the amplitude and phase components of the nonlinear polarization, which has a tendency to oppose each other for emission buildup associated with QPM \[^{21,33}\]. In the current case, taking all modulation factors into account, the modulation depth of the amplitude and the phase of the nonlinear polarization are different; thus, the overall modulation can be used, as we verify here, to realize an efficient all-optical copropagating QPM scheme.

The quasi-phase matching condition for the \( q \)th harmonic order can be achieved when \( \Delta k_{0} = \Delta k_{q} \). For a given focal length \( f \), the radius of the imaged ring, \( R \), determines the angle \( \theta \) of the Bessel wave vectors and consequently determines the value of \( \Delta k_{0} \):

\[
\Lambda = \frac{2\pi}{\Delta k_{0}} = \frac{\lambda}{1 - \cos(\theta)}.
\]

The asymptotic limit for the periodicity that can be achieved with such a setup is equal to the wavelength being used, which is twice the periodicity available with counterpropagating fields. Practical setups would set this limit to between a few to ten wavelengths. The number of periods within the modulation is given with \( L_{\text{ND}}/\Lambda \), where \( L_{\text{ND}} \) in turn is dependent on the thickness of the imaged ring [see Eq. (2)]. Therefore, \( L_{\text{ND}} \) determines the effective phase
matching bandwidth $\delta k$, which roughly behaves as $\delta k = \pi/L_{ND}$. This follows from approximating the intensity buildup of the harmonic emission for a given modulation depth as proportional to [34]

$$L_{ND}^2 \text{sinc}^2\left(\frac{(\Delta k - \Delta k_0)L_{ND}}{2}\right)$$

with $\Delta k$ being any given value of the phase mismatch. For an efficient use of the available number of periods (as well as for the last approximation to be of any value), it is essential that the effective interaction length is at least as long as $L_{ND}$. This is easily accomplished by using a shallower focusing for the Gaussian beam than for the Bessel beam. The depth of the modulation is determined by the ratio of the intensities of the two beams. Using thinner rings increases $L_{ND}$ but decreases the intensity of the Bessel beam and so reduces the modulation depth. The modulation depth is also dependent on the ring radius—due to the Gaussian profile of the beam reflecting of the SLM. Larger ring radius decreases the intensity of the Bessel beam. Finally, the modulation depth can also be tuned by adjusting the overall transmission of any given ring, which is also controllable with the SLM. In our experiment, the ring transmission was set to its maximal value. As the radius and width of the rings are controlled by the software operating the SLM, it is very easy to exert on-the-fly control on the geometric parameters of the quasi-phase matching modulation—effectively changing the center of the phase matching curve $\Delta k_0$, its bandwidth $\delta k$, and its modulation depth.

Experimental results.—To attain a relatively long Rayleigh range for the Gaussian beam and significant cone half angles $\theta$ for the Bessel beam, different focusing conditions are required for the two beams. As such, the output beam of a 1 kHz, 35 fs Ti:sapphire amplifier (Coherent Legend USX) is split into two paths (see Fig. 2). The first beam, having 0.6 mJ per pulse, retains its spatial Gaussian profile and is used as the main pump beam. This intense beam also drills a through hole at the aluminum foil terminating a semi-infinite gas cell (SIGC) filled with argon. The second beam, with a typical peak intensity which is only about 1.5% of the first beam (when material dispersion and broadening by achromatic pulse front delay during focusing are taken into account [35]), undergoes an amplitude modulation using two perpendicularly oriented polarizers and a phase-only SLM (Holoeye Pluto) in between. This beam acquires a spatial distribution of a ring which is then imaged and focused to form a Bessel beam [see Fig. 1(a)]. A half wave plate is used to set the polarization of the Bessel beam to be parallel to the polarization of the Gaussian beam. Both beams are combined using a holed mirror and are focused close to the output of the SIGC. The use of a holed mirror is possible, because the Bessel beam retain its ring shape before the focus. In the interaction region, the beams form a periodic modulation of the amplitude and phase according to Eq. (3). Modifying the position of both focusing lenses (LG and LB3 in Fig. 2) allows for a careful selection both of the location of the interaction region with respect to the exit hole and of the location around which the modulation is applied.

The Rayleigh range of the Gaussian beam that was used is 5 mm. The nondiffracting Bessel region $L_{ND}$ varies between 1.4 and 8.3 mm, while the periodicity of the induced periodic modulation varies from 400 to 860 $\mu$m (corresponding to $\theta$ in the range of 0.043–0.062 rad). Thus, the range of the number of periods in the modulation can vary between 2 and 21.

The HHG spectrum when only the Gaussian beam is present and focused to 1.5 mm before the exit hole of the SIGC is shown in Fig. 3 (labeled with “no Bessel”). In this case, the harmonic orders are not phase matched and therefore do not build up efficiently. The addition of a

![FIG. 2. Schematic representation of the experimental setup, where LB1–3 are lenses along the Bessel beam path, LG the Gaussian beam lens, BS the beam splitter, P1 and 2 polarizers, HWP a half wave plate, SLM the spatial light modulator, HM a holed mirror, and SIGC a semi-infinite gas cell. The delay stage allows us to compensate for the difference between the optical paths of the two beams, assuring the temporal coincidence of the two pulses at the interaction region.](image)

![FIG. 3. HHG enhancement: HHG spectra produced by a Gaussian beam only (black) and with a modulation caused by an addition of a copropagating Bessel beam establishing different $\Delta k_0$ values (other colors) with a fixed phase matching bandwidth of $\delta k = 0.83 \pm 0.16 \text{ mm}^{-1}$. Results are shown for 50 (a) and 100 Torr (b). Insets: Integrated intensity enhancement of each harmonic order for different $\Delta k_0$.](image)
Bessel beam, focused to 1 mm before the exit hole, with specific cone angles for generating modulations with the appropriate periodicity needed to phase match the harmonic orders under scrutiny, led to a substantial enhancement of these harmonics. The results of such enhancements are shown for two cases of ambient argon pressure, at 50 [Fig. 3(a)] and 100 Torr [Fig. 3(b)].

When only the Gaussian beam is present, the set of conditions at which the harmonic radiation is most efficient is a function of the time-dependent intensity: Higher intensity yields a stronger dipole moment; however, it also increases the ionization rate, which changes the phase mismatch. Following Eq. (5), with the addition of the perturbing Bessel beam, the applied periodic modulation which provides the highest enhancement of the harmonic radiation matches the actual phase mismatch. For example, at 50 Torr, by applying a modulation of $\Delta k_0 = 10 \text{ mm}^{-1}$, the strongest enhancement was observed for the 39th harmonic; therefore, we conclude that $\Delta k_{39} = 10 \text{ mm}^{-1}$. In effect, we are thus using the perturbation as an in situ probe for the actual phase mismatch of a given harmonic order [36]. Interestingly, by using Eq. (1), we find that this value of the phase mismatch corresponds to an ionization rate of 0.3, while the maximum ionization rate is 0.87 (see Supplemental Material [37]). This suggests that the best conditions for the generation of this harmonic order are at the leading edge of the pulse and not at its maximum, corroborating a recent theoretical prediction [43] related to the temporal dynamics of the phase mismatch [44–46]. Increasing the frequency of the modulation $\Delta k_0$, by modifying the radius of the ring on the SLM (for a fixed ring width, in which case the phase matching bandwidth is $\delta k = 0.83 \pm 0.16 \text{ mm}^{-1}$), we clearly observe a displacement of the band of phase matched harmonics to higher orders. This is easily seen for the integrated intensity enhancement shown in the insets in Fig. 3. The enhancement was calculated by dividing the integrated intensity of each harmonic order by the integrated intensity of the same harmonic order produced by the unperturbed Gaussian beam. The observed displacement is in accordance with the expected linear dependence of the phase mismatch on the harmonic order [see Eq. (1)] with the slope rising with the pressure. Overall, the most enhanced emission is scanned over ten harmonic orders, while an enhancement of up to 30 times is observed for this particular case of the focal position of both beams relative to the exit hole of the SIGC. The observed linear dependence of the modulation period on the most enhanced harmonic order validates that the enhancement mechanism observed here is due to QPM. Furthermore, we recall that the intensity of the perturbing Bessel beam is about 2 orders of magnitude lower than that of the Gaussian beam, and so the added intensity alone cannot account for any observed enhancement.

Next, at a pressure of 50 Torr, we modify the phase matching bandwidth $\delta k$ by changing the ring thickness on the SLM while keeping a constant value of $\Delta k_0 = 12 \text{ mm}^{-1}$. The results are shown in Fig. 4, with the integrated enhancement presented in the inset. Theoretically, according to the simplest QPM models [34], the phase matching bandwidth should grow linearly with $\delta k$. In the inset in Fig. 4, we can see this linear dependency quite clearly as long as $\delta k < 1 \text{ mm}^{-1}$. For higher values of $\delta k$, the length of the modulation becomes too small to observe any enhancement. Notably, for this set of measurements, the highest enhancement observed is 100-fold, obtained for the longest modulation length as expected. Notice also that this largest enhancement is achieved for the case at which the width of the ring on the SLM is the smallest; hence, the modulation depth is the weakest. This establishes that the geometric properties of the modulation (length and period of the modulation) are more important than the modulation depth. The set of measurements at 50 Torr was repeated while setting the region of QPM at different locations along the total interaction length, and the results were compared to a numerical model [37] further affirming the utility of our method.

Conclusions.—We have driven the process of HHG using a pump beam made of the superposition of a Gaussian and a tunable perturbing Bessel beam, establishing a periodic modulation along the interaction cell. This modulation is able to perform on-the-fly, tunable quasi-phase matching of the harmonic spectra—with a controlled band and bandwidth at different conditions of ambient gas pressure. The flexibility of this method and its relatively easy implementation would allow us to use more complicated beams in order to control further parameters of the emitted radiation. For example, the use of aperiodic modulations could allow for the phase matching of several different bandwidths [47]. It would also be very interesting to extend the scheme presented here to control other degrees of freedom of the harmonic emission, such as the polarization state [48] and orbital angular momentum [49,50]. Additionally, similar perturbed beams might be helpful to analyze in situ conditions (such as the ionization level, coherence length, etc.) at different locations during the process of HHG. Finally, the
all-optical technique presented here might also be applicable to perturbative nonlinear optical frequency conversion in nonlinear crystals [47,51].

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L. H. and L. L. contributed equally to this work.

*alonb@eng.tau.ac.il

[37] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.183902 for more information about the experimental setup and data analysis, which includes Refs. [38–42].