Anomalous phase matching through inter sub-cycle interference

Georgiy Shoulga* and Alon Bahabad

1Department of Physical Electronics, School of Electrical Engineering, Fleischman Faculty of Engineering, Tel-Aviv University, Tel-Aviv 69978, Israel

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Providing efficient nonlinear optical frequency conversion is essential for numerous applications. Fundamentally, dispersion limits the efficiency of such processes through mismatch of the phase velocities and the group velocities of the involved optical waves. We reveal a dispersion condition at which, surprisingly, phase and group velocity mismatch act together to provide efficient optical frequency conversion. We demonstrate this condition numerically for the process of high-harmonic generation. This finding could be valuable to the theory and practice of nonlinear optical frequency conversion in general.

Introduction. Optical frequency conversion is fundamental to many applicable nonlinear optical processes [1] such as second-harmonic generation, sum and difference frequency generation, supercontinuum generation, parametric light amplification, parametric down conversion, optical rectification for Thz generation, Raman conversion and high-harmonic generation (HHG). The efficiency of these processes can be severely limited by dispersion leading to two mismatch phenomenon. The first, is phase-mismatch (PM) - namely the difference between the phase velocities of the different frequency components involved in the process, a difference which can also be regarded as a momentum imbalance between the interacting photons [2]. The phase mismatch limits the interaction length to the coherence length which is the distance over which emissions can still be added constructively. The second mismatch mechanism is group-velocity-mismatch (GVM) which leads to the spreading of the generated field outside the temporal span of the pump pulse, reducing its intensity. However, GVM can also be treated as a delay mechanism, contributing to the phase of the emission, and as such can be used to alleviate the deleterious effect of the PM as we show here, when a condition we name Anomalous Phase Matching (APM) is satisfied. The word ”anomalous” indicates that APM mandates a non-zero phase mismatch value.

We chose to investigate APM for HHG [3, 4] as the latter supply a platform where we can study and compare generation of high harmonics under the APM conditions with other harmonics which are not phase matched for the same environmental setting. In HHG radiation is emitted in the form of high harmonics of the fundamental pump pulse extending to the x-ray part of the spectrum [5]. PM in HHG was extensively studied. It is known that with intense pump beams leading to high levels of ionization and thus plasma dispersion, it is impossible to impose phase matching for HHG [6]. In such cases it is beneficial to apply quasi-phase-matching (QPM) [2, 7] where a parameter of the interaction is modulated spatially, temporally or spatio-temporally [2, 8, 9] in order to bring back the emissions into phase over extended length scales as was demonstrated for HHG [10–22]. QPM becomes challenging as the coherence length becomes very short.

It is interesting that compared to the vast body of work dedicated to PM in HHG, very little attention has been given to GVM in this field. It was realized that for HHG driven with a long wavelength pump at pressures of at least hundreds of Torrs, GVM starts to be a significant limiting factor [5, 23]. This should also be the case for HHG in a pre-ionized medium [24, 25] (even at pressures of dozens of Torrs) due to large plasma dispersion. We also mention that an aperiodic QPM scheme was suggested as a mean to realize attosecond pulse shaping by utilizing GVM [26]. As we now show APM, working through inter sub-cycle interference, can be achieved for HHG through tuning of global parameters such as gas pressure and light intensity to realize efficient up-conversion over extended length scales. This allows to achieve efficient HHG in parameter regimes were it was previously thought to be very unlikely, without satisfying PM conditions and without applying any QPM technique.

Theory. The premise of our suggested condition is described using Fig.1 in which continuous (dot-dashed) lines represent the propagation of the pump (harmonic order q) in space-time. When there is phase mismatch but no GVM, the field of the q-th harmonic can be built constructively by adding contributions emitted along the propagation of the pump pulse (say from point A to C) only up to the coherence length \( l_{c,q} \) at which the emission at C is exactly out of phase with respect to the emission at A. With GVM, the emission in point C is interfering with the emission arriving from point B (and not with that from point A). If the time difference between points A and B is equal to an odd number of \( T_q/2 \) (half the period of oscillation of this harmonic order), the emission from point B would interfere constructively with the emission at point C, realizing the condition for Anomalous Phase Matching (APM). The word ”anomalous” is used as this condition requires a non-zero phase mismatch value. More generally, if we denote with \( \ell_q \) the distance after which GVM causes a time-delay of value \( T_q/2 \) between the pump and the nonlinear emission, then...
the condition for APM is: $l_g = m \cdot l_{c,q}$, with $m$ being an odd positive integer, describing the order of the APM process.

Numerical simulations. APM requires high enough dispersion so that the length scales associated with the ∆ω and $\tau_T$ in time are the result of interference of local emission with radiation delayed by GVM. This can be described in Fig.1 if the delay between points A and B as well as between C and G is $T_0/2$ and $l_g$ is multiplied by $q$. In this case, between points C and G the phase difference is $2\omega_0T_0/2 = 2\pi q$ - twice of that expected from harmonics generated at the single atom level and obeying the dynamical symmetry $E(t + T/2) = −E(t)$ [27]. Hence, once the propagation gets to $q \cdot l_g$ the dynamical symmetry due to APM is replaced with $E(t + T/4) = E(t) \cdot e^{i\phi_{PM}}$ which supports harmonic orders $q = 4p + 2\phi_{PM}/\pi$ (with $p \in \mathbb{N}$), where $\phi_{PM}$ is a term whose value is determined by the exact APM condition (that is - which frequency is exactly phase-matched). The significance of this observation is that within the phase-matching bandwidth of APM, the harmonics that would be phase-matched would be spaced $4\omega_0$ apart, while their exact position would depend on the exact phase matching conditions. This can be clearly seen in Fig.4(b) below.

To investigate the conditions for this effect further, we are helped by Figure 2 which shows graphical solutions for Eq.1. The solutions (indicated by white circles) are the intersection of the left hand side (solid lines) with the right hand side of of Eq.1. The left hand side has been evaluated for a constant ionization of $p = 0.006$ and for $m = 1$ while other parameters are taken from Table 1[B]. In addition, the refractive index was calculated using a known plasma-neutral atoms equation [28] while $\Delta v_g$ was obtained through the wave-vector $k$ given with [29](neglecting the nonlinear term):

$$k = \frac{2\pi}{\lambda} + \frac{2\pi P(1 - p) n(\lambda)}{\lambda} - PpN_{\text{atm}} r_e \lambda - \frac{\mu_{mn}^2 \lambda}{4\pi a^2},$$

where $n(\lambda)$ is the refractive index of neutral atoms at the fundamental wavelength $\lambda$, $\mu_{mn}$ is the root of the Bessel function, corresponding to the waveguide mode, $P, p, N_{\text{atm}}, r_e, a$ are the pressure in atmospheres, ionization fraction, number density at 1 atm, classical electron radius and the waveguide radius, respectively.

As one can see from Fig.2, a general behavior suggests the need for larger pressures (dispersion, in general) to satisfy APM for a lower order harmonic. In addition, noticing that for higher harmonic orders, smaller changes in pressure (or other parameter that influences dispersion) would bring other harmonic orders into APM we can conclude that the phase-matching bandwidth would be wider for higher harmonic orders.

Numerical simulations. APM requires high enough dispersion so that the length scales associated with the
FIG. 2. (Color online). Conditions for APM: graphic solutions (white circles) of Eq.1 (for \(m = 1\)): left hand side is shown by solid lines for \(q = 401, 501, 601\) and 701. The right hand side is shown by a dashed line. The solutions are marked by circles. The parameters used for the calculation are listed in Table I[B].

All cases were numerically investigated using a semi-classical one-dimensional numerical model of high harmonic generation. The propagation of the pump along the optical axis \(z\) is calculated using a known extreme nonlinear propagation equation which takes ionization dispersion into account [30] supplemented to account for neutral atoms dispersion as well. At each propagation step we solve a 1D time-dependent Schrödinger equation (TDSE) with absorbing boundary conditions using a Crank-Nicolson method [31]. For the TDSE solution we used a soft-core Coulomb potential \(V(x) = -2I_p/(1 + \gamma |x|)^{-1}\); \(\gamma = \sqrt{2I_p m_e/\hbar}\) (which is a generalization for a potential from [32]) with its ground state wave function \(\psi_0(x) = \sqrt{2\gamma/\pi} (1 + \gamma |x|) e^{-\gamma|x|}\).

The polarization in each step is calculated using the expectation value of the electron’s position along the direction of polarization of the electric field \(x\) multiplied by the electron’s charge \(e\) and the atomic density of the medium \(N\): \(P = Ne \langle \psi | x | \psi \rangle\). Finally, the emitted field is calculated by integrating the propagation equation \(\frac{\partial E(z,\tau)}{\partial z} = -\frac{2\pi}{c} \frac{\partial P}{\partial \tau}\) [30] using a 4th order Runge-Kutta method.

All three cases show the expected general behavior for APM: as a relevant parameter is changed to increase the dispersion a lower harmonic order gets phase-matched, if the dispersion parameter is varied over a wide enough range, then at some point the next APM order would be observed (in Fig.3 we observe the orders \(m = 3, 5, 7\)). Furthermore, following the discussion of Fig.2, indeed the range of matched harmonic orders is wider for higher APM harmonic orders (this is most obvious in Fig.3).

We can estimate the phase-mismatch or coherence...
length of the harmonics undergoing APM. Simply, the phase mismatch is approximately linear in the harmonic order \([35]\), so observing the coherence length for several low harmonic orders (observing half an oscillation of their spatial evolution) we can approximate the coherence length for different cases. For example, the coherence length of the 45th harmonic in model \([C]\) which undergoes APM (see supplemental) is around 5 \(\mu m\). The coherence length of harmonic orders undergoing APM for models \([A]\) and \([B]\) can be as short as 9 \(\mu m\) for \(q = 271\) in case \([A]\) (for a waveguide radius of 28 \(\mu m\)) and 102 \(\mu m\) for \(q = 501\) in case \([B]\) (for a pressure of 700 torr).

In Fig.4 we show the evolution of the spectrum and of the time-dependent intensity of harmonic orders which undergo APM for a case similar to that listed in Table I\([A]\) with a waveguide radius of 28 \(\mu m\) but with a longer pump pulse with FWHM duration of 40 fs. The temporal signal shown is the intensity of the HHG emission in the bandwidth between harmonic orders 263 and 293. It is clear that after propagating about 2.5 mm, the emission from adjacent half-cycles of the pump pulse converges and the phase-matched harmonic orders become spaced 4\(\omega_0\) apart, as was anticipated following the analysis of Fig.1.

Finally we consider the effect of increasing the pump pulse length without changing the peak intensity. This allows more emissions to be added constructively and we should expect that the APM spectrum would be further enhanced as well as that the phase-matched bandwidth would be decreased (phase-matched selectivity increased). This is clearly demonstrated when we use again model A, fix the waveguide parameter to 28 \(\mu m\) and vary the pulse duration without changing the peak intensity (see Fig.5.). In a way - where regular phase-matching becomes more prominent for longer propagation lengths, for APM both propagation length and pulse duration play a similar role, since the latter allows a larger number of sub-cycle interferences. Notice that the spacing of every 4th harmonic in the APM bandwidth is also quite clear in this figure.

In conclusion, we have shown numerically that Anomalous Phase Matching (APM) can occur due to the interplay of both phase mismatch and GVM, providing an over-all phase matching mechanism, with no need for employing external macroscopic perturbations as with Quasi Phase-Matching techniques. The numerical demonstration in this work was done for HHG, but APM should be valid for all optical frequency conversion processes where phase mismatch is an issue. Importantly APM can address large values of phase-mismatch (very short coherence lengths) and thus supply enhanced, phase matched and selective radiation in parameter regimes in which this was thought to be highly unlikely because either phase mismatch or GVM alone would have reduced the efficiency of the frequency conversion process and QPM would have been too challenging to implement.

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* Georgiy.Shoulga@gmail.com
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In this work we analyze three different cases of HHG setups that show the APM effect (see Table I). The general trends in all three systems are quite similar. The results of case [A] were given in the main text. Here we bring the results of cases [B] and [C]. In case [A] we changed the dispersion by varying the waveguide radius. In the other two models we change other parameters to vary the dispersion. In case [B] we vary the pressure of the gas medium (Fig.1) while in case [C] we use a preionized gas medium and we vary the peak intensity of the pump pulse (Fig.2). Parameters used in these models are listed in Table I. All models clearly show the APM effect.

![Fig. 1.](image1)

![Fig. 2.](image2)

![Fig. 3.](image3)

**TABLE I. Models parameters**

| Parameter                     | [A]     | [B]     | [C]     |
|-------------------------------|---------|---------|---------|
| Fund. wavelength \( \lambda_0 \) [nm] | 2000    | 2500    | 800     |
| Fund. pulse FWHM [fs]         | 20      | 20      | 20      |
| Peak intensity \( [10^{18} \text{ W/m}^2] \) | 2       | 5       | 2.2 to 6.5 |
| Medium (gas)                  | He      | He      | Ar⁺     |
| Medium pressure P [torr]      | 100     | 500 to 1000 | 50     |
| Medium temperature T [°C]     | 24      | 24      | 24      |
| Ionization probability \( p \) | \( 1 - |\langle \psi_0 | \psi \rangle |^2 \) | \( 1 - |\langle \psi_0 | \psi \rangle |^2 \) | \( 1 - |\langle \psi_0 | \psi \rangle |^2 \) |
| Absorption / Transmission     | CXRO database [1] | [2]     |         |
| Waveguide diameter a [µm]    | 14 to 35 | 1000    | No WG   |
| Propagation length L [mm]     | 5       | 50      | 5       |

The evolution of the energy of various harmonic orders from model [C] for various peak intensities of the fundamental pulse: (a) \( I_{\text{peak}} = 2.2 \times 10^{18} \text{ Wm}^{-2} \), (b) \( I_{\text{peak}} = 3.3 \times 10^{18} \text{ Wm}^{-2} \) and (c) \( I_{\text{peak}} = 3.8 \times 10^{18} \text{ Wm}^{-2} \). Harmonic orders that undergo APM are highlighted on a background of other harmonic orders.

The evolution of the energy of various harmonic orders as a function of propagation for case [C] is shown in Fig.3 for various peak intensities (which is used as the dispersion control parameter for this model). The harmonic orders which undergo APM are highlighted on a background of the evolution of all the other harmonic orders which are not phase matched. It is clear that the APM
harmonics are subject to coherent constructive interference, while the other harmonic orders show alternating constructive and destructive interference, characteristic of phase mismatch conditions. We have checked that other models ([A] and [B]) show similar behavior upon changing their respective dispersion control parameter (actually they show cleaner results as their dispersion control parameter does not change the yield of radiation in the single emitter level as happens when the pump intensity is varied).

* Georgiy.Shoulga@gmail.com

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