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# Phase-matched absorption-limited HHG using pre-ionization

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**Abstract:** We have discovered that ionization-induced defocusing is the main cause of inefficient high-order harmonic generation in long absorption-limited media. A novel method utilizing pre-ionization to overcome this bottleneck was validated both experimentally and with numerical simulations. © 2022 The Author(s)

High Harmonic Generation in rare gases as a source of ultrashort coherent extreme ultraviolet (XUV) pulses is increasingly popular, requiring novel approaches for enhancing its performance. Many works have focused on increasing the HHG efficiency by controlling the phase-matching via different methods, for example: manipulation of the laser wavefront [1] or mixing multiple gases in the target medium [2]. However, not many works have focused on impact of the laser non-linear propagation on the generation itself yet.

In this contribution, we first determine the impact of non-linear laser propagation that compromises HHG in long absorption-limited medium and then we present a novel method utilizing pre-ionization of the generating medium to solve this issue.

The ionization-induced defocusing influences substantially laser propagation in gaseous medium [3]. To illustrate this behavior in more detail, we have performed numerical simulations utilizing cylindrically symmetric unidirectional solver [4] for different IR intensities at the entrance of a 15-mm long krypton medium (Fig. 1a). Two regions can be quickly observed from these simulations. Region I, where intensity drops drastically due to defocusing and region II, where intensity stabilizes for the rest of the propagation.

During absorption-limited generation (where medium length is much longer than absorption length) XUV reabsorption plays a critical role, therefore Region II is the region, where most of the detected XUV originates from. Furthermore, a crucial condition of efficient generation in long media is to fulfil the phase-matching [5], i.e., to minimize the phase mismatch  $\Delta k = k_q - qk_1$  between the generated q-th harmonic and the laser wave. In the Region II, the laser intensity is too low to sufficiently ionize the medium. We illustrate this effect for the 19<sup>th</sup> harmonic order of 800-nm laser in krypton (Fig. 1b full lines). In these conditions, the  $\Delta k$  is around 3 mm<sup>-1</sup> and phase-matching is not achieved; hence the HHG is inefficient.

We consider the dominant processes for each wave, which is dispersion of neutral particles for both fields and the effect of plasma for the laser field. Using the knowledge of phase-mismatch, we can estimate the degree of ionization  $\eta$  as a function of atomic density  $\rho$  and required coherence length  $L_{coh}$

$$\eta = \frac{\omega_1 \epsilon_0 m_e}{e^2} \left( \Delta\chi \mp \frac{2\pi c}{q\omega_1 \rho L_{coh}} \right), \quad (1)$$

where  $\Delta\chi = \chi_1^{(n)} - \chi_q^{(n)}$  is the difference of electric susceptibilities of neutrals at laser and its q-th harmonic frequency (at standard temperature and pressure). The coherent length  $L_{coh} = \pi/|\Delta k|$  can be also considered as a length along which all microscopic emitters add constructively. We consider ideal phase-matching as a situation, where coherent length is at least the same length as the medium. Using (1), we can estimate the optimal degree of ionization for the case of 19<sup>th</sup> harmonic of 800 nm laser generated in krypton (Fig. 1c). Results of the laser propagation simulations with homogeneously pre-ionized medium to 8% (Fig. 1b, dashed lines) reveal dramatic decrease of phase mismatch of more than one order of magnitude, thus, enabling ideal phase-matched generation in region II.

To evaluate this method experimentally, we developed a special target employing a weak 15-mm long capillary discharge to achieve homogeneous pre-ionization across the medium. A capillary discharge driven by a RLC circuit was selected based on MHD simulations [6], which confirmed homogeneity of the plasma. The experiment was performed on the HHG Beamline at ELI Beamlines facility [7], utilizing 10 mJ, 40 fs, 798 nm Ti:Sapphire laser as a driver. The laser was focused by a 5 m mirror into the target and emerging harmonic radiation was characterized with a flat-field XUV spectrometer.

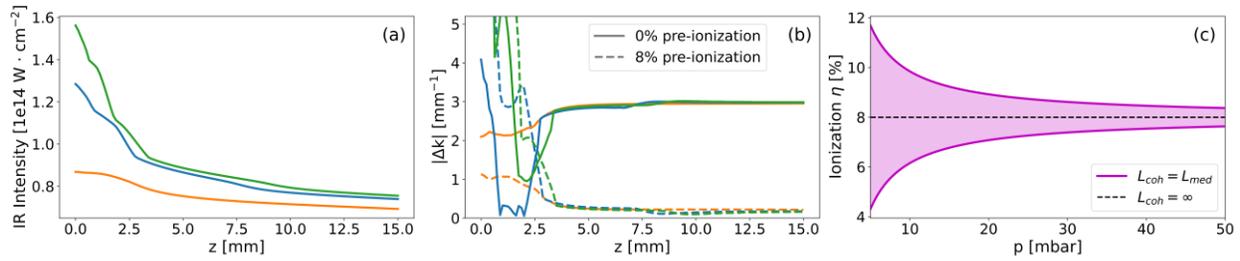


Fig. 1. (a) Evolution of on-axis intensity for different pulse intensities at the entrance of a 15-mm long medium with 36 mbar of Kr. (b) Comparison of phase-mismatch for the same pulses as in (a) and 19<sup>th</sup> harmonic for situation without pre-ionization (full lines) and with 8% pre-ionization (dashed lines). (c) Optimal degree of ionization for 19<sup>th</sup> harmonic generated by 800 nm laser in krypton as a function of pressure in 15-mm long medium.

Experimental results of 19<sup>th</sup> harmonic intensity as a function of gas pressure are presented in Fig. 2a. Varying the gas pressure modified the medium kinetics and resulted in different values of pre-ionization. The maximum XUV signal is reached at 35 mbar with pre-ionization, where signal is roughly 4 times stronger than without pre-ionization. Similar results (not shown here) were observed for other harmonic orders in Kr and Ar showing the broadband character of the method.

To simulate the experimental conditions, we inserted the laser field profiles from the unidirectional solver into the 1D time-dependent Schrödinger equation [8] to compute the quantum dipoles. The dipoles were used for a numerical XUV propagation in far-field analogical to [9]. The initial degree of ionization by the discharge was calculated from MHD simulations [6]. A significant signal enhancement can be observed, which agrees with the experimental results. Slight shift in the position of the maximum is probably caused by possible underestimation of degree of ionization in the MHD simulations of the Kr discharge.

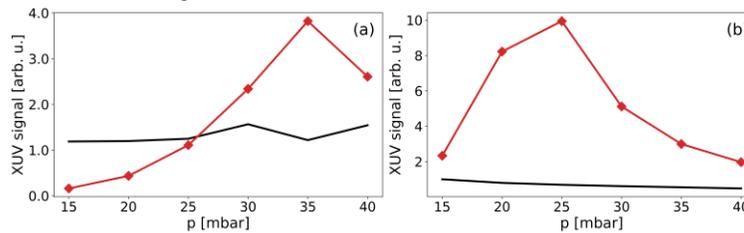


Fig. 2. Signal of the 19<sup>th</sup> harmonic in Kr without (black) and with 8% pre-ionization (red) (a). H19 signal from rigorous numerical simulations without (black) and with the pre-ionization (red) (b).

In summary, we have shown that HHG in long media is inefficient due to ionization induced defocusing and we proposed a novel method of controlling the phase-matching by pre-ionizing the medium. We have confirmed the ability of this method to increase the harmonic yield theoretically and experimentally. We believe the employment of our method will further enhance the applicability of ultrafast compact XUV sources for photon-hungry applications.

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