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Absorption-limited XUV generation in noble gases

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Abstract

We demonstrate absorption-limited high-order harmonic generation by an 800 nm femtosecond laser in argon and krypton. Phase-matching is achieved by pre-ionizing the gas using a weak capillary discharge.

High harmonic generation (HHG) in noble gases is nowadays an indispensable source for coherent extreme ultraviolet (XUV) radiation used in various fields of research fields [1]. Besides effects influencing the microscopic phenomenon of HHG, which have been intensively studied for almost 30 years [2], the overall XUV signal strength is mainly affected by the phase-mismatch between the driving IR laser pulse and the generated high harmonic radiation [3]. Thus, the effective target thickness for efficient XUV generation is usually limited by the coherence length $L_{coh} = \pi/|\Delta k|$, with phase-mismatch $\Delta k = k_q - qk_1$, and k_q and k_1 being the wavenumbers of q -th harmonic and fundamental, respectively. Here, we present a strategy to reduce the phase-mismatch Δk significantly and achieve absorption-limited HHG, that is, the effective target thickness is limited only by the absorption length $L_{abs} = 1/\sigma\rho$, given by the medium density ρ and the absorption cross-section σ for the XUV radiation in the target medium.

When the focused laser pulse enters the target chamber it is known that due to ionization-induced defocusing the laser intensity drops drastically along the direction of propagation, provided that the entry intensity is higher than the clamping value [4]. To illustrate such behavior, we have performed numerical simulations of an 800-nm laser pulse propagation in a $L_{med} = 15$ mm long gaseous krypton medium for different laser intensities and gas pressures. Figure 1a) clearly shows that the intensity drop (region I) is followed by a stable region with almost constant laser intensity (region II). The value of clamped intensity depends only on the medium type and density, and is practically independent of the laser intensity at the entrance of the medium.

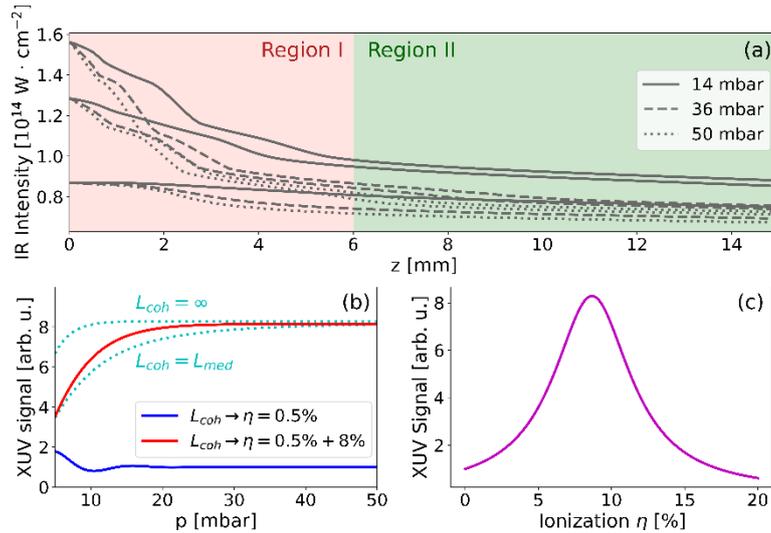


Fig. 1 (a) Results of numerical simulations of a 30 fs pulse propagating in krypton demonstrating the stabilization of the on-axis intensity in region II for different medium pressures and input laser intensities. In region I the intensity drops rapidly. (b) 17th harmonic signal versus gas pressure obtained from the analytical model of Ref. [5], when the optimal signal is in the range $L_{coh} > L_{med}$ (cyan dotted curves). The expected signal from region II shown in Fig. 1a) is denoted by the blue curve. By adding a constant pre-ionization of 8% to achieve phase-matching, the expected signal moves into desired region (red curve). (c) The absorption-limited XUV signal as function of ionization ratio η according to the same model of Ref. [5], which surprisingly does not depend on the medium density [6].

When effects of the dipole phase on the harmonic generation and the geometrical phase due to the spatial beam propagation are neglected, the phase-mismatch for the q -th harmonic can be expressed as

$$\Delta k = \frac{q\omega}{2c} \left(\Delta\chi - \eta \frac{\rho e^2}{\epsilon_0 m_e \omega^2} \right), \quad (1)$$

where $\Delta\chi = \chi_{1n}^{(a)} - \chi_{qn}^{(a)}$ is the difference of electric susceptibilities and η is the degree of ionization. Inversely, Eq. (1) can be used to compute the η required to achieve phase-matching, that is, $\Delta k \approx 0$. For the 17th harmonic in krypton as shown in Fig. 1 a degree of ionization of about 8% yields a sufficiently large coherence length.

In order to produce the desired 8% of ionization by the laser pulse itself, an intensity much higher than the clamping one is required. This is the very reason why phase-matching of the HHG process by self-ionization of the intense laser pulse is not possible: the clamping intensity is not tunable. Therefore, we chose to pre-ionize the gas to achieve phase-matching in the upcoming experiments. To this end, a 15 mm long glass capillary with a 1 mm inner diameter filled with Kr or Ar and hollow electrodes on both sides to pre-ionize the medium was placed in the laser focus (High Harmonic Beamline at the ELI Beamlines facility employing 40 fs pulses @ 798 nm with an energy of 10 mJ and 5 m focal length). Note that the inner diameter of such a short capillary was too large for the laser beam to be guided, so its propagation can be considered as free-space propagation. The electrical discharge was generated by a RLC circuit, where energy is stored in a capacitor bank and then discharged through a 100 Ω resistor into the capillary. The current pulse had a 45 ns rising edge followed by an exponential decay with a characteristic time of 0.5 μ s. The peak current were either 40 A or 50 A. MHD simulations [7] were used to find a current pulse that generates a discharge with the desired ionization degree.

The strength of the 17th harmonic signal measured in the experiment with Kr together with the estimate of degree of ionization (Fig. 4a dashed purple line) indicate that for low gas pressures (below 25 mbar), the ionization is too high for efficient HHG. For pressures higher than 25 mbar, the combined ionization from the discharge and the laser propagation falls exactly into the phase-matching region, leading to optimum phase-matched generation. In particular, the highest XUV signal of the 17th harmonic is seen at 35 mbar. Figure 2b summarizes the enhancement factor for the H15, H17, and H19 in krypton with 40 A current discharge as a function of pressure. Similarly, Fig. 2c represents the measurements with 50 A discharge in argon for the H17, H19, and H21.

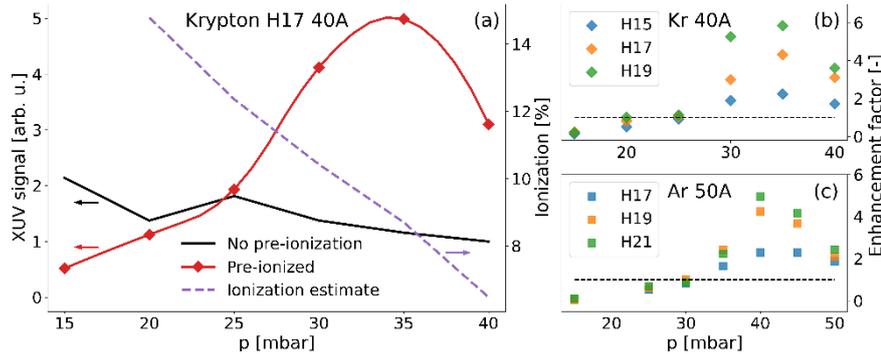


Fig. 2 (a) Experimentally measured intensity of the 17th harmonic order as a function of pressure for HHG in Kr without pre-ionization (black curve) and in pre-ionized medium with discharge peak currents of 40 A (red diamonds) [6]. Red line (spline) is added to guide the eye. Estimate of degree of ionization in the (purple dashed curve). (b) Enhancement factor for H15 (blue), H17 (orange), and H19 (green diamonds) in krypton with 40 A peak current. (c) Enhancement factor for H17 (blue), H19 (orange), and H21 (green squares) in argon with 50 A peak current. Each experimental point corresponds to the strongest shot from a set of ten laser shots.

In summary, we have demonstrated a new way of controlling the phase-matching of the HHG by pre-ionizing the generating medium. It increases the HHG conversion efficiency in the absorption-limited generation in long media. As the pre-ionization does not affect any other aspect of the generation, this mechanism turns out to be an independent tuning parameter of the experiment, paving the way for enabling photon-hungry applications with compact HHG sources.

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