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Observation of Third-Harmonic Saturation in Helium due to Ionization Depletion

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Abstract: We report the saturation of the third harmonic generated in Helium by ultrashort laser pulses, and describe the observed plateau based on the depletion of the electrons generating the harmonics in a restricted interaction volume. © 2022 The Author(s).

1. Introduction

In the last decades, the generation of coherent light into the extreme ultraviolet and x-ray range has experienced a fast development due to the potential applications. This radiation is usually generated by ultrashort pulses in gases inside vacuum chambers due to the strong absorption by solids and the atmosphere, and the main mechanism explored has been the High Harmonic Generation (HHG) [1]. In this method, the laser pulse ionizes the gas, and the freed electrons emit harmonics upon recombination with the parent atom [2]. More recently, the Below Threshold Harmonics (BTH) [3-5] have received attention for generating coherent light in the vacuum ultraviolet (VUV) to provide tools for ultrafast spectroscopy [6], frequency comb technology for precision measurements [7], and to boost HHG efficiency [8]. The BTH are generated by bound electrons [7], and their wavelengths are restricted to those shorter than the third harmonic of the laser, and longer than the energy associated with the ionization edge of the gas used, limited to 50.4 nm for Helium, which has the highest ionizing energy at 24.6 eV.

BTH are generated by the gas atoms valence electrons; we recently demonstrated [9] that, when the harmonics are generated in a gas in a limited region of space (such as a gas nozzle or a gas cell), laser-induced ionization depletes the BTH generating electrons, saturating the harmonic yield. This saturation can be understood considering a laser beam propagating in the z direction, focused on the origin $z = 0$, and gas filling the space from $z = -\ell/2$ to $z = +\ell/2$, as indicated by the shadowed region in Fig. 1. For the third harmonic, without loss of generality, the harmonic will be generated by all the atoms inside the volume limited by the isointensity surface defined by the third harmonic generation intensity threshold, $I_{3\omega}$, indicated by blue in Fig. 1 [9, 10], but its yield will be decreased if ionization happens. The ionization will occur inside the volume limited by the isointensity surface determined by the ionization intensity threshold, I_{ion}, indicated by red in Fig. 1.

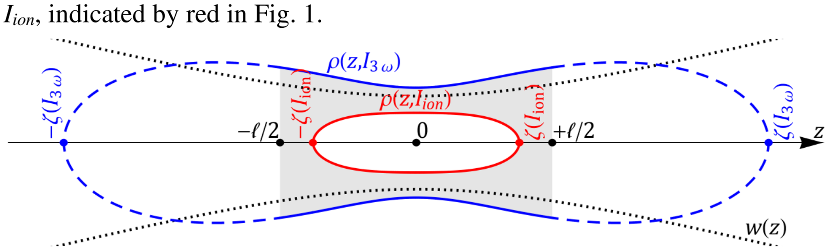

Fig. 1. Representation of the radius, ρ, of the third harmonic (blue) and ionization (red) thresholds isointensity surfaces, and the region with gas (shadowed), in which the third harmonic is generated. The points ζ mark the longitudinal limits of the surfaces, and the dashed lines indicate the region in which no harmonic is generated due to gas absence. The dotted lines indicate the laser beam $1/e^2$ radius, $w(z)$.

We showed [9] that, as the laser intensity, I_L, grows, the harmonic yield increases and the harmonic and the ionization isointensity surfaces get longer; when the ionization isointensity surface length equals the gas region length, $\pm\zeta(I_{ion}) = \pm\ell/2$, the harmonic emission is capped, creating a plateau, and cannot be increased by raising the laser intensity. This limitation occurs for a plateau intensity threshold, I_{p_th}, given by [9]:

$$I_{p\_th} = \frac{I_{ion}}{2}\left(1 + \frac{\ell^2}{b^2}\right), \quad (1)$$

where $b = (\pi\, w_0^2)/\lambda$ is the laser beam confocal parameter, with w_0 being the laser beamwaist, and λ its wavelength.

We demonstrated [9] that our theory correctly described the plateau intensity threshold for two noble gases, krypton and argon, but we could not observe the saturation for neon nor helium, since our setup could not reach the needed intensities. Here we show the observation of the plateau for helium, and correctly predict its intensity threshold.

2. Results

Ultrashort pulses from a CPA system (Femtolasers Femtopower Compact Pro HR/HP) were used to generate the third harmonic from helium flowing at 100 mbar through a nozzle inside a vacuum chamber. The pulses were centered at $\lambda = 785$ nm, and their duration could be varied from 25 fs up to ~200 fs. The laser pulses were focused to $w_0 = 14$ µm, and a short gas nozzle with length $\ell = 0.5$ mm was employed. The use of a tighter focusing and a shorter nozzle than in our previous setup [9] increased the laser intensity and lowered the plateau threshold intensity, according to Eq. (1), allowing us to reach it. Fig. 2 shows the experimental results for 3 different energies (colored dots), and the numerical calculation based on our theory [9] (black line), and the agreement is quite good. The orange vertical line indicates the plateau intensity threshold calculated by Eq. (1), which is $I_{p\_th} = 0.8\times10^{15}$ W/cm^2. The experimental data deviation from the theory curve (black) at low intensities is due to the small experimental signal under these conditions, and the plateau decrease at high intensities is due to the onset of other nonlinear phenomena not included in our model.

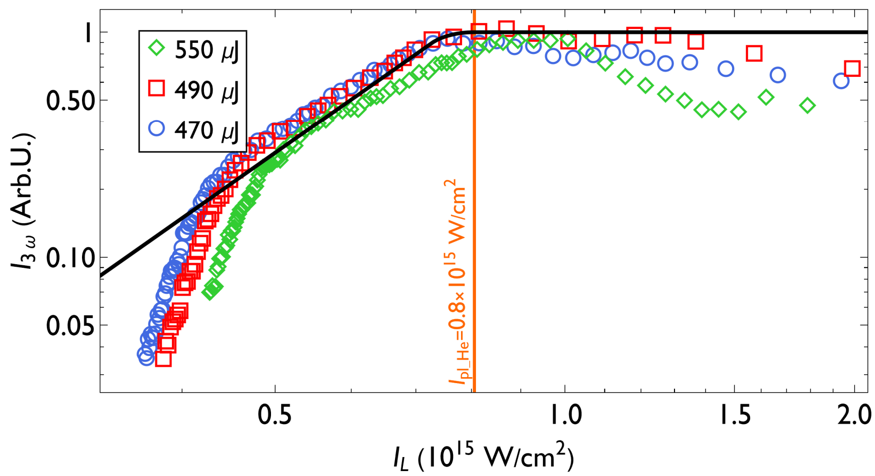

Fig. 2. Experimental results for the 3rd harmonic yield from helium (colored dots), and theoretical prediction for the experimental parameters used (black line).

3. Conclusions

We introduced a theory based on the depletion of electrons due to laser-induced ionization that explains the saturation of BTH emission, but could not observe it in helium [9] due to its high ionization energy. The theory predicted that tightening the laser focus and shortening the nozzle in which the harmonics are generated would lead to the harmonic yield saturation in this gas, which was experimentally observed and correctly quantified by the theory.

4. Acknowledgements

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5. References

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