Molecular Control of the Evolution of Capillary-Generated Soft X-ray High Harmonics

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Abstract: High harmonic generation from targets of Ar, N_2 and N_2O in a gas-filled capillary has been studied. A clear shift in the weighting of the harmonic intensity distribution with decreasing ionization energy is reported.

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1. Introduction

High harmonic generation (HHG) in gas-filled capillaries is an efficient method of producing a well-collimated, coherent, EUV/soft x-ray beam [1]. HHG is a complex process in which both single-atom generation and macroscopic phase matching must be understood. The majority of previous work has examined the HHG process from the noble gases with the gas jet geometry [2,3], with only limited numbers of studies employing a gas-filled capillary (see [4,5] for examples). HHG from molecules allows effects such as alignment and molecular symmetry on the HHG process to be examined in detail, as well as providing a new set of parameters for capillary phase matching. To date, experiments on HHG in molecules, such as H_2, O_2, N_2 and CO_2 [6,7] have been limited to the gas jet geometry. In this work we have extended these observations by studying and comparing the HHG from Ar, N_2 and N_2O in a gas-filled capillary. Theoretical predictions of the spectra are shown, which reproduce well the behavior seen in these gases.

2. Results and Discussion

The experimental set-up has been described in detail elsewhere [5]. 35 fs, 1 mJ laser pulses at a repetition rate of 1 kHz and central wavelength of 790 nm were focused to a spot of radius 50 μm and peak intensity of 2×10^{14} W cm^{-2} at the entrance of a fused silica capillary of radius 75 μm. The central, active region of the capillary is filled with the target gas through two 150 μm radius holes in the capillary wall. Following the capillary a 200 nm thick Al foil is used to block the laser beam while allowing 10% of the generated x-ray beam to be transmitted and subsequently detected on a grazing incidence spectrometer. Both the x-ray spectra from Ar, N_2 and N_2O and the corresponding spectra of the pump laser pulse after propagation through the capillary, are presented as a function of pressure in figure 1. Also plotted are the theoretical envelopes for the harmonics predicted from phase-matching calculations in which the radial variation of the laser intensity across the capillary mode is either averaged [8], or explicitly included.

At 15 mbar the x-ray spectra, figure 1(a), of Ar and N_2 are almost identical (except for a factor of three in intensity) with eight narrow, well-defined harmonic peaks up to an energy of ~50 eV. Both theoretical curves are similar and predict the correct maximum energy. The similarities in the Ar and N_2 x-ray spectra are mirrored in their near identical laser output spectra, figure 1(e). As the pressure is increased, the behaviour of Ar and N_2 diverges. The weighting of the harmonic intensities tends towards lower energies with decreasing ionization energy (IE), which becomes more apparent at 50 mbar, figure 1(c). Here the most intense harmonics generated from Ar (IE=15.7eV) and N_2 (IE=15.58eV) are concentrated around 35-50 eV and 20-30 eV respectively. The theory of the present work, including radial integration, provides a good description of these trends and predicts the highest observed harmonics for both gases accurately. Based on examination of individual terms in the calculations, the change in x-ray yield between Ar and N_2 is principally due to differences in the absorption of the target gas. The role of absorption in HHG will be discussed in greater detail at the conference. Increasing the pressure to 110 mbar, it is observed that the theoretical curves no longer model the x-ray spectra of Ar or N_2 well. In Ar, the theoretical envelopes underestimate the harmonic intensities. In the case of N_2 both theories predict harmonics with energies above 35 eV, although no significant x-ray signal was measured in this region. These discrepancies are probably due to nonlinear propagation of the pump laser pulse through the capillary arising from the high density of plasma generated. These effects become more significant as pressure is increased and are not accounted for in the theory. A further effect of increasing the gas pressure of Ar and N_2 is increased spectral broadening of the harmonic peaks, particularly for Ar.

This broadening is mirrored in the corresponding laser spectra for both gases, which are both blue-shifted and broadened by interaction with the plasma generated by the laser ionization, consistent with previous results [5].

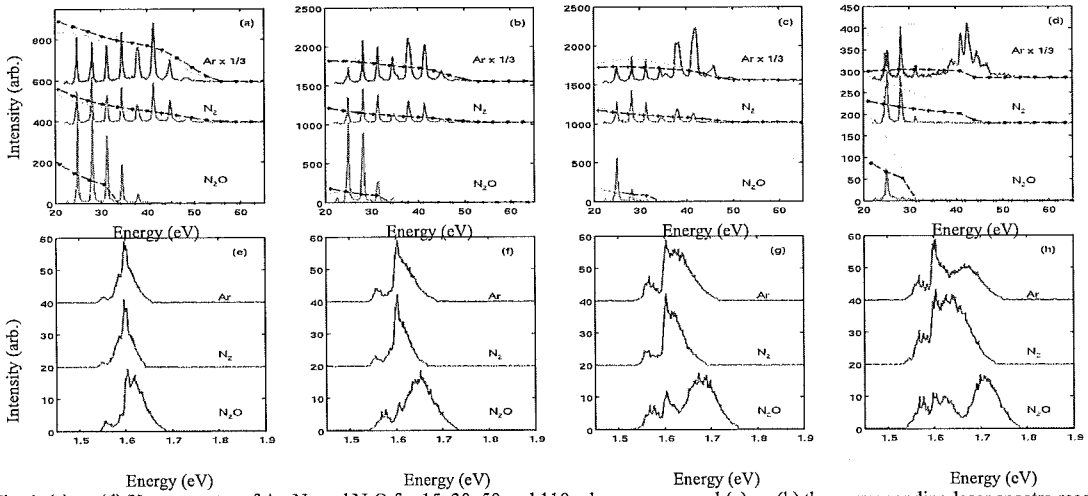

Fig. 1. (a) → (d) X-ray spectra of Ar, N_2 and N_2O for 15, 30, 50 and 110 mbar pressure and (e) → (h) the corresponding laser spectra recorded at the capillary exit under identical experimental conditions. The spectra are shifted vertically for clarity and the Ar spectra intensities are dived by 10. Averaged and radially integrated theoretical envelopes for harmonic intensities are given by (·· +) and (— *) respectively.

In N_2O, the high-energy harmonics are limited to <33eV by breakdown of phasematching due to above-barrier ionization producing very high plasma densities. However, at 15 and 30 mbar harmonics were observed slightly above this energy. The probable reason is due to the simplistic over-the-barrier model used in determination of the cut-off energy - although it is valid for atomic potentials, no allowance is made for possible subtleties present in a molecule. The harmonic peaks generated from N_2O display the least amount of broadening of all three gases, yet their corresponding laser spectra show the greatest amount of the three gases. This implies that the x-ray generation and broadening are occurring at different distances along the capillary, with Ar generating strongly at the end of the gas-filled region where the laser pulse has undergone the most broadening.

3. Conclusions

We have presented and compared HHG in Ar, N_2 and N_2O in a gas-filled capillary geometry. The spectral properties are dominated by phasematching considerations, and well described by a phasematching theory that includes the radial intensity variation of the laser. The spectral broadening of the pump laser at the capillary output has been measured, and used to interpret the distortion of the x-ray spectra seen at high gas pressures. Ongoing work aims to improve the theoretical calculations, and be able to determine the exact interactions between the spectral broadening of the laser and the generated high harmonic spectra and efficiencies.

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4. References

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