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Coherence Properties of High Order Harmonic Radiation

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Abstract. The results of a series of experiments to characterize the coherence properties of xuv radiation produced by high-order harmonic generation in helium, neon and argon are reported and compared to predictions from an effective order model. The harmonics exhibit smooth, near gaussian spatial profiles, and have a divergence that is approximately constant (< 12 mrad) in the plateau region and decreases (≈ 4 mrad) in the cutoff for $f/17$ focusing. For a bandwidth limited, 140 fsec incident pulse, we measure a harmonic line width of $\Delta\lambda/\lambda \approx 2 \times 10^{-3}$ at 30.3 nm. By reducing the spectral width of the driving pulse, harmonics with $\Delta\lambda/\lambda \approx 2 \times 10^{-4}$ can be produced. Absolute conversion efficiency as high as 10^{-7} for harmonic radiation as short as 20 nm has been achieved by using 400 fsec, 526 nm pulses from an Nd:Glass laser.

1. Introduction

The understanding of high order harmonic generation in rare gases by a very intense, short pulse laser has progressed significantly in the past few years. The detailed characterization of the generated harmonic fields, however, is only now being addressed. In particular, the spatial and temporal coherence of the high order harmonics in addition to the absolute conversion efficiency must be optimized for use in applications. Spatial profile measurements of high order harmonics produced by a 1 ps laser have been recently reported by Tisch et. al.¹, and L' Huillier et. al. reported measurements of the spatial profiles of the 13th harmonic of a 40 ps Nd:YAG laser.² Some work has also been reported on the spectral characteristics of the harmonics. In particular, the spectral blue shift of the harmonics due to the ionization and plasma formation have been reported by two groups.^{3,4} In this paper, we report on measurements of the absolute conversion efficiency, spatial distributions and the spectral line widths of high order harmonics generated in helium, neon and argon and compare these results to predictions from an effective order model. Measurements of coherent xuv radiation at 20 nm with a peak brightness on the order of 10^{24} photons/(mm²-mRad-sec-0.1% bandwidth) are presented.

2. Spatial Distribution Measurements

The harmonic spatial distribution experiments were conducted by generating harmonics of a short pulse Cr:LiSrAlF₆ (LiSAF) laser⁵ operating at 825 nm and capable of producing pulse energies in excess of 200 mJ. Special care has been taken to characterize the far-field focal profile of the laser which is approximately two-times diffraction limited with a 70 μm focal spot ($1/e^2$) using an f/17 focusing lens. For the spatial profile measurements the laser was focused at f/17 into a gas plume provided by a pulsed gas jet. The estimated atomic density for these experiments could be varied between 10^{17} atoms/cm³ and 10^{18} atoms/cm³. The measured divergence of the focused, 825 nm beam was 62 mrad ($1/e^2$). The harmonics produced were collected by a flat field spectrometer consisting of a flat, variable line spaced gold grating at grazing incidence and a single, flat, carbon folding mirror. A CsI coated dual micro-channel plate (MCP) detector was located at the image plane of the spectrometer, 59 cm from the source of the harmonics. A phosphorus screen coupled to the MCP detector was read by an 8-bit CCD camera yielding simultaneous spectral and spatial information on each laser shot.

The experiments were conducted by averaging 5 laser shots in a 10% energy window. For intensities below the ionization saturation intensity, the images are extremely reproducible on a shot to shot basis. In figure 1, the line outs of three harmonic profiles generated in argon at an intensity of 4.0×10^{14} W/cm² are shown. The 25th and the 29th harmonics represent harmonics in the plateau region of the spectrum and the 33rd harmonic is the highest order harmonic detectable in the cutoff with the MCP detector. The harmonic distributions exhibit near gaussian profiles with little or no substructure. The divergence of the harmonic radiation decreases slightly with increasing order in the plateau region and rapidly in the cutoff. The divergence for the harmonics in argon range from 12 mrad for the orders from the 17th to the 25th down to 3 mrad for the 33rd harmonic.

Figure 1: Integrated line outs of selected harmonics (25th, 29th and 33rd) produced in argon at a laser intensity of 4×10^{14} W/cm².

Harmonics produced in neon show a similar behavior as those produced in argon. Like harmonics produced in argon, the distributions are smooth and exhibit a divergence that decreases from around 13 mrad for the lower orders down to 6 mrad for harmonics at the cutoff. In general, we find that the harmonics exhibit a divergence that is less than that predicted by lowest order perturbation theory.⁶

This can be explained by considering the fact that the harmonic yields typically scale with the laser intensity with an order that is typically much less than the actual process order⁴. Under our experimental conditions, the confocal parameter, b , is much longer than the length of the generating medium (gas jet), L . In this limit ($b \gg L$), perturbation theory predicts that a harmonic with order q will have a divergence that is $1/\sqrt{q}$ times the laser divergence. If, however, the harmonic varies with an effective nonlinear order, p , then the divergence of the harmonic is $\sqrt{p/q}$ times the laser divergence. Since p is typically much smaller than q for the harmonics out in the plateau ($p = 5-8$) we would expect the higher order harmonics to exhibit a smaller divergence than predicted by perturbation theory. This prediction is consistent with our measurements throughout the rare gases provided the laser intensity is less than approximately twice the saturation intensity for ionization, I_{sat} .

When the peak laser intensity is greater than approximately twice I_{sat} , plasma formation occurs early in the pulse and refraction of the incident light produces structure on the harmonic spatial profiles and rapidly increases the divergence (along with the previously reported spectral blue shifting). Similar observations have been reported by Tisch, et al.¹ A detailed model accounting for these effects is currently in development.

3. Spectral Profile Measurements

The spectral distributions of selected high order harmonics produced by the Cr:LiSAF laser were conducted using a reimaging monochromator, (described in ref 4). The monochromator had an estimated resolution of .03 nm, and the spectral width of the laser for these experiments was 8.5 nm, $\Delta\lambda/\lambda = 1 \times 10^{-2}$. Profiles of the 27th harmonic in argon and neon produced at an incident intensity, near I_{sat} for each atom are shown in figures 2a and 2b respectively. Accounting for the resolution of the spectrometer we find that the fractional bandwidth of the harmonics was $\Delta\lambda/\lambda = 2 \times 10^{-3}$ in argon and was $\Delta\lambda/\lambda = 5 \times 10^{-3}$ in neon. This range of spectral line widths is comparable to the line widths predicted by perturbation theory which predicts a fractional bandwidth of $\Delta\lambda/\lambda = 2 \times 10^{-3}$ for the 27th harmonic.

As the intensity increases above the saturation intensity, the bandwidth of the harmonic also increases, similar to the increase in divergence. This can be explained self-phase modulation of the laser pulse created by the time-dependent refractive index experienced by the leading edge of the pulse resulting from ionization of the gas.⁴ At an intensity of $3 \times 10^{15} \text{ W/cm}^2$, the bandwidth of the 27th harmonic produced in neon increases to $\Delta\lambda/\lambda = 8 \times 10^{-3}$, well above the transform limited prediction of either perturbation theory or the effective order model which predicts that the normalized

spectral width of the harmonic should exhibit a $\sqrt{p}/q$ dependence on the fractional bandwidth of the laser, $(\Delta\lambda/\lambda)_q = \sqrt{p}/q (\Delta\lambda/\lambda)_{\text{laser}}$.

Figure 2: Spectral profile of the 27th harmonic produced near the saturation intensity in argon (a) and in neon (b).

4. Conversion Efficiency Measurements

The absolute energy yields of harmonics produced in helium and neon by a frequency doubled Nd:Glass laser were studied under conditions identical to those in which the spatial and spectral distributions were studied. The harmonic yields were measured using an absolutely calibrated X-ray CCD camera which can accurately determine the yield of incident photons over the spectral range 2 to 40 nm. The absolute yields of harmonics produced in helium with three different focal ratios are shown in figure 3 for a peak intensity of $6 \times 10^{15} \text{ W/cm}^2$. The harmonic yield increases linearly with the increasing confocal parameter, b obtained with weaker focusing. Energies in excess of 10 nJ can be generated for harmonics in the plateau. We have generated individual harmonics with an energy as high as 58 nJ at 25 nm with 500 mJ of green light. An optimum conversion efficiency of 2×10^{-7} was obtained at 25 nm (40 nJ produced) decreasing to 1×10^{-7} at 20 nm (25th harmonic) with 200 mJ incident at 526 nm and $f/70$ focusing into helium. Measured harmonic yields in the plateau in neon are very close to those values plotted in figure 3 for helium with the exception that the harmonics exhibit a cut-off at the 21st harmonic.

Figure 3: Absolute energy yield of harmonics produced in helium by 526 nm laser light at a peak intensity of $6 \times 10^{15} \text{ W/cm}^2$ for three focal ratios. Squares ($\square$) are for $f/25$ focusing, circles ($\circ$) for $f/50$ focusing, and diamonds ($\diamond$) are for $f/70$.

5. Conclusion

Measurements of the angular distributions, spectral line width and absolute conversion efficiency of high order harmonic radiation indicate a source peak brightness on the order of 10^{24} photons/(mm²-mrad-sec-0.1% bandwidth) at 20 nm produced by table-top subpicosecond lasers. We find that the angular distributions are smooth, near Gaussian distributions, without structure when the incident intensity is less than approximately $2xI_{\text{sat}}$. The harmonics exhibit a fractional line width of 2×10^{-3} for a 140 fsec laser ($\Delta\lambda/\lambda_{\text{laser}} = 1 \times 10^{-2}$). Measurements of the absolute harmonic conversion efficiency of a 600 fs, 526 nm laser pulse in neon and helium under similar conditions show that individual harmonics can be produced in excess of 20 nJ can be generated at 20 nm with a conversion efficiency $> 10^{-7}$.

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