

Generation of 35.5-nm coherent radiation

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Tunable coherent radiation was produced at 35.5 nm by seventh-harmonic conversion of 248-nm radiation from a krypton fluoride excimer laser. The nonlinear interaction took place at the intersection of the laser focus and a pulsed, supersonic helium gas jet. Third- and fifth-harmonic generation produced coherent outputs at 83 and 50 nm in both helium and xenon gas jets.

Thus far, harmonic generation and parametric mixing processes using high-power, ultraviolet laser sources have provided the only practical means of producing coherent radiation in the extreme-ultraviolet (XUV) region below 100 nm.¹⁻⁸ Several types of laser sources, including fixed-frequency Nd lasers^{1,2} and tunable dye lasers,^{3,4} have been upconverted to this wavelength region. The recent development of high-intensity, tunable excimer lasers with high spectral and spatial purity as well as ultrashort pulse width has been a particular stimulus in this area.⁵⁻⁸ We report the production of coherent radiation at 35.5 nm by seventh-harmonic conversion using a krypton fluoride (KrF) excimer laser tuned to 248.4 nm. To our knowledge, this is the shortest wavelength at which the production of coherent radiation has been reported in the open literature and is the first report of seventh-harmonic generation using an excimer laser. The third and fifth harmonics of the KrF laser were also generated at 82.8 and 49.7 nm, respectively.

The most important technical problems that must be addressed in XUV harmonic-generation experiments are the lack of a suitable transparent window material and self-absorption of the generated harmonic by the nonlinear medium. These problems have usually been overcome by employing a differential pumping system.^{1,4-8} A new method for confining the nonlinear medium was employed in these experiments; the nonlinear interaction took place at the intersection of the laser focus with an orthogonally directed, pulsed supersonic gas jet.⁹ Such an arrangement provides a localized region of high gas density in the vicinity of the nozzle orifice while substantially reducing the gas load on the pumping system. A related benefit is the reduced gas-consumption rate, which is an important consideration when costly rare gases are being used. The pulsed jet device used in these experiments is an automotive fuel injector modified for minimum gas pulse width, high vacuum compatibility, and high backing pressure.¹⁰ The device was tested to a maximum backing pressure of 7.76×10^3 Torr. Interchangeable nozzles with orifice diameters of 0.25 and

0.5 mm were used. The gas pulse duration was typically 500–1000 μ sec.

A schematic of the optical system is shown as Fig. 1. A 1-m focal-length, normal-incidence vacuum monochromator was modified to accept the pulsed jet. The entrance slit was removed, and the jet was installed with the nozzle tip approximately at the former position of the slit. The gas flowed orthogonally to the light-propagation direction and into a 150 liter/sec turbomolecular pump. No differential pumping aperture was interposed between the gas jet and the monochromator main chamber. Even at the highest backing pressures with gas pulse durations of up to 1 msec, the background pressure in the monochromator main chamber remained below 10^{-4} Torr. The monochromator was equipped with a 1200 line/mm gold-coated grating blazed at 80 nm. A windowless electron multiplier with CuBe dynodes was used to detect the XUV

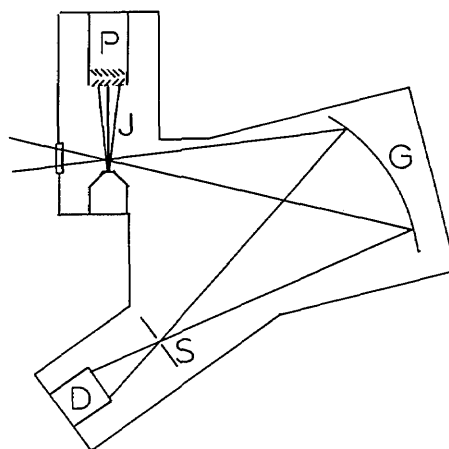


Fig. 1. Schematic diagram of the optical arrangement for the observation of XUV harmonic generation in a pulsed gas jet. J, gas jet; P, 150 liter/sec turbomolecular pump; G, 1200 line/mm gold-coated diffraction grating; S, monochromator exit slit; D, windowless electron multiplier tube with XUV filter.

radiation. Additional filtration of scattered laser light was necessary, and a 100-nm-thick aluminum or indium foil filter was placed in front of the detector. The KrF laser beam was focused ($f/10$) with a 100-mm focal-length quartz lens to a spot of approximately 10 μm in diameter, giving a peak intensity at the focus of approximately 10^{15} W/cm^2 . The position of the laser focus was adjusted to intersect the gas jet at a minimum distance of about 1 mm from the nozzle tip. The maximum densities obtained in the interaction region were estimated¹¹ to be in the 10^{17} - cm^{-3} range.

The laser system has been described in detail elsewhere,¹² so only a brief description is given here. The output of an amplified, mode-locked visible dye-laser system is first upconverted to 248 nm by using KDP crystals and is then spatially filtered. This signal is used as input to a single-stage, discharge-pumped, KrF excimer amplifier. At the final output of this system, 15-psec pulses are obtained with energies of up to 20 mJ/pulse. The value of 20 mJ/pulse is obtained at the peak of the KrF gain curve at 248.4 nm, whereas the full tuning range of the system is from 247.0 to 250.0 nm. Output-beam divergence is no more than 50% greater than the diffraction limit. The repetition rate of the system is 10 Hz.

The harmonic spectrum resulting from the interaction of the laser beam with a pure-helium jet is shown in Fig. 2. The backing pressure was 5.69×10^3 Torr, and the density in the interaction region was calculated¹¹ to be approximately 2×10^{17} cm^{-3} . This is a low-resolution scan taken with the exit slit width set to 2 mm, and the line shapes are solely determined by the monochromator optics. The third, fifth, and seventh harmonics appear in first order at 82.8, 49.7, and 35.5 nm, respectively. The seventh harmonic appears in second order at 71 nm. In Fig. 3 the relative dependence of the seventh-harmonic output on input laser energy is shown. No saturation is evident at even the highest intensities available. The scatter in the data arises from the extreme sensitivity of the conversion efficiency to small fluctuations in medium density.

The observation of the third harmonic in pure helium, which is positively dispersive for this process, is of some

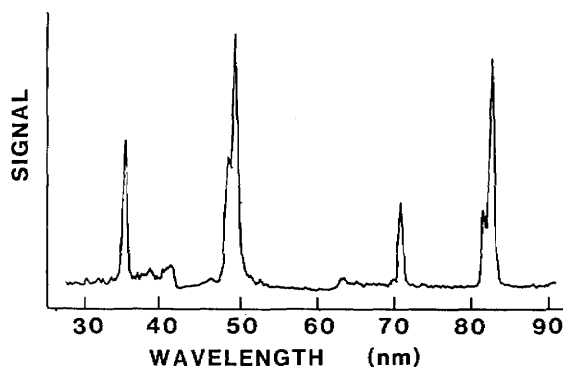


Fig. 2. Harmonic spectrum obtained using a helium gas jet showing third, fifth, and seventh harmonics. The peak at 71 nm is the second diffracted order of the seventh harmonic. The detector gain was reduced by a factor of several hundred between 42 and 63 nm.

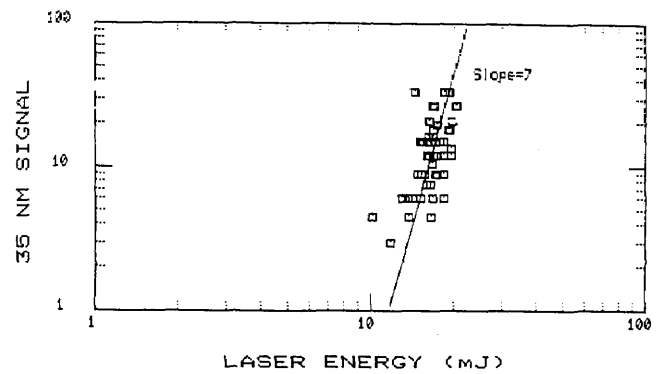


Fig. 3. Seventh-harmonic output in relative units as a function of input laser energy.

interest, since third-harmonic generation in a tight-focusing geometry (beam confocal parameter \ll length of nonlinear medium) and a positively dispersive isotropic medium is forbidden.¹³ In this experiment, the confocal parameter is 0.6 mm, the width of the gas jet at a distance of 1 mm from the nozzle is estimated to be 0.8 mm, and the tight-focusing approximation breaks down. Third-harmonic generation is allowed for both signs of dispersion in this case. Thus the pulsed jet permits a unique combination of extremely high laser intensity with a collimated-beam geometry.

Pure-xenon gas jets were studied as well. Xenon has a two-photon resonance at 249.6 nm, which has been exploited in previous experiments on third-harmonic generation using a KrF excimer laser.⁶ At the intensities used in this experiment, however, the resonance is expected to be completely saturated, and the principal problem is optical breakdown. We found that, by reducing the density in the jet to approximately 10^{16} cm^{-3} , we could essentially eliminate the breakdown. Under these conditions, third and fifth harmonics were observed, but no seventh harmonic was detected.

An order-of-magnitude estimate was made of the maximum output flux generated at 35.5 nm based on the typical values for filter transmission, CuBe photoelectric quantum efficiency, grating efficiency, and reflectivity of the gold coating given by Samson¹⁴ and the electron-multiplier gain given by the manufacturer. The multiplier gain was roughly consistent with the average size of individual dark counts. Approximately 100 photons/pulse were incident upon the photocathode of the detector, implying that 10^5 photons per pulse were actually generated. This corresponds to an energy-conversion efficiency of 3×10^{-11} . The maximum outputs occurred with the maximum helium density available. Based on previous XUV harmonic-generation experiments in helium, the output would be expected to increase as a high power of the density until the absorption length at the harmonic wavelength becomes comparable with the interaction length.¹ In helium at 35 nm, the absorption cross section¹⁵ is 4×10^{-18} cm^2 , and thus the density could be increased to $\sim 10^{19}$ cm^{-3} before this regime is reached, roughly 100 times the density used in this experiment. Such densities should be attainable with an improved jet nozzle design, substantially improving the conversion effi-

ciency. With comparable intensity, the maximum efficiency for seventh-harmonic conversion of 266-nm radiation to 38-nm was 10^{-7} to 10^{-8} at a helium density of $4.5 \times 10^{18} \text{ cm}^{-3}$.

In summary, tunable coherent radiation at 82.8, 49.7, and 35.5 nm was produced by third-, fifth-, and seventh-harmonic conversion of 248.4-nm radiation from a high-power KrF excimer laser. The nonlinear interaction took place in a pulsed, supersonic gas jet in a collimated-beam geometry. Significant output fluxes were obtained, and the output scaling relations with input laser intensity and medium density suggest that increases of several orders of magnitude may be achieved with relatively straightforward improvements in the apparatus.

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