Generation of high-power subpicosecond pulses at 155 nm

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Subpicosecond vacuum-ultraviolet radiation at 155 nm with pulse energies above 0.2 mJ has been obtained by near-resonant four-wave difference-frequency mixing in a Xe gas jet. Laser fields for the mixing process have been generated by a short-pulse KrF dye excimer laser system and a Raman converter. The process permits tuning in a broad vacuum-ultraviolet range and can be scaled up to higher output energies.

Frequency conversion of high-intensity laser radiation is a well-established technique for the generation of coherent vacuum-ultraviolet (VUV), extreme-ultraviolet (XUV), and soft-x-ray radiation. Currently, short-pulse high-power lasers in the IR, visible, and UV spectral ranges are used as primary lasers. With these laser systems wavelengths shorter than 10 nm have been obtained by high-order harmonic generation,[1] and high-order mixing has led to the generation of tunable XUV radiation.[2] However the output power and conversion efficiency of these high-order interactions are generally small. Therefore it seems to be advantageous to start from high-power laser systems operating at VUV wavelengths and generate coherent radiation below 100 nm by efficient low-order nonlinear processes, such as frequency tripling and four-wave mixing. The development of powerful laser sources at VUV wavelengths with gigawatt output power, however, has been demonstrated only for ArF excimer systems at 193 nm.[3–8] By frequency conversion techniques in metal vapors[9–11] and LiB_3O_5 crystals,[12] VUV peak powers up to 11 MW have been achieved.

Recent investigations[13–16] have shown that tunable subpicosecond VUV radiation can be efficiently generated by near-resonant four-wave difference-frequency mixing (FWDM) in Xe. The excitation of Xe by a short-pulse KrF hybrid dye excimer laser[17] (248.5 nm, 20 mJ, 400 fs) close to the $5p^6-5p^5 6p[1/2]_0$ two-photon resonance and additional injection of laser pulses with wavelengths between 330 and 1900 nm has resulted in short-pulse radiation in the range of 200–133 nm, with several microjoules of output energy.[14]

In this Letter we report on further progress to increase output power and conversion efficiency of the FWDM process in Xe. For experimental convenience experiments have been performed at the specific wavelength of 155 nm, produced according to $\omega_{155} = 2\omega_{248} - \omega_{626}$. Output energies above 200 μJ and peak powers in the range of 0.5 GW have been achieved. To our knowledge this is the first demonstration of a subpicosecond laser system in the mid-VUV spectral range operating at energy levels well above 100 μJ.

Efficient FWDM requires, in addition to the strong laser field at 248 nm, a powerful injection field that is synchronized to the KrF system. A straightforward approach would be to use the 497-nm radiation (100 μJ, 360 fs) generated internally in the KrF laser system.[17] Instead, we have frequency shifted this radiation by Raman conversion in H_2 to 626 nm (first Stokes line) to generate VUV radiation at a shorter wavelength and take advantage of a higher nonlinear susceptibility at 155 nm.[14] The experimental setup is shown in Fig. 1. Because Raman conversion of subpicosecond pulses becomes easily affected by other power-dependent processes, such as self-focusing and self-phase modulation,[4] the Raman converter is operated as a seeded Raman amplifier, which significantly improves the spectral and spatial quality of the Raman beam. Seed photons for the Raman amplifier are generated by a tunable dye laser (DCM in methanol) that is tuned to 626 nm. The dye laser is pumped by 1.7 mJ of a frequency-doubled YAG laser (Quanta-Ray DCR-11) and generates seed pulses at 626 nm with an energy of 240 μJ and a pulse duration of 8 ns. The YAG laser is synchronized to the KrF laser with a temporal jitter of ± 2 ns between the 497-nm and the 626-nm dye laser pulses. The 626-nm seed pulse and the 497-nm pulse are slightly noncollinear overlapped in the Raman cell, which permits separation of the seed and Stokes pulse at mirror M1. The Raman amplifier is operated at a H_2 pressure of 32 bars, just below the self-oscillation threshold. Pulse energies of $\sim$16 μJ have been obtained for the first Stokes line. A second dye cell pumped by 50 mJ of 532-nm radiation is used for further amplification of the Stokes signal in a double-pass configuration. The 626-nm output pulses from the dye amplifier (DCM in methanol) have a typical energy of 300 μJ and a pulse duration of 330 fs (sech^2 pulse shape). The spectral width of the amplified Stokes pulse is ~ 3.7 nm, resulting in a time–bandwidth product of $\Delta\nu\Delta t = 0.9$. Although the Raman converter generates pulses only at a fixed wavelength and, correspondingly, the VUV output of the FWDM process is not tunable; a set of VUV wavelengths is accessible by the use of different gases for the Raman cell.

Unlike in previous experiments,[13–16] a Xe jet is used instead of a gas cell for the mixing process to minimize absorption of the generated VUV radiation at 155 nm. Mixing experiments in a gas cell have demonstrated that VUV radiation at 155 nm is absorbed mainly by two-photon ionization of Xe and two-photon absorption at the exit window (MgF_2) of the gas cell. The VUV transmission of the exit window is further affected by color-center formation. For a MgF_2

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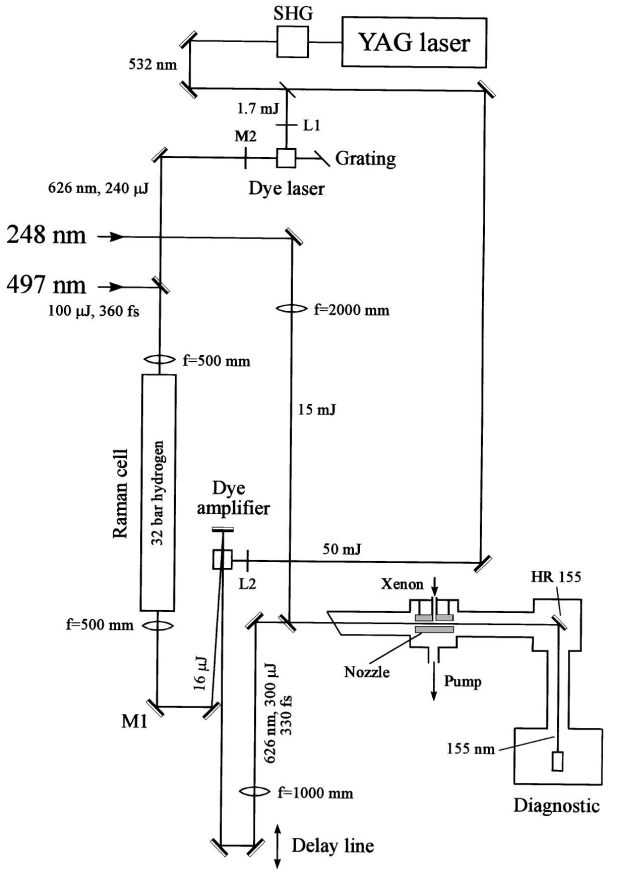

Fig. 1. Schematic layout of the laser system. L1, f = 80 mm cylindrical lens; L2, f = 35 mm cylindrical lens; M2, uncoated quartz plate; HR 155, dielectric mirror; SHG, second-harmonic generator.

window with a thickness of 2 mm a degradation of the transmission from 80% to 50% has been observed after several laser shots. Another advantage of the gas jet is the localized interaction region defined by the length of the nozzle (see Fig. 1). The input fields for the mixing process propagate without distortion to the interaction region, which further increases the conversion efficiency. A comparison of the VUV output between the gas jet and a gas cell (equipped with a new MgF_2 window) has yielded more than twice the energy for the gas jet. A vacuum of 10^{-3} mbars has been maintained within the propagation region of the input fields and the VUV radiation. The injection of Xe gas into the nozzle is controlled by a pulsed gas valve (General Valve Corporation). The valve is operated with a backing pressure of 5 bars and triggered approximately 700 μs before the lasers. Optimum VUV output has been obtained for a nozzle length of 5 cm and a diameter of 3 mm.

Phase matching of the FWDM process is accomplished by a noncollinear mixing geometry with an angle of 1° between the 248- and 626-nm beams. A Xe pressure of 54 mbars in a gas cell corresponds to an angle of 1° and can be used as an estimate of the pressure in the nozzle. The 248-nm beam with a pulse energy of 15 mJ is focused by a 2-m focal-length lens into the nozzle. A 1-m focal-length lens is used for focusing of the 626-nm beam. The generated VUV radiation is separated by a 155-nm high-reflecting dielectric mirror from the input fields.

Figure 2 shows a spectrum of the generated VUV radiation. Because of the multishot recording technique, however, no information can be extracted about the spectral structure. The spectrum suggests an upper limit of ~1 nm (FWHM) for the spectral bandwidth. Although no pulse duration measurement at 155 nm has been performed yet, autocorrelation of 193 nm radiation generated by FWDM in Xe (Ref. 14) has indicated comparable pulse durations between the FWDM signal and the input pulses. This is further supported by a recent pulse duration measurement of 147-nm radiation generated by four-wave parametric oscillation in Xe.[18] If we assume a Fourier-limited VUV pulse with a duration of 400 fs and a sech^2 pulse shape, we can calculate a minimum bandwidth of 0.06 nm.

A maximum output energy of 195 μJ at 155 nm has been measured by a pyroelectric energy detector, calibrated at 200 nm. Because the spectral sensitivity of pyroelectric detectors tends to decrease at wavelengths below 200 nm, the actual pulse energy is higher. The loss of sensitivity of the pyroelectric detector has been estimated in the following way. According to Manley–Rowe relations,[16] the FWDM process amplifies the 626-nm pulse. By measuring the gain in energy of the 626-nm pulse after FWDM, we can calculate the number of generated 626-nm photons and in turn the number of generated photons at 155 nm. This gives an estimate of the generated VUV energy, if we assume no significant absorption of the VUV radiation within the nozzle. If we compare this value with the measured VUV energy, we obtain a drop in sensitivity at 155 nm of ~25% compared with the sensitivity at wavelengths above 200 nm. The corrected maximum output energy would be 260 μJ, which corresponds to a peak power of 650 MW for a pulse duration of 400 fs.

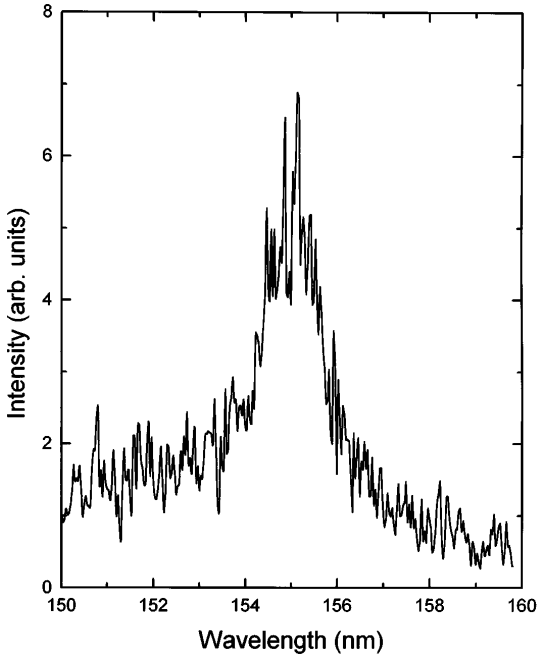

Fig. 2. Multishot spectrum of FWDM radiation at 155 nm.

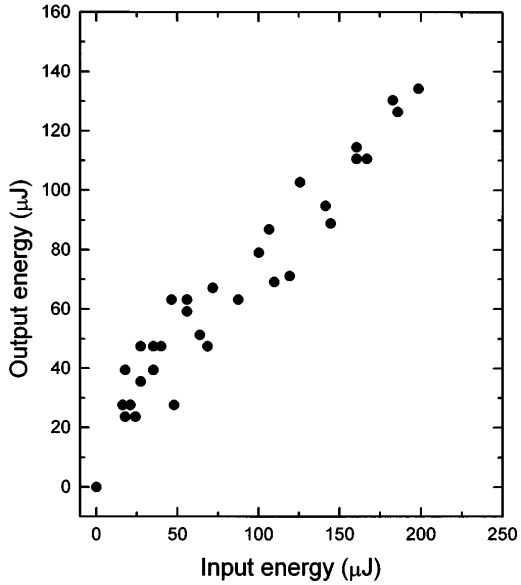

Fig. 3. Output energy at 155 nm versus input energy at 626 nm (detector calibration at 200 nm).

As indicated in Fig. 3, the energy dependence between the 155- and 626-nm radiation of the present setup shows no saturation. Therefore even higher VUV output energies can be expected at higher input energies at 626 nm. This can be achieved by a more powerful YAG laser for the dye amplifier.

In conclusion, we have developed a high-power laser system at 155 nm based on four-wave difference-frequency mixing in Xe. The system generates subpicosecond pulses with an energy of more than 200 μJ and can be scaled up to higher VUV energies. Experiments are now in progress to replace the Raman converter by an optical parametric generator to introduce continuous tunability of the VUV radiation. With further improvements the mixing process has the potential of providing tunable VUV powers in excess of 1 GW.

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