

# Generation of high-brightness coherent radiation in the vacuum ultraviolet by four-wave parametric oscillation in mercury vapor

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Coherent radiation at several wavelengths in the vacuum ultraviolet (VUV) has been generated by four-wave parametric oscillation in mercury vapor. When a powerful ultraviolet pump laser of frequency  $\omega_p$  is tuned to a two-photon resonance, VUV signal photons at frequency  $\omega_s$ , as well as idler photons at frequency  $\omega_i$ , are generated such that  $\omega_s + \omega_i = 2\omega_p$ . A frequency-doubled dye laser tuned to two-photon resonance with the  $6s6d\ ^1D_2$  level near 280.3 nm produced output at 184.9 and 143.5 nm. A tunable krypton fluoride excimer laser tuned to two-photon resonance with the  $6s10s\ ^1S_0$  level near 248.7 nm produced output at 140.1, 130.7, 130.1, 125.9, and 125.0 nm. Approximately 1- $\mu$ J, 200-W pulses were observed at 143.5 and 125.9 nm.

Third-harmonic generation and four-wave sum-frequency mixing in atomic vapors have been used extensively for the production of coherent radiation in the vacuum-ultraviolet (VUV) region of the spectrum.<sup>1</sup> These nonlinear processes involve the combination of three input frequencies to produce output at the sum frequency. When fundamental radiation derived from visible- and near-ultraviolet-wavelength laser sources has been used, such mechanisms have provided the means for the generation of tunable coherent radiation in the region below 200 nm. However, with the availability of powerful laser sources in the 200–300-nm range, another type of four-wave interaction may be considered for the generation of coherent VUV radiation, namely, four-wave parametric oscillation (FWPO). In FWPO, pairs of optical waves at frequencies  $\omega_s$  and  $\omega_i$  are generated on irradiation of an appropriate nonlinear medium by an intense pump wave at frequency  $\omega_p$  such that  $\omega_s + \omega_i = 2\omega_p$ .

In atomic vapors, resonance enhancements of the nonlinear susceptibility are generally exploited. Thus  $2\omega_p$  is tuned to a two-photon resonance while  $\omega_i$  and  $\omega_s$  are generated such that  $2\omega_p - \omega_i$  is near a hyper-Raman resonance. This type of resonantly enhanced FWPO leading to up-conversion of visible radiation to the near ultraviolet has been observed by several groups.<sup>2–6</sup> Because of the resonance enhancements of the nonlinear susceptibility, the conversion efficiency of pump photons to signal photons can be quite high; up to 5% conversion of 579-nm pump radiation to 330-nm output radiation was observed by Hartig<sup>5</sup> in sodium vapor.

We have performed two experiments in which FWPO has been used for the first time to our knowledge to generate coherent radiation in the VUV. In the first case, outputs at 184.9 and 143.5 nm were produced in mercury vapor by using 280-nm pump radiation. The tunable pump radiation was produced by frequency doubling the output of a Nd:YAG-pumped dye laser in a KDP crystal. Pulses of up to 20 mJ at 280 nm, with a 4–5-nsec duration and 0.3-cm<sup>-1</sup> bandwidth, were available at a repetition rate of 10 pulses per second.

The pump radiation was focused by a 250-mm focal-length lens into a 450-mm-length cell containing mercury at a density of  $10^{16}$ – $10^{17}$  cm<sup>-3</sup> and helium buffer gas at a pressure of up to 300 Torr. Peak intensities in the focal region were not measured but are estimated to be between  $10^{10}$ – $10^{11}$  W/cm<sup>2</sup>. When the pump radiation was tuned to two-photon resonance with the  $6s^2\ ^1S_0$ – $6s6d\ ^1D_2$  transition in mercury at 280.3 nm, strong coherent emission at 143.5 and 184.9 nm was observed, corresponding to additional resonance with the  $6s7p\ ^3P_1$  and  $6s6p\ ^1P_1$  levels, respectively [see Fig. 1(a)]. The VUV wavelengths were measured with a 0.3-m focal-length scanning monochromator to an accuracy of 0.2 nm. The relative magnitude of the output at each wavelength was also measured by using the monochromator.

Other characteristics of the output were determined

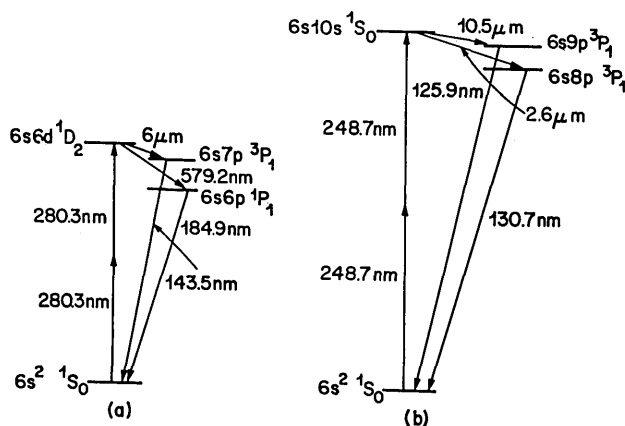


Fig. 1. Partial energy-level diagram of mercury showing resonances involved in generation of coherent VUV radiation by four-wave parametric oscillation. (a) Generation of 184.9- and 143.5-nm radiation by pumping with a frequency-doubled dye laser at 280.3 nm. (b) Generation of 125.9- and 130.7-nm radiation by pumping with a tunable krypton fluoride excimer laser at 248.7 nm. In this configuration, output was also observed at 140.1, 130.1, and 125.0 nm (see text).

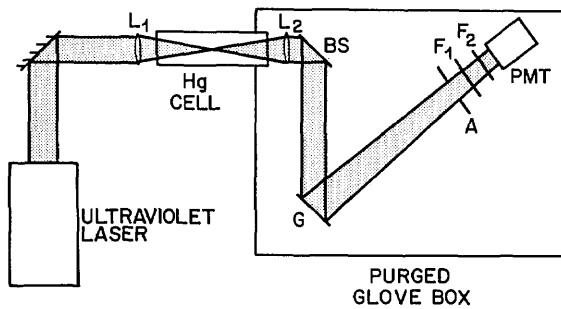


Fig. 2. Schematic diagram of experimental setup used to generate and characterize the VUV outputs. The ultraviolet laser represents either the frequency-doubled dye laser or the tunable krypton fluoride excimer laser. L1, 250-mm focal-length quartz lens; L2, 400-mm focal-length calcium fluoride lens or lithium fluoride lens; BS,  $\sim 10\%$  reflecting beam splitter; G, 1200-groove/mm grating; A, variable aperture; F<sub>1</sub>, F<sub>2</sub>, calibrated VUV filters; PMT, solar-blind photomultiplier tube.

by using a nitrogen-purged glove-box arrangement, as shown in Fig. 2. The VUV outputs were found to be highly directional and were generated in the forward direction only. Strong directional emission at 579 nm, the idler wavelength associated with the 184.9-nm signal, was observed in both the forward and backward directions. No attempt was made to detect 6- $\mu\text{m}$  radiation, which is the idler wavelength associated with the 143.5-nm signal. Measurements of the absolute VUV output pulse energies were obtained by using the optical system shown in Fig. 2. Emission from the cell was recollimated by a 400-mm focal-length calcium fluoride lens L2, then split by a flat, uncoated calcium fluoride beam splitter BS and directed onto a 1200-groove/mm grating G, blazed for 120 nm in the first order. The dispersed radiation was attenuated by using calibrated VUV filters and detected by a solar-blind photomultiplier tube. The beam splitter BS was used to attenuate both the signal and the pump radiation, preventing optical damage to the grating by the strong pump. Our purging system was imperfect, and residual absorption in the VUV optical paths was always present. This absorption was accounted for by translating the phototube over a measured distance while noting the change in signal and then extrapolating to obtain the total absorption over the full path length using Beer's law. With this adjustment, and including the absorption loss at L2 and the reflectivity of BS and using manufacturer's data for the grating efficiency,<sup>7</sup> phototube quantum efficiency, and gain,<sup>8</sup> we inferred an output energy of 1  $\mu\text{J}$  at 143.5 nm with an estimated uncertainty of a factor of 3. Output at 184.9 nm was a factor of 3 smaller than that at 143.5 nm, with a relative uncertainty of 50%.

The VUV output was found to be relatively insensitive to the helium buffer-gas pressure over the range of 5–100 Torr. Above 100 Torr, the output began to decrease, and it was undetectable above 250 Torr. As is shown in Fig. 3, the output at 143.5 nm increased approximately as the square of the mercury density until a value of 0.1 Torr was reached, at which point the output leveled off to a constant value. Tuning of the

pump radiation was found to be critical; with a detuning of more than  $\pm 0.5 \text{ cm}^{-1}$ , the signal was completely extinguished.

We have also used a tunable krypton fluoride excimer laser to observe FWPO in mercury vapor. In this case, output at several discrete wavelengths from 125.0 to 140.1 nm was generated by using 249-nm pump radiation. The tunable excimer-laser system is based on the design of Hawkins *et al.*<sup>9</sup> A tunable dye laser operating near 498 nm is frequency doubled by a temperature-tuned ammonium dihydrogen phosphate (ADP) crystal. The second-harmonic radiation is then amplified in a single pass through a krypton fluoride discharge device (Lambda Physics model EMG 200). In this way, tunability over the krypton fluoride gain band from 247 to 250 nm may be obtained. In our case, approximately 100 mJ of radiation per pulse were produced at the peak of the krypton fluoride gain band (248 nm) with 100  $\mu\text{J}$  per pulse of input second-harmonic radiation. The output bandwidth was approximately  $0.4 \text{ cm}^{-1}$ , the pulse duration was 5–7 nsec, and the pulse repetition rate was 10 pulses per second.

When this laser system was used as the pump source for FWPO in mercury by tuning it to the  $6s^2 \ ^1S_0$ – $6s10s \ ^1S_0$  two-photon resonance at 248.7 nm, strong coherent emission at 125.0, 125.9, 130.1, 130.7, and 140.1 nm was observed, corresponding to resonance with the  $6s9p \ ^1P_1$ ,  $6s9p \ ^3P_1$ ,  $6s8p \ ^1P_1$ ,  $6s8p \ ^3P_1$ , and  $6s7p \ ^1P_1$  levels, respectively [see Fig. 1(b)]. By using the optical system shown in Fig. 2, but substituting lithium fluoride optics for the calcium fluoride optics, an output energy of 1  $\mu\text{J}$  per pulse was measured at 125.9 nm. The measurement uncertainty is again a factor of 3. Output at each of the other wavelengths was about an order of magnitude smaller. As in the first experiment, no attempt was made to detect the infrared and far-infrared idler wavelengths. The dependence of the 125.9-nm output on helium buffer and mercury densities, as well as on pump detuning, was found to be similar to that found for the 143.5-nm output described above.

It is somewhat surprising that triplet intermediate states lead to the dominant output signals. However, spin-orbit mixing is well known to be strong in mercury.

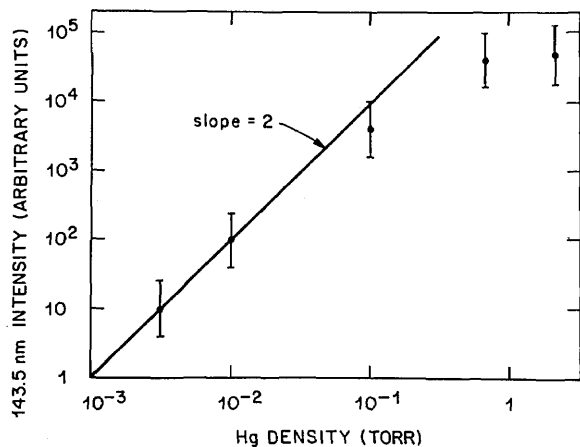


Fig. 3. Dependence of 143.5-nm output signal on Hg density.

Recent measurements<sup>10</sup> have shown that the Einstein  $A$  coefficient for the intercombination line  $6s6d\ ^1D_2-6s6p\ ^3P_1$  is actually 4.1 times larger than that for the  $6s6d\ ^1D_2-6s6p\ ^1P_1$  transition. This provides a qualitative explanation for the dominance of intercombination pathways in our experiments.

In the course of these experiments, we have also observed second-harmonic generation (SHG) of VUV radiation in mercury. Although collinear SHG is strictly forbidden for plane-wave propagation in isotropic media,<sup>1,11</sup> Freeman *et al.*<sup>12</sup> have recently demonstrated that SHG may be observed by using a focused single beam in an isotropic vapor. With our pump laser tuned near the  $6s6d\ ^1D_2$  level in Hg, tunable radiation at the second harmonic near 140 nm was observed. This signal was strongly peaked at the resonance but was observable at pump-frequency detunings as large as  $100\text{ cm}^{-1}$ , with the output wavelength appropriately tracking with pump-laser tuning. At its peak, this signal was about a factor of 10 smaller than the 143.5-nm FWPO signal. When the pump laser was tuned near the  $6s10s\ ^1S_0$  level in Hg, no SHG could be observed. This restriction of the observation of resonantly enhanced SHG to states of angular momentum greater than 1 is in agreement with the observations of Freeman *et al.*<sup>13</sup>

In summary, we have observed the generation of coherent radiation at several wavelengths in the vacuum-ultraviolet region by four-wave parametric oscillation in mercury vapor. Approximately  $1\text{-}\mu\text{J}$ , 200-W pulses were obtained at 143.5 nm using 10-mJ, 2-MW input pulses at 280.3 nm for an overall conversion efficiency of  $10^{-4}$ . Approximately  $1\text{-}\mu\text{J}$ , 200-W pulses were obtained at 125.9 nm using 100-mJ, 20-MW pulses at 248.7 nm for an overall conversion efficiency of  $10^{-5}$ .

These radiation levels are of immediate utility in such applications as photolithography, photochemistry, spectroscopy and interferometric diagnostics of high-density plasmas.

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