Generation of high-spectral-brightness tunable XUV radiation at 83 nm

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High-spectral-brightness coherent XUV radiation has been produced by third-harmonic generation of a transform-limited-bandwidth KrF* laser in gaseous xenon. The observed XUV output, which was continuously tunable from 82.8 to 83.3 nm, had a peak power of 40 mW, a bandwidth <0.01 cm^{-1}, and absolute frequency control to within 0.04 cm^{-1}. The utility of this XUV source for high-resolution spectroscopic applications is demonstrated by absorption studies in molecular hydrogen.

The rapid development of rare-gas–halogen excimer lasers is permitting the detailed study of many nonlinear processes in the ultraviolet region. These include high-resolution multiquantum spectroscopy of high-lying atomic and molecular states,[1–3] state-selective collisional processes,[1,2] isotopically selective mechanisms involving excited molecular electronic levels,[3] and third-harmonic generation.[4] In this Letter, we report new results on third-harmonic conversion of excimer-laser radiation. The excimer-laser systems provide highly attractive fundamental sources for frequency conversion to the XUV because of their demonstrated ability to deliver tunable, very-high-spectral-brightness radiation in the ultraviolet region. This ability was clearly demonstrated by the early work of Hutchinson *et al.*,[5] who produced tunable, coherent radiation at ~57 nm by frequency tripling a xenon excimer laser.

The shortest-wavelength coherent radiation produced to date is at 38 nm, the 28th harmonic of the Nd:YAG laser.[6] That result was obtained in a cascade process by first generating the fourth harmonic (266.1 nm) of the Nd:YAG laser in two stages of frequency doubling and then directly generating the seventh harmonic of the 266.1-nm radiation in gaseous helium. The earlier work[6] stressed the need for new, powerful, narrow-bandwidth pump sources in the ultraviolet region for frequency conversion to the XUV.

Recently, a tunable, narrow-bandwidth KrF* laser was developed that exhibits performance parameters closely approaching the fundamental limits governing spectral linewidth, beam divergence, and absolute wavelength control.[7] This instrument combines the property of continuous tunability over the full KrF* gain profile with the following experimentally established characteristics: pulse energy ~60 mJ, pulse duration ~10 nsec, spectral linewidth 150 ± 30 MHz, absolute frequency control to within 300 MHz, and beam divergence ~50 μrad.

In this Letter, we report the generation of high-spectral-brightness, coherent, and continuously tunable XUV radiation in the vicinity of 83 nm by two-photon resonant frequency tripling in gaseous xenon. The experimental apparatus is shown schematically in Fig. 1. The ultraviolet laser radiation is focused by a 10-cm focal-length lens into a 350-μm-diameter pinhole, through which xenon flows from the tripling cell to the differentially pumped chamber. The generated third-harmonic radiation is dispersed by a 1-m scanning vacuum-ultraviolet monochromator (MacPherson model 225) and is detected with a windowless electron multiplier (EMI model 9603/2B), which has a quantum efficiency of 17% at 83 nm and less than 0.01% at 250 nm. This configuration is similar to that used in previous studies.[5,6] The optimum xenon pressure was found to be ~10 Torr, independent of input wavelength or intensity.

No attempt was made to compensate for the phase mismatch, Δk, in these initial experiments. Since 83 nm, the third-harmonic wavelength, is above the photoionization threshold for xenon, the index of refraction at this wavelength is complex. (The absorption cross section at 83 nm in xenon is[8] $\sigma_{\mathrm{abs}} \cong 6.5 \times 10^{-17}$ cm^2.) For lossless nonlinear media, the variation in third-harmonic output as a function of gas density may be related to the variation in phase mismatch.[6,9] For absorbing media, the third-harmonic output peaks and then begins to drop as a function of pressure when the absorption length becomes comparable with the confocal parameter of the fundamental beam.[6] Because appreciable harmonic conversion is observed in our case,

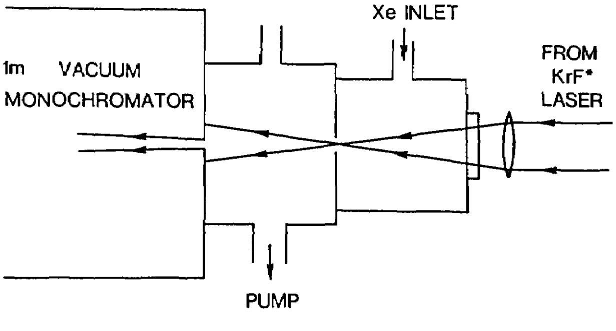

Fig. 1. The experimental apparatus for tripling KrF* radiation in xenon (see text).

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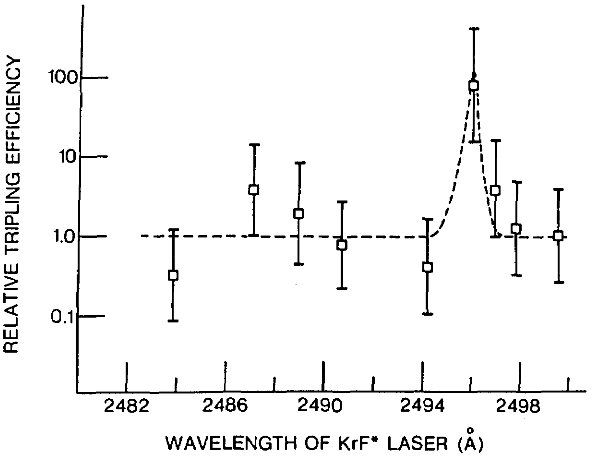

Fig. 2. Relative tripling efficiency as a function of the fundamental wavelength. Peak absolute efficiency is estimated to be $\sim 10^{-8}$.

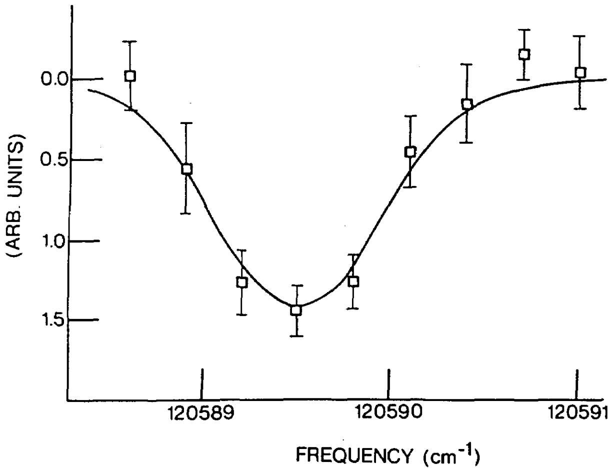

Fig. 3. Absorption cross section of H_2 at the $D \leftarrow X(4,0)$ $Q(3)$ line as observed in transmission of the tripled light. The solid line is a Gaussian profile with Doppler width (31 GHz).

we may at least infer that the sign of Δk is negative.[6,9]

Tunable XUV radiation was obtained from 82.8 to 83.3 nm. Figure 2 illustrates the conversion efficiency versus fundamental wavelength. More than an order-of-magnitude enhancement is observed at a fundamental wavelength of 249.63 nm, which arises from the two-photon resonance involving the Xe $6p(\frac{1}{2})_0$ state. The detected XUV output exhibits a cubic dependence on the input power, except close to the two-photon resonance, where an approximately linear dependence is observed. This linear behavior may be due to either Kerr-induced dispersion[10] or saturation effects.[11] The signature of the Kerr-induced dispersion mechanism is a variation in optimum gas density as a function of power density or detuning from resonance because of variations in phase mismatch.[10] Neither behavior was observed. As noted above, since the conversion efficiency is limited by loss at the third harmonic before optimal phase matching is achieved, it is believed that the variation in the power dependence near the two-photon resonance condition is primarily due to saturation. To support this view we note that the laser intensity in the focal region is estimated to be $\sim 10^{13}$ W/cm^2. Using the estimated two-photon absorption parameter $\alpha \cong 10^{-27}$ cm^4/W applicable for the $^1S_0 \rightarrow 6p(\frac{1}{2})_0$ transition[12] with the narrow-linewidth 248-nm radiation, we confirm that saturation behavior is expected.

In order to demonstrate explicitly the spectroscopic utility and narrow linewidth of the tunable XUV radiation, several transitions in molecular hydrogen have been studied. This was done by introducing ~100 mTorr of H_2 into the differential pumping chamber, as shown in Fig. 1. At the wavelength λ = 82.926 nm, strong absorption was observed that was due to the $Q(3)$ line of the $D^1\Pi_u \leftarrow X^1\Sigma_g{}^+$ (4,0) band. In Fig. 3, the absorption profile for this line is shown. The linewidth of the absorption is 31 ± 3 GHz, in agreement with the Doppler width of the transition at room temperature. Since the gas-flow characteristics in the differentially pumped chamber are not well known, we are only able to give a lower limit to the absorption cross section $\sigma \gtrsim 7 \times 10^{-16}$ cm^2. Many other absorption lines in H_2, HD, and D_2 are within the available XUV tuning range. Some of these lines are expected to be Doppler broadened, like the one shown in Fig. 3, whereas others are appreciably broadened and distorted by predissociation.[13] Detailed studies of these predissociation linewidths and line profiles will furnish considerable detailed information regarding diabatic interactions among the Rydberg states in hydrogen. Such an investigation is currently under way in this laboratory.

In conclusion, a coherent, tunable 83-nm light source has been developed using third-harmonic generation of tunable KrF* radiation in xenon and has been demonstrated to be a useful spectroscopic tool. We note that this source, in comparison with the outputs obtained from the brightest synchrotron radiation sources[14] at the same wavelength, provides a spectral brightness approximately 2 orders of magnitude greater.

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