A tunable, ultrahigh spectral brightness KrF* excimer laser source

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An extremely high spectral brightness KrF* (248 nm) excimer source is described. This instrument combines the property of continuous tunability over the full gain profile with the following output pulse characteristics: pulse energy ~60 mJ, pulse duration ~10 nsec, spectral width 150 ± 30 MHz, absolute frequency control to within 300 MHz, and beam divergence ~50 μrad. Within the uncertainty of measurement, the spectral width of the output radiation is Fourier transform limited, and the beam divergence corresponds to the diffraction of the radiating aperture.

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Rare-gas halide excimer sources have proven to be extremely useful in studies of multiphoton processes.[1] Nevertheless, the utility of these sources in such studies could be considerably enhanced. Heretofore, limitations of available sources have been (1) a broad (~100 cm^{-1}) emission profile, (2) the absence of a convenient, accurate, and reliable tuning system for control of the output wavelength, and (3) an output beam divergence on the order of one hundred times the diffraction limit. Overall, an enhancement of several orders of magnitude in spectral brightness, the key parameter in multiquantum processes, is achievable if the output parameters of these sources are made to conform to the most stringent limits fundamentally possible.

Previous studies[2,3] have shown that substantial improvement in source properties can be obtained by using the output of a tunable, intracavity etalon line-narrowed, discharge-excited oscillator to injection lock a high-energy, unstable resonator oscillator. The best linewidth reported was ~10^{-1} cm^{-1} in a beam with a divergence approximately twice the diffraction limit.[3]

In this letter, we report the properties of a KrF* excimer source with performance parameters which closely approach the fundamental limits governing spectral width, beam divergence, and absolute wavelength control. The basic laser system is illustrated schematically in Fig. 1. The output of a frequency-stabilized, cw dye laser (Coherent 599-21, $\Delta\nu < 5$ MHz, ~30 mW at 500 nm) is pulse amplified in a 3–4 stage XeF* pumped (30–50 mJ at 351 nm) dye amplifier, producing a 7-nsec visible pulse with an energy of ~3 mJ at a repetition rate up to 1 sec^{-1} (limited by thermal recovery time in the dye cuvettes). The linewidth of this visible radiation was examined with a scanning interferometer (Tropel 240) and was found to be 85 ± 10 MHz. Frequency-doubled radiation corresponding to any wavelength within the KrF* gain profile may readily be generated in a temperature-tuned, 90° phase-matched ADP crystal, producing ~5 nsec second-harmonic pulses with energy > 100 μJ. This spectrally narrow (120 ± 20 MHz FWHM, Fourier transform limited) second-harmonic radiation is subsequently amplified in a single pass through a discharge-pumped KrF* amplifier (Lambda Physik EMG 500) to produce output pulses of ~10-nsec duration, and energies up to 60 mJ.

A study of the spatial properties of the 2×0.5-cm-output beam reveal it to have a diffraction limited divergence of ~50 μrad. We note that at the peak measured power, ~6 MW, focal intensities at 248 nm in excess of 10^{15} W/cm^2 can be generated with f1 optics. In preliminary studies, we have observed optical breakdown in pure helium at pressures above 1.85 bars, with a KrF* laser intensity of ~3×10^{14} W/cm^2.

Furthermore, since the frequency of the cw dye laser can be electronically scanned over 1 cm^{-1}, continuously tunable coverage of the ultraviolet radiation over a 2-cm^{-1} interval is readily accomplished. In addition, the dye laser frequency is known to within 300 MHz by interferometric comparison with a stable HeNe laser source.[4] In our current experiments, we have observed amplification at wavelengths λ_{KrF} in the range 248.2 nm $< \lambda_{\mathrm{KrF}} <$ 250.3 nm. In this range, the output pulse energy was observed to monotonically decrease with increasing wavelength. In contrast to previous reports of a strong absorption centered at ~248.8 nm in both *e*-beam pumped KrF* amplifiers[5] and discharge-pumped tunable KrF* lasers,[2,6] no reduction in output energy was observed at that wavelength in our apparatus.

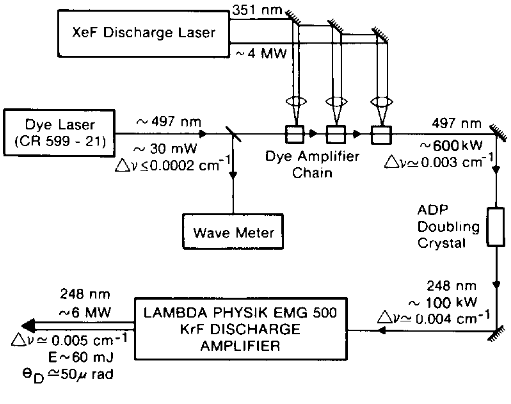

FIG. 1. Ultraviolet laser system illustrating the cw dye laser, the dye amplifier chain, the ADP doubling crystal, and the final KrF* amplifier stage.

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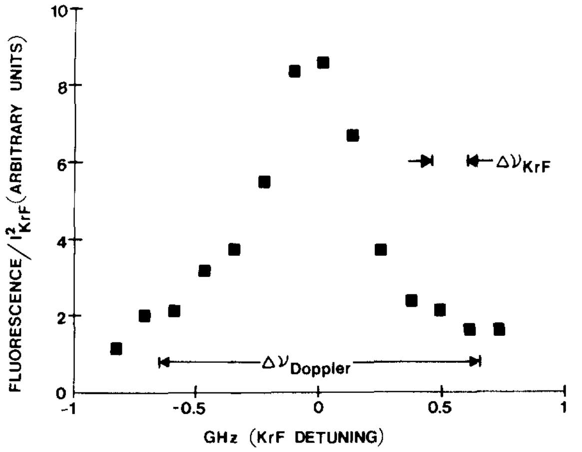

FIG. 2. Observed fluorescence at 828 nm following two-photon absorption in Xe($6p[\frac{1}{2}]_0 \leftarrow {}^1S$) at 249.63 nm, 0.25 Torr Xe pressure, and $\sim 5\times10^5$ W/cm^2 KrF* intensity (unfocused).

In order to establish the linewidth of our source, direct interferometry has been used. A 1 GHz FSR Fabry-Perot interferometer was constructed with two dielectric coated mirrors of 98% reflectivity, resulting in a finesse of ~20 for our typical 10-nsec pulse duration. With this interferometer, the amplified ultraviolet linewidth was measured to be $\Delta\nu_{KrF} = 150 \pm 30$ MHz. We observe that this linewidth is equivalent, within our experimental uncertainty, to the Fourier-transform-limited linewidth $\Delta\nu_{SHG} = 120 \pm 20$ MHz of the second harmonic output of the ADP crystal.

In order to further verify the narrow linewidth of the laser output, a preliminary study of the xenon ($6p[\frac{1}{2}]_0 \leftarrow {}^1S$) two-photon absorption at 249.63 nm was performed under Doppler-free conditions through detection of the resulting ($6p[\frac{1}{2}]_0 \rightarrow 6s[\frac{3}{2}]_1$) fluorescence at 828 nm. Upon scanning the KrF* source frequency across the resonance, a single feature was observed, centered at $80\,118.73 \pm 0.10$ cm^{-1}, with a width of ~450 MHz (FWHM, at the KrF* frequency), as shown in Fig. 2. These measurements were made at a xenon pressure of 0.25 Torr and an ultraviolet intensity of $\sim 5\times10^5$ W/cm^2. Although this width is a factor of 3 less than the Doppler width, the resonance is still a factor of ~3 broader than would be expected with a source linewidth of ~150 MHz. Since we experimentally determined the pressure broadening coefficient of this xenon two-quantum transition to be 25 ± 15 MHz/Torr, which gives a minor contribution to the linewidth at 0.25 Torr, we have concluded that unresolved isotope splittings (five xenon isotopes with natural abundances > 8%) lead to the observed width. An estimate[7] of the isotope shift for such a ($5p^5 6p \leftarrow 5p^6$) transition yields a relative shift of ~50–100 MHz for two Xe isotopes whose masses differ by 1 amu. This estimate agrees well with both the linewidth of the observed absorption, and the absence of additional resonances within $\pm$ 6 GHz of the one observed.

In conclusion, we state that an ultrahigh spectral brightness ultraviolet source, continuously tunable over the KrF* excimer band, has been constructed with the following demonstrated performance characteristics: $P \sim 6$ MW, $\Delta\nu_{KrF} = 150 \pm 30$ MHz, pulse repetition rate ~1 sec^{-1}, and divergence $\theta_D \simeq 50\,\mu$rad.

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