High power ultraviolet source with extreme frequency stability

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A continuous wave ultraviolet source with extreme frequency stability is described. A ring dye laser with internal electro-optic phase modulator reaches sub-Hertz linewidths relative to a passive reference cavity with the help of an FM stabilization technique. A barium- β -borate frequency doubler crystal inside a second injection-locked ring dye laser generates output powers of 30 mW near 243 nm.

We report on a source of continuous wave ultraviolet radiation which combines extreme frequency stability with high power. A ring dye laser with internal fast electro-optic phase modulator is locked to a passive reference cavity by an FM sideband technique, as described earlier [1]. With improvements in the frequency response of the servo loop, the laser reaches sub-Hertz linewidths relative to the passive cavity. This oscillator injection-locks a second ring dye laser with an internal angle-tuned barium β -borate (BBO) frequency doubler crystal. The cavity of this second laser is kept in resonance with the injecting light by a second FM servo system, so that stable optical phase locking becomes possible. Output powers of 30 mW have thus been produced near 243 nm with a linewidth limited only by the master oscillator. To our knowledge, the resulting spectral brightness is the highest reported so far for a tunable cw ultraviolet source.

This work has been motivated by our interest in high resolution two-photon spectroscopy of the extremely narrow 1S-2S transition in atomic hydrogen [2]. In past experiments, cw ultraviolet powers of a few milliwatts near 243 nm have been produced by second harmonic generation of 486 nm cw dye laser radiation in angle-tuned BBO. The doubler crystal was placed inside an external build-up cavity [2] or inside the dye laser cavity [3]. In either case, the circulating visible power was limited by losses in the many tuning and control elements inside the cavity of the stabilized dye laser in addition to the losses in the crystal. Even small gains in visible intensity can translate into large improvements in spectroscopic signal, since the two-photon excitation probability grows with the square of the UV intensity and thus with the fourth power of the fundamental intensity. In separate experiments, we have explored doubly resonant second harmonic generation inside an external resonator, where both the fundamental and the second harmonic radiation are resonantly enhanced [4]. The output power, however, remained limited to a few milliwatts by UV-induced damage in the dielectric antireflection coatings of the BBO crystal.

To overcome these problems, the system reported here uses a Brewster-cut BBO crystal without coatings inside an active ring dye laser cavity where a high circulating power is realized by removing most of the lossy intracavity elements. Wavelength selection, frequency stabilization, and unidirectional operation are achieved by injecting the light from a separate highly stable ring dye laser oscillator into this second cavity. A schematic overview of the system is shown in fig. 1.

It is well known that a master laser can control a slave laser by injection locking if one of the axial modes of the slave is tuned close to resonance [5-7]. If the injected power exceeds a threshold, the slave oscillators only at the injected frequency, and its original free running frequency is extinguished. Optical phase locking occurs over a frequency range [7]



Fig. 1. Scheme for generating ultraviolet radiation with extreme frequency stability and high power. The light from a highly stabilized dye laser oscillator 1 is injected into a second ring dye laser cavity with a BBO frequency doubler. An FM servo scheme is employed to control the frequency of the second laser cavity so that optical phase locking occurs.

$$\Delta f = \eta \, \frac{TF}{\pi} \left(\frac{P_{\text{master}}}{P_{\text{slave}}} \right)^{1/2},\tag{1}$$

where P_{master} and P_{slave} are the power of the first and second laser, T is the transmission of the input coupling mirror, F is the free spectral range of the slave, and η is an efficiency factor for the mode overlap.

Two commercial ring dye lasers (Coherent 699-21) were available for our experiments. Each is operated with coumarin 480 and pumped by a krypton ion laser with about 5 W at 415 nm. One laser is stabilized to a reference resonator by an FM locking technique, as described elsewhere in detail [1]. The frequency response of the electronic servo loop has been improved since the earlier report and is illustrated in fig. 2. Fig. 3 shows the new Allen variance of the optical spectrum, as derived from the servo signal. For sampling times of a few hundred milliseconds, the linewidth is reduced below 1 Hz relative to the reference resonator.

The second dye laser is operated without its intracavity etalons, Lyot filter, and optical diode. An acousto-optic modulator (AOM) operated near 80 MHz serves as optical isolator to decouple the two laser resonators. A lens of 500 mm focal length matches about 200 mW of the frequency shifted light



Fig. 2. Frequency response of the FM servo system used to stabilize the dye laser oscillator to a reference cavity.



Fig. 3. Allan variance of the spectrum of the laser oscillator obtained from the frequency discriminator signal of the FM servo system.

from the first laser into a mode of the second laser. The radiation is injected through the standard 2% output coupler.

From eq. (1) we estimate that the slave ring dye laser must follow the frequency of the injecting oscillator to within about 0.5 MHz in order to assure reliable optical phase locking. This is a somewhat narrower range than can be readily realized with the factory-provided stabilization system. An available second FM sideband servo loop was therefore employed which is almost identical to that of the master oscillator [1]. An external electro-optic modulator (EOM) generates FM sidebands at 18 MHz in the injecting beam which remain outside the slave cavity resonance frequency and serve as a phase reference. Any detuning of the carrier from exact resonance introduces an amplitude modulation of the reflected light which provides the frequency discriminator signal needed to adjust the three tuning elements, a fast internal electro-optic phase modulator (EOM), a piezo-mounted "tweeter" mirror, and a galvanometer-driven Brewster plate. With a unity gain bandwidth of about 10 MHz, this servo system achieves a frequency noise spectral density of less than 1 Hz/\sqrt{Hz} outside the optical phase locking bandwidth and reduces the relative phase fluctuations of the two lasers to a few milliradian, as estimate from the integrated frequency noise density [7].

The BBO frequency doubler crystal available for the present experiments has a length l=8 mm, and its faces are cut at Brewster's angle. The crystal is oriented for a phase matching angle of 55° at 486 nm. Double refraction causes a "walk-off" angle $\rho=4^{\circ}$ for the 243 nm radiation [8,9]. According to Boyd and Kleinmann [8], the efficiency of second harmonic generation can be predicted with a function $h_{\rm m}(B, \xi)$, which depends on the inverse confocal parameter ξ of the beam waist inside the crystal and on the walk-off parameter $B=\rho\sqrt{lk_1}/2$, where k_1 is the absolute value of the wave vector of the fundamental beam. The estimated optimum value for the waist size in our crystal is 16 µm.

This optimum waist size is smaller than the 50 μ m value at the so-called auxiliary waist in the standard ring dye laser. The location of this waist inside the cavity is illustrated in fig. 4. The waist size w_2 can be changed by adjusting some mirror distances. An analysis of the ring cavity with the usual ABCD matrix approach [10] shows that w_2 can be reduced to as little as 12 μ m by decreasing the mirror distance d_1 from 125 to 110 mm and simultaneously increasing the distance d_2 from 200 to 260 mm. Unfortunately, the same adjustment decreases the waist size w_1 at the dye jet from about 80 μ m to 50 μ m, so that is is not easy to optimize the two waist sizes independently.

The BBO crystal is mounted on a rigid mechanical stage that permits fine adjustment of all degrees of freedom. In order to insert the BBO crystal at the auxiliary waist, it is mainly necessary to move the upper folding mirror in order to compensate for the different optical path length. The mirror distances



Fig. 4. Active dye laser ring cavity with BBO frequency doubler inserted at the auxiliary beam waist w_2 . A quartz rhomb provides astigmatic compensation. A galvanometer driven Brewster plate, a piezo transducer (PZT) mounted mirror, and a fast electro-optic phase modulator (EOM) serve as frequency control elements to keep the cavity in resonance with the injected radiation.

are then optimized while observing the UV power. To match the beam diameter of the ring resonator to the pump focus in the dye jet, both lower folding mirrors can be translated, so that the tight focus can be moved out of the jet. So it is possible to compensate for the fact that in the chosen geometry a very small waist size in the BBO crystal also implies a small waist size near the jet.

To extract the UV output, the usual high reflector M6 is replaced by a similar mirror with high transmission near 243 nm and high reflection at 486 nm. At the fundamental wavelength, its losses as well as those of the intracavity Brewster plate and astigmatic compensation rhomb remain negligible compared to those of the output coupler. The biggest cavity losses are introduced by the BBO crystal. We have measured a single-pass transmission loss of 5% despite repeated repolishing of its optical surfaces. Even better results can therefore be expected with a different crystal of lower scattering losses.

With an open servo loop, the free running second dye laser reaches a circulating fundamental power of 15 W. After closing the servo loop, this power increases to 22 W, and 30 mW of second harmonic power are generated at 243 nm. Without the BBO crystal, the intracavity fundamental power reaches 80 W. The injection threshold is about 80 mW.

The ultraviolet output beam appears severely astigmatic and distorted. However, it is possible to compensate for much of this asymmetry by using offaxis reflection from a spherical mirror. In this way, one can couple more than 40% of the light into a clean gaussian TEM_{00} mode inside a separate external standing wave cavity [3].

To investigate the frequency stability of the injection locked ring dye laser, we have analyzed the 80 MHz beat note between its AOM shifted fundamental frequency and the driving laser oscillator. The observed spectral width of 200 Hz is entirely limited by the resolution of the rf spectrum analyzer. These results confirm that the closed frequency servo loop ensures stable phase locking of the two lasers.

Although the described system is complex and expensive, it offers a spectral brightness not available before in a tunable UV source. Such a system opens novel possibilities for precision experiments in non-linear high resolution laser spectroscopy or for laser cooling and manipulation of atoms and ions.

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