GENERATION OF CONTINUOUS-WAVE 243-nm RADIATION BY SUM-FREQUENCY MIXING

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We have generated tunable cw radiation near 243 nm with a linewidth of less than 4 MHz by sum-frequency mixing the 351 nm radiation from an argon-ion laser with the 789 nm radiation from a ring dye laser in a crystal of ammonium dihydrogen phosphate held at moderate temperature. An external ring cavity, resonant with the dye laser, gives a power enhancement of about 12 in the sum-frequency generated radiation. Thermal lensing due to laser heating of the nonlinear crystal, distorted the 351 nm mode structure. This effect could limit the efficiency of the sum frequency mixing process.

Several years ago, two-photon spectroscopy with a pulsed laser yielded the first measurements of the 1S Lamb shift of atomic hydrogen and deuterium [1]. In these experiments the precision was limited to ~0.1% by the transform-limited pulsed amplified source of radiation at 243 nm. A thousend-fold improvement in the experimental precision would be possible with a narrowband, continuous-wave 243 nm radiation source with sufficient power to drive the narrow 1S-2S two photon transition. Gouillaud et al. [2] have recently reported the generation of cw radiation at 243 nm by sum-frequency mixing (SFM) of two lasers at 590 nm and 413 nm in a crystal of ammonium dihydrogen phosphate (ADP). However, the output was seriously limited by damage to the ADP crystal, presumably due to the necessity of operating the crystal near its Curie temperature (−115°C) in order to obtain 90° phasematching. We have recently reported the generation of tunable continuous wave radiation at 194 nm by frequency mixing of 792 and 257 nm radiation in a potassium pentaborate (KB5) crystal [3]. In this communication we report a method for generating about 0.3 mW of stable cw radiation at 243 nm by slightly modifying the 194 nm experimental setup.

The very low temperature necessary to obtain 90° phasematched SFM of 590 nm and 410 nm radiations to 243 nm may be avoided by increasing the disparity between wavelengths of the two mixed radiations. For example [2,4], the 243 nm radiation can be generated at moderate ADP crystal temperatures by mixing the radiation from a dye laser operating in the 730–800 nm region with one of the two strong UV lines of the Ar-ion laser. In particular, 90° phasematched SFM of 351 nm and 789 nm radiation to 243 nm is obtained near 8°C in ADP. We chose these wavelengths since we already had an external, enhancement ring cavity with mirrors that were optimized for reflectance near 790 nm. Passive external resonant cavities have recently proved valuable in enhancing the output power of sum-frequency generated radiation at UV wavelengths [2,3].

A schematic of the experimental setup is shown in fig. 1. The primary radiation sources are a ring dye laser operating with the dye Rhodamine-700 near 789 nm and an argon-ion laser operating on the UV lines. Both lasers operate in a single-axial mode and are frequency stabilized by separate reference cavities. Single mode operation of the Ar-ion laser is achieved with a temperature tuned uncoated solid etalon (UV grade fused silica) 15 mm in length, but with no intracavity prism. The 351.1 and 363.7 nm lines are observed to be single frequency and stable. There is a 30 to 40 percent loss in the output power with the insertion of the etalon tuned for single mode operation. (We have tried this etalon with two separate Ar lasers; one

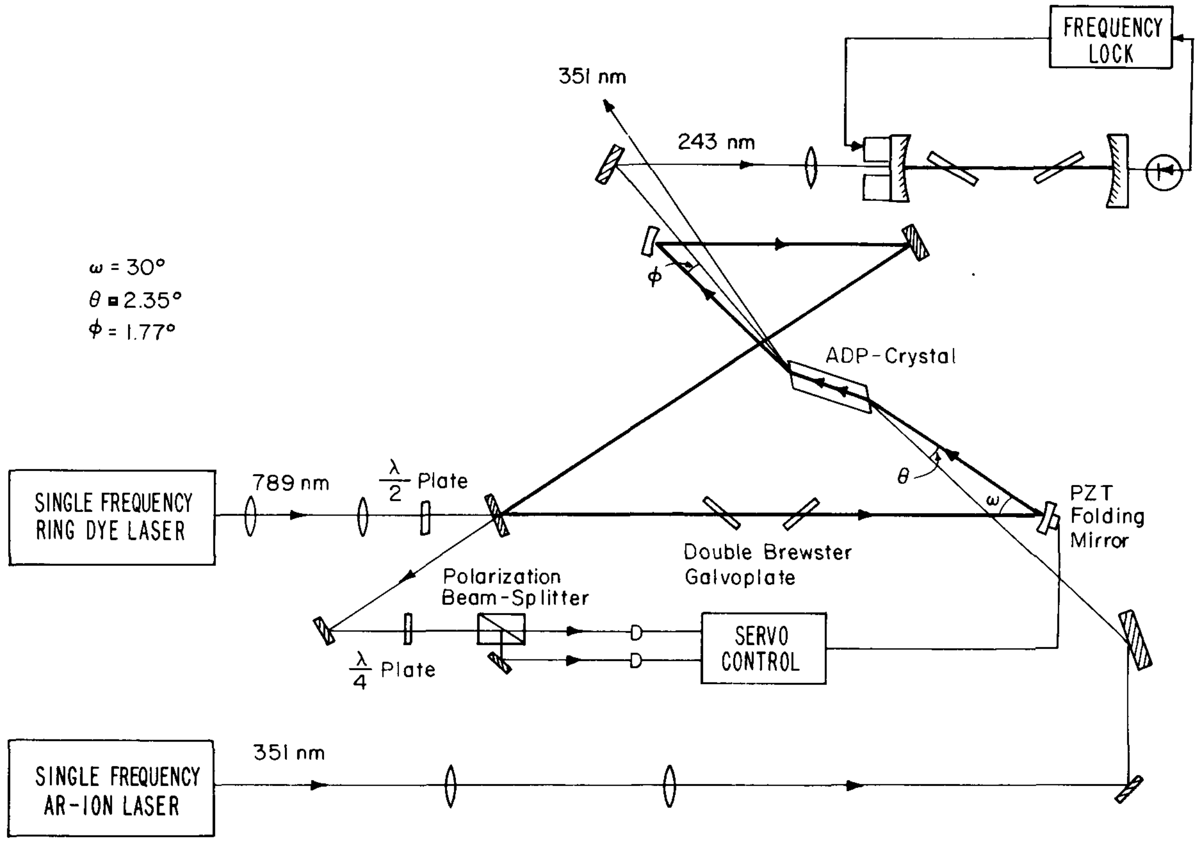

Fig. 1. Experimental setup for generation of 243 nm radiation in an external ring cavity.

laser demonstrated complete single mode operation at the 351 nm line, while the second was nearly always accompanied by an additional axial mode with ~5–10% of the dominant mode power). A fraction of the argon-ion laser's output beam is directed through an external prism to separate the UV lines. The ion laser is then stabilized to a few MHz by locking the 351 nm line to the half-power point of a transmission fringe of a Fabry-Perot cavity [5].

A 50 mm × 5 mm × 5 mm Brewster-cut ADP crystal is used for the SFM. To enhance the 789 nm power inside the crystal, this crystal is placed inside an external ring cavity, which is locked to resonance with the dye laser. The Brewster-cut crystal minimizes Fresnel reflection losses for the input beams. The ring cavity is completely compensated for astigmatism and to some extent for coma [6]. The 789 and 351 nm beams are collinear in the crystal and polarized in the same direction. The dye laser beam is introduced into the cavity through a partially reflecting mirror ($R \approx 90$ percent). Fifteen centimeters from the ADP crystal the 243 nm radiation is well separated from the 789 nm radiation since the refraction angles differ by ~1.77°. Because of this, no special mirrors or dichroic beam splitters are necessary inside the 789 nm enhancement cavity to extract the 243 nm radiation. Maximum power enhancement is obtained by locking the cavity to resonance, using the polarization technique first discussed by Hänsch and Couillaud [7,8]. A more detailed description of the enhancement ring cavity is given in ref. [3].

The ADP crystal is located inside a small housing for temperature tuning and regulation [3]. The ADP crystal temperature must be maintained near 8°C to achieve 90° phasematched SFM for the wavelengths used. The crystal is positioned in the beam waist of the cavity, which is about 50 μm in diameter. The 351 nm radiation is focused to about the same diameter and superposed with the 789 nm radiation for maximum UV generation. With input powers of 500 mW and 150 mW at 351 and 789 nm respectively, and a power buildup factor of 10 in the enhancement cavity, about 0.3 mW of single-frequency 243 nm radiation is obtained. This corresponds to an efficiency parameter of about 4×10^{-4} W^{-1} for the SFM process. The output power remained stable for many hours of operation. The UV power is further boosted by injecting the 243 nm radiation into an enhancement cavity which is locked to resonance. This cavity is formed by two dielectric mirrors, both with a radius of curvature of 200 cm and separated by 50 cm. The mirror reflectances are 98% and 85% at 243 nm. The mirror with 85% reflectance is used as the input coupler. Despite the fact that the reflectances of the available mirrors were far from optimal, this cavity yields a power enhancement of nearly 12. Thus standing wave radiation of about 3 mW circulating power is produced inside the cavity. The insertion of two high quality Brewster windows into the cavity degrades the power buildup by only a few percent.

We anticipate that the output power can be raised by an order of magnitude when an ADP crystal of better quality, and better surface finish is used. About 350 mW of 789 nm radiation is now readily attainable from our dye laser; this may also improve the 243 nm power. However, the power-enhancement factor inside the ring cavity and the efficiency of SFM may be limited by thermal gradients that arise in the ADP crystal from absorption heating by the laser beams. Thermal gradients stemming from localized heating have been shown to cause a self-defocusing effect on cw beams passing through materials with finite loss [9,10]. In our experiment, the thermal lensing effect is particularly apparent on the 351 nm beam profile. If the 789 nm enhancement ring cavity is not locked to resonance, the 351 nm beam exits the ADP crystal with a clean TEM_{00} mode structure. However, when the ring cavity is locked to resonance with the dye laser and nearly 2 W of 789 nm radiation circulates through the crystal, the outgoing 351 nm beam spreads in diameter and displays a doughnut mode structure (fig. 2). This result is independent of the presence of 243 nm radiation. The observed blooming effect may be attributed to interference effects caused by the spherical aberration of the thermal lens. Because of this, the 351 nm, and 789 nm beams no longer remain completely overlapped along the length of the crystal, thus reducing the expected efficiency. The thermal lensing effect could also be a limiting mechanism in the amount of buildup achieved from the 789 nm enhancement ring cavity. Surprisingly, at the present power levels, the mode structure of the 243 nm radiation appears to be dominantly TEM_{00} in spite of the distortion of the 351 nm mode structure. Thermal blooming was also observed in sum-frequency mixing of 257 nm and 790 nm radiation in a KB5 crystal.

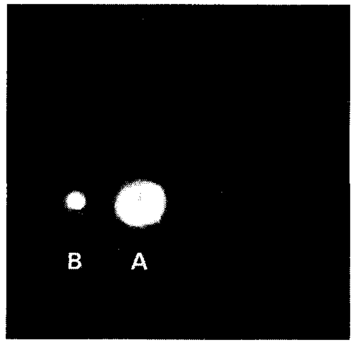

Fig. 2. Thermal lensing in ADP crystal. (A) The 351 nm radiation after passing through an ADP crystal located in an external ring cavity locked to resonance with the 789 nm dye laser. (B) The 243 nm sum-frequency generated output.

Such a source of 243 nm radiation not only promises to improve considerably the measured value of the ground state Lamb shift of atomic hydrogen; in addition, through direct frequency measurements of the 1S-2S two-photon transition in hydrogen and deuterium, a more-precise value of the Rydberg constant and of the electron-to-proton mass ratio may also be obtained [11]. Furthermore, relatively high fluxes of cold metastable atomic hydrogen can be generated with this source of 243 nm radiation, which might be valuable in parity non-conservation experiments.

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