Generation of continuous-wave 194-nm radiation by sum-frequency mixing in an external ring cavity

H. Hemmati, J. C. Bergquist, and Wayne M. Itano

Time and Frequency Division, National Bureau of Standards, Boulder, Colorado 80303

Received November 4, 1982

Several microwatts of tunable cw radiation near 194 nm in a linewidth of less than 2 MHz have been generated by sum-frequency mixing the radiation from a frequency-doubled argon-ion laser with the radiation from a ring dye laser in a crystal of potassium pentaborate. An external ring cavity resonant with the dye laser gives an enhancement factor of about 14 in the sum-frequency-generated radiation power. The Doppler-limited absorption spectrum of the $6s\ ^2S_{1/2}$–$6p\ ^2P_{1/2}$ first resonance line of natural Hg II has been resolved, and the vacuum wave number for the mass-202 isotope has been measured to be 51485.904(20) cm^{-1}.

High-resolution spectroscopy of atomic and molecular lines in the UV region of the spectrum requires tunable sources of radiation. Many of the tunable UV sources can only be obtained by second-harmonic generation (SHG) or sum-frequency mixing (SFM). For radiation pressure cooling and optical pumping of electromagnetically confined mercury ions, which have great potential in optical and microwave frequency standards,[1] a narrow-band tunable cw laser near the $6s\ ^2S_{1/2}$–$6p\ ^2P_{1/2}$ first resonance line (194.2 nm) is required. For optimum cooling, the frequency, linewidth, and stability of the radiation source must be less than the natural linewidth of the resonance line of Hg^+ (about 70 MHz) with a minimum cw power requirement, which is estimated, from experiments on Mg^+ ions,[2] to be about 1 μW.

In this Letter we describe a method for producing 194-nm radiation by SFM, in a potassium pentaborate (KB5) crystal, the 257-nm second harmonic of the output of a cw 515-nm argon-ion laser with the output of a tunable cw dye laser in the 792-nm region. Previously, Stickel and Dunning,[3] using pulsed dye lasers, generated coherent radiation tunable between 185 and 217 nm by SFM in KB5. However, cw sources of radiation can demonstrate better frequency stability and smaller linewidths. The shortest-wavelength cw radiation that was previously produced by SFM in a crystal is approximately 211 nm.[4] The only other technique that has been demonstrated for generation of coherent cw radiation below 200 nm is four-wave mixing in strontium vapor, at approximately 170 nm, by Freeman *et al.*[5]

A schematic of the experimental setup is shown as Fig. 1. The primary radiation sources are a ring dye laser operating with the dye LD700 near 792 nm and a 257-nm source that is the frequency-doubled output of the 515-nm line of an argon-ion laser. The 257-nm second harmonic is generated in an ammonium dihydrogen phosphate (ADP) crystal, which is placed in an external ring cavity that is resonant at the fundamental frequency.[6] Both lasers operate in a single-frequency mode and are frequency stabilized to separate reference cavities. For the argon-ion laser, additional long-term stabilization is provided by locking the reference cavity to a hyperfine component of a corresponding molecular iodine line. A 30-mm × 5-mm × 5-mm Brewster-cut KB5 crystal is used for SFM. To enhance the 792-nm radiation power inside the crystal, this crystal is placed inside an external ring cavity, similar to the frequency-doubling cavity, which is locked on resonance with the dye laser. The Brewster-cut crystal minimizes Fresnel losses for the input beams and compensates for beam astigmatism inside the ring cavity. The 792- and 257-nm input beams are polarized along the *a* axis of the crystal. The 194-nm beam is polarized along the *c* axis, and all three beams propagate along the *b* axis. For the biaxial KB5 crystal, this is the 90° phase-matched condition. The dye-laser beam is introduced into the cavity through a partially reflecting mirror ($R \cong 90\%$). The sum-frequency radiation is well separated from the 792-nm radiation at 15 cm from the KB5 crystal since the refraction angles for these radiations differ by ~2.5°. Because of this, no special mirrors or dichroic beam splitters are necessary inside the 792-nm enhancement ring cavity to extract the 194-nm radiation. Maximum

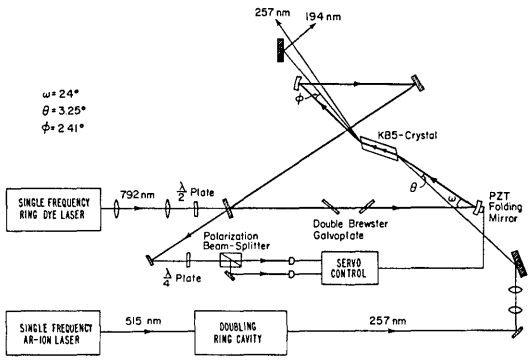

Fig. 1. Experimental setup for generation of 194-nm radiation in an external ring cavity.

0146-9592/83/020073-03$1.00/0

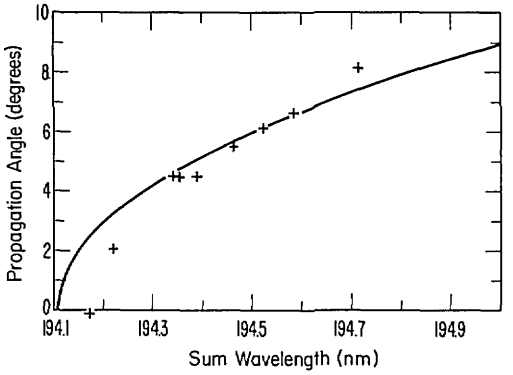

Fig. 2. Angle tuning in KB5 crystal. The crosses represent our data; the solid curve is from calculations.

enhancement is obtained inside the cavity by stabilizing the cavity on resonance, using the polarization technique discussed by Hänsch and Couillaud.[7] A similar enhancement cavity was recently used by Couillaud *et al.* for SFM in an ADP crystal.[8]

As was previously discussed for SHG of the 257-nm radiation,[6] the external ring cavity offers several attractive features, including the possibility of achieving higher enhancement factors than is generally possible with laser intracavity SFM. In this experiment, buildup factors as high as 14 were obtained using an input mirror with a reflectivity of about 90%. This buildup factor is presumably limited by the quality of the surface polish of our KB5 crystal.

The KB5 crystal is located inside a small housing that is equipped with electrical heaters to provide the temperature tuning necessary for the 90° phase-matched condition. This crystal is positioned in the beam waist of the cavity, which is about 50 μm in diameter and is close to the optimum spot size for sum-frequency generation. The 257-nm radiation is focused to about the same diameter and superposed with the 792-nm radiation for maximum UV generation.

With input powers of 25 and 220 mW at 257 and 792 nm, respectively, and a buildup factor of 12 in the enhancement cavity, about 2 μW of single-frequency 194-nm radiation is obtained. This corresponds to an efficiency parameter of about 3×10^{-5} W^{-1} for the SFM process. This is about a factor of 2 smaller than the value calculated from the previously measured nonlinear coefficients of KB5.[9] The output radiation was measured by a calibrated EMR Type 541F-05M-14 solar-blind photomultiplier tube (PMT). About 80 mW of 257-nm and 300 mW of 792-nm radiation were obtained previously in our experiments. At these input power levels, a fourfold improvement to 8 μW of 194-nm radiation is expected. At the 2-μW power level, we see no degradation of the 194-nm radiation power with time. Since the 257- and 792-nm radiation beams are also well separated outside the crystal, again because of different refraction angles, a second ring cavity may be added to enhance the power of the 257-nm radiation, which would further increase the 194-nm radiation power. The linewidth of the 194-nm output is less than 2 MHz, inferred from the linewidth of the primary beams, each of which is about 1 MHz.

Figure 2 shows the propagation angle for the input beams in the a–b plane, relative to the b axis, versus the wavelength for which angle phase-matched SFM occurs at a temperature of about 25°C. The solid curve is calculated from the published indices of refraction.[10]

Figure 3 shows the wavelength for which 90° phase-matched SFM occurs, as a function of temperature. In both cases (Figs. 2 and 3) the 257-nm wavelength is fixed and the 792-nm wavelength is varied. The greatest conversion efficiency occurs when the 90° condition is met. For the mercury-ion resonance line at 194 nm, the required 90° phase-matching temperature is about 34°C.

A preliminary study was made of the isotope shift and hyperfine structure of the $6s\ ^2S_{1/2}$–$6p\ ^2P_{1/2}$ 194.2-nm transition of singly ionized mercury by absorption spectroscopy. The argon-ion laser was stabilized at a fixed frequency while the dye laser was tuned in frequency. The dye laser could be continuously scanned over 40 GHz. The external enhancement ring cavity is locked and synchronously scanned with the dye laser. Both cavities are frequency scanned by double tipping Brewster plates, which have nearly zero insertion loss and do not cause any beam displacement as the cavities are tuned.

The absorption spectrum is obtained by probing the ground state of the singly ionized Hg ions that are created in a glass cell excited by an electrodeless rf discharge, using 150-MHz rf radiation with power of less than 10 W. The cell contained the naturally occurring Hg isotopes with Xe gas(10^{17} cm^{-3}) as the buffer and was heated to about 70°C to increase the density of the Hg atoms. To eliminate laser intensity noise, the signal was differentially detected by using two photomultiplier tubes. A fraction of the 194-nm beam was split off and directed into one PMT. The remaining radiation was directed through the Hg cell and subsequently detected by the second PMT. By using a differential amplifier, the gains of the two PMT's were adjusted to balance the intensity fluctuations of the 194-nm radiation. No attempt was made to normalize the spectrum against laser-power variations.

To calibrate the frequency scan, a portion of the 792-nm dye laser beam was directed into a 250-MHz free-spectral-range confocal interferometer. This in-

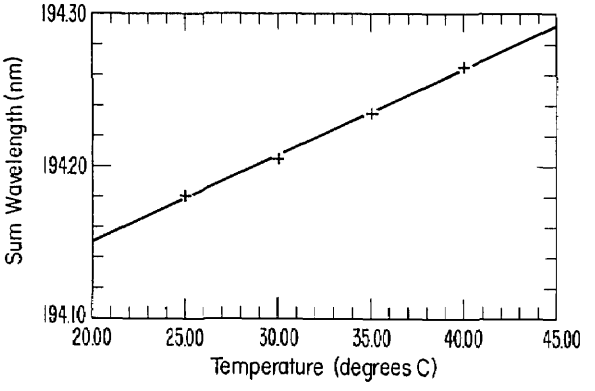

Fig. 3. Temperature tuning in KB5 crystal. The crosses represent our data; the line is a linear least-squares fit.

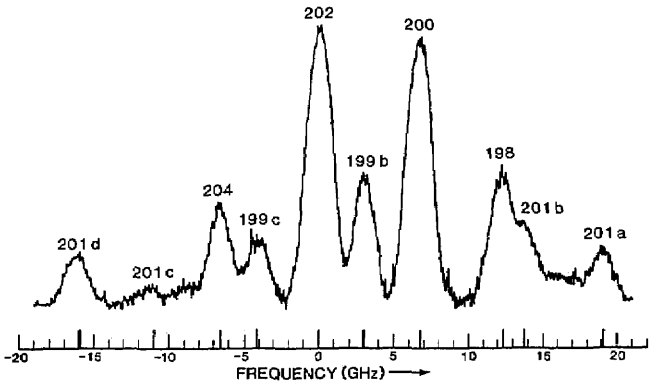

Fig. 4. Isotope and hyperfine-structure components of the 194.2-nm resonance line in natural Hg II.

terferometer gave simultaneous frequency markers with the recording of the atomic-absorption spectrum as a function of the dye-laser frequency. A single scan of the isotope and hyperfine structure of the 194-nm transition recorded in approximately 24 sec is shown in Fig. 4. The most abundant isotopes of mercury are masses 202 (30%), 200 (23%), 199 (17%), 198 (10%), 201 (13%), and 204 (7%), which appear clearly in our scan. The contribution from the 199*a* hyperfine component, which is located at about 24.7 GHz, to the blue of the 201*a* hyperfine component is not included in this figure. The relative line spacing of the absorption spectra is in good agreement with the previous work by Guern *et al.*, in which Fabry–Perot interferometric techniques were used.[11]

The wavelength of the $^{202}Hg^{+}$ component was determined from the wavelengths of the input lasers. The argon-ion laser was locked to the a_2 component of the 43-0, $P(13)$ transition of $^{127}I_2$, whose vacuum wavelength is known to be 514.67352 nm.[12] An interferometric wavelength meter,[13] which has an accuracy of a few parts in 10^7, was used to measure the wavelength of the dye laser. The reference for the wavelength meter was a He–Ne laser stabilized to the *h* component of the 11-5, $R(127)$ transition of $^{127}I_2$, which has a vacuum wavelength of 632.99137 nm. Proper corrections were made for the refractive index of air for our laboratory physical conditions, using the formulas of Edlén.[14] The dye-laser vacuum wavelength necessary to generate UV radiation in resonance with the $^{202}Hg^{+}$ component was measured to be 791.9965 ± 0.0002 nm, where the accuracy is limited by our inability to determine the line center. This corresponds to the UV sum-frequency wave number of 51485.904 ± 0.020 cm^{-1}. The uncertainty in the UV wave number has been increased because of possible line shifts caused by the electric fields of electrons and ions in the discharge. The measured width of an individual component is 1.8 GHz. If we assume that the line profile is the convolution of a Lorentzian line shape and a Gaussian Doppler profile, calculated for the temperature measured at the cell walls, we find that the width of the Lorentzian is 620 MHz, compared with the natural width of 70 MHz. The line shift can be of the same order.

In conclusion, we have demonstrated the possibility of generating narrow-band tunable cw coherent radiation below 200 nm by SFM in KB5. If the argon-ion laser were replaced with a tunable dye laser, or if other ion laser lines were used, tunable cw UV radiation could be generated at even shorter wavelengths, possibly down to 185 nm or below. The 194-nm radiation produced by the SFM was used to resolve the isotope and hyperfine structure of the $6s\ ^2S_{1/2}$–$6p\ ^2P_{1/2}$ transition in Hg II and to measure the absolute wave number for this line in $^{202}Hg^{+}$.

We wish to thank D. J. Wineland, R. E. Drullinger, F. L. Walls, and K. B. Persson for many stimulating and useful discussions. We also thank J. Faller for the loan of the National Bureau of Standards iodine-stabilized He–Ne laser. We gratefully acknowledge the support of the U.S. Air Force Office of Scientific Research and the Office of Naval Research.

References

1. D. J. Wineland, W. M. Itano, J. C. Bergquist, and F. L. Walls, in *Proceedings of 35th Annual Symposium on Frequency Control* (Electronic Industries Associates, Washington, D.C., 1981), pp. 602–611; F. G. Major and G. Werth, Phys. Rev. Lett. **30,** 1155–1158 (1973); M. D. McGuire, R. Petsch, and G. Werth, Phys. Rev. A **17,** 1999–2004 (1978); M. Jardino, M. Desaintfuscien, R. Barillet, J. Viennet, P. Petit, and C. Audoin, Appl. Phys. **24,** 107–112 (1981); L. S. Cutler, R. F. Gifford, and M. D. McGuire, in *Proceedings of Thirteenth Annual Precise Time and Time Interval Applications and Planning Meeting, Washington, D.C., 1981,* NASA Conf. Pub. 2220 (National Aeronautics and Space Administration, Washington, D.C., 1982), pp. 563–578; P. L. Bender, J. L. Hall, R. H. Garstang, F. M. J. Pichanick, W. W. Smith, R. L. Barger, and J. B. West, Bull. Am. Phys. Soc. **21,** 599 (1976).
2. D. J. Wineland, R. E. Drullinger, and F. L. Walls, Phys. Rev. Lett. **40,** 1639–1642 (1978).
3. R. E. Stickel, Jr., and F. B. Dunning, Appl. Opt. **17,** 981–982 (1978).
4. R. E. Stickel, Jr., S. Blit, G. F. Hildebrandt, E. D. Dahl, F. B. Dunning, and F. K. Tittle, Appl. Opt. **17,** 2270 (1978).
5. R. R. Freeman, G. C. Bjorklund, N. P. Economou, P. F. Liao, and J. E. Bjorkholm, Appl. Phys. Lett. **33,** 739–742 (1978).
6. J. C. Bergquist, H. Hemmati, and W. M. Itano, Opt. Commun. (to be published).
7. T. W. Hänsch and B. Couillaud, Opt. Commun. **35,** 441–444 (1980).
8. B. Couillaud, Ph. Dabkiewicz, L. A. Bloomfield, and T. W. Hänsch, Opt. Commun. **7,** 265–267 (1982).
9. H. J. Dewey, IEEE J. Quantum Electron. **QE-12,** 303–306 (1976).
10. W. R. Cook, Jr., and L. M. Hubby, Jr., J. Opt. Soc. Am. **66,** 72–73 (1976); F. B. Dunning and R. E. Stickel, Jr., Appl. Opt. **15,** 3131–3134 (1976).
11. Y. Guern, A. Bideau-Méhu, R. Abjean, and A. Johannin-Gilles, Phys. Scr. **14,** 273–276 (1977).
12. F. Spieweck, IEEE Trans. Instrum. Meas. **IM-29,** 361–363 (1980).
13. J. L. Hall and S. A. Lee, Appl. Phys. Lett. **29,** 367–369 (1976).
14. B. Edlén, Metrologia **2,** 71–80 (1966).