Lasers for an Optical Frequency Standard using Trapped Hg^+ Ions[1]

Brenton C. Young, Flavio C. Cruz,[2] Dana J. Berkeland,[3] Robert J. Rafac, James C. Bergquist, Wayne M. Itano, and David J. Wineland

National Institute of Standards and Technology, Boulder, Colorado 80303

Abstract. We are developing an optical frequency standard based on the narrow 281.5 nm transition of trapped $^{199}Hg^+$ ions. A major step toward the completion of this standard is the construction of an isolated high-finesse Fabry-Pérot cavity to stabilize the local oscillator. The cavity system that we have assembled has enabled the creation of an optical frequency source with good short-term stability. Eventually, this frequency source will derive long-term stability from a lock to the Hg^+ transition. We have recently demonstrated an improved linewidth of 0.6 Hz (40 s averaging time) for a 563 nm dye laser locked to our stable cavity. Additionally, we are developing solid-state laser replacements for gas and dye lasers presently used for driving 194 nm and 281.5 nm Hg^+ transitions.

INTRODUCTION

The next major advance for frequency standards probably lies in the development of optical frequency standards. Optical frequency standards are attractive since the potential fractional frequency instability of a quantum system is inversely proportional to the transition frequency. Because optical frequencies are approximately 10^5 times higher than the 9.2 GHz microwave transition used in cesium standards, much higher fractional stability might be achieved in a given measurement time.

In the 1970s, Dehmelt noted that single trapped and laser-cooled ions might be nearly ideal references for optical frequency and time standards [1,2]. High resolution is possible because perturbations can be made small and interrogation times long [1–4]. In addition, laser cooling considerably reduces first- and second-order Doppler shifts [5,6].

Several groups are developing optical frequency standards based on a variety of ions [7–20]. Among proposed standards that use trapped and laser-cooled ions, $^{199}Hg^+$ ions are attractive because they offer both a suitable microwave and optical transition. Figure 1(a) shows the $^{199}Hg^+$ electric dipole transitions at 194 nm used for laser cooling, optical pumping, and detection, and the electric quadrupole transition at 281.5 nm that is the reference for the optical frequency standard. Our group has recently demonstrated an accurate microwave frequency standard based on the 40.5 GHz, $F = 0 \rightarrow F = 1$, ground-state hyperfine splitting in trapped and cooled $^{199}Hg^+$ ions [21]. We expect to achieve significant gains in statistical precision, and likely in accuracy, for an optical frequency standard that interrogates the ultraviolet transition, with a frequency over 25 000 times that of the microwave frequency standard.

The optical standard is based on the $^2S_{1/2}\rightarrow{}^2D_{5/2}$, 281.5 nm electric-quadrupole transition [20]. An optical oscillator locked to this transition can have a fractional frequency instability approximately equal to 1×10^{-15} at 1 s even for a single laser-cooled ion. However, reaching such low instabilities requires a laser (local oscillator) whose frequency fluctuations are less than approximately 1 Hz during time intervals as long as a few seconds.

One of the main technical difficulties of working with Hg^+ is the development of reliable and economical ultraviolet laser sources. Recent advances in solid-state lasers have made possible optical sources at these

---

[1] Work of the U.S. Government, not subject to U.S. copyright.
[2] Present address: Universidade Estadual de Campinas, Campinas, SP, 13083-970, Brazil.
[3] Present address: Los Alamos National Laboratory, Los Alamos, NM 87545.

CP457, *Trapped Charged Particles and Fundamental Physics*
edited by Daniel H. E. Dubin and Dieter Schneider
1999 The American Institute of Physics 1-56396-776-6/99/$15.00

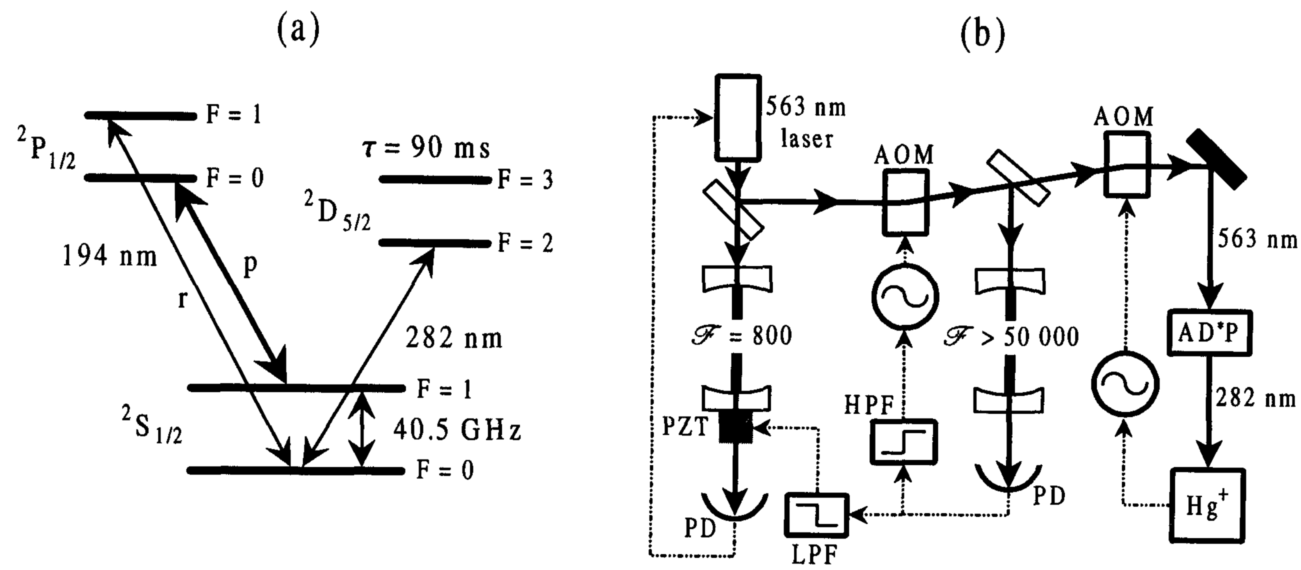

FIGURE 1. (a) Energy level diagram of $^{199}Hg^+$. We cool the ions using the $^2S_{1/2} \rightarrow {}^2P_{1/2}$ transitions at 194 nm. Because the $^2P_{1/2}$, F=0 $\rightarrow {}^2S_{1/2}$, F=0 transition is forbidden, transition p is a cycling transition. A second laser on transition r repumps atoms that decayed to $^2S_{1/2}$, F=0 after off-resonant excitation to $^2P_{1/2}$, F=1 by the first laser. (b) Simplified schematic of the proposed optical frequency standard. A dye laser is prestabilized to a Fabry-Pérot cavity ($\mathcal{F}$ = 800). Further stabilization to a much higher finesse cavity ($\mathcal{F} > 50\,000$), and eventually to a narrow transition of trapped Hg^+ ion(s) should provide a highly stable frequency source. Solid lines denote optical paths and dotted lines represent electrical connections. AD*P, deuterated ammonium dihydrogen phosphate crystal for frequency doubling, AOM, acousto-optic modulator; $\mathcal{F}$, finesse; HPF, high-pass filter; LPF, low-pass filter; PD, photodiode, PZT, piezoelectric transducer.

wavelengths with lower initial costs and operating costs, higher reliability and efficiency, and lower intrinsic noise than for Ar^+ and dye lasers. Consequently, we are developing solid-state replacements for our present laser systems.

OVERVIEW OF THE OPTICAL FREQUENCY STANDARD

Figure 1(b) shows a simplified diagram of our proposed optical frequency standard. When interrogating a narrow atomic resonance, the laser must have a frequency width narrower than the transition linewidth to prevent the frequency instability of the laser from limiting the performance of the frequency standard. Consequently, one of the major steps in the development of the optical frequency standard is the construction of an optical local oscillator with sufficient spectral purity. For the 1.7 Hz linewidth $^{199}Hg^+$ transition, the stability of the standard will not be significantly degraded if the laser linewidth is below 1 Hz for interrogation times as long as a few seconds.

A central component of this system is a high-finesse ($\mathcal{F} > 50\,000$) Fabry-Pérot cavity [20], which is described in detail in a later section. We use a dye laser at 563 nm as the optical source that is locked to this reference cavity. (The light is frequency-doubled to 281.5 nm in a crystal close to the Hg^+ trap.) Not shown in Fig. 1(b) is an iodine reference cell that we use to locate the Hg^+ transition whenever changes are made to the high-finesse reference cavity.

Rather than locking the laser directly to the high-finesse cavity, we first prestabilize it to a cavity with a finesse of approximately 800 using a Pound-Drever-Hall FM lock [22]. This prestabilization provides several advantages, including an increased locking range, a higher loop bandwidth for the lock, and improved versatility and tunability of the laser. An intracavity electro-optic modulator (EOM) in the dye laser provides high-frequency correction of laser frequency noise. A piezoelectric transducer (PZT) behind one of the dye-laser cavity mirrors eliminates long-term frequency drifts between the dye laser and the cavity. A loop bandwidth of approximately 2 MHz in this prestabilization stage narrows the dye laser short-term ($\tau < 1$ s) linewidth to approximately 1 kHz.

An optical fiber delivers light from the dye-laser table to a vibrationally isolated table that supports the high-finesse cavity. An acousto-optic modulator (AOM) mounted on the isolated table shifts the frequency of the incoming light to match a cavity resonance. Again, we implement the lock using the Pound-Drever-Hall technique. The feedback loop performs corrections at low frequencies by adjusting a PZT on the prestabilization cavity, and at higher frequencies as high as approximately 90 kHz by varying the AOM drive frequency. With the lock enabled, the light entering the cavity has a spectral width less than 1 Hz, as we demonstrate later.

Finally, the frequency-stabilized light is transported to the table holding a cryogenic Hg^+ trap. The 563 nm radiation is frequency-doubled to 281.5 nm and is focused onto the trapped ion(s). The final AOM in Fig. 1(b) shifts the frequency of the light to match the ion transition. We plan to interrogate the transition using the Ramsey technique [23], with a Ramsey time of approximately 30 ms. A digital servo loop will adjust the AOM frequency to step between both sides of the central fringe, and will periodically record the values of the center frequency [20].

SOLID-STATE LASERS

The inherent frequency stability of solid-state lasers makes them attractive for metrological applications and precision spectroscopy. In addition, a solid-state laser can be compact, reliable, and long-lived. Reliable, commercial diode lasers do not yet exist in the uv, near the transitions needed for the Hg^+ system, but high-power, near-infrared diode lasers are available. Consequently, some groups have frequency-quadrupled the output of near-infrared diode lasers that oscillate at a single frequency and in a single spatial mode to obtain cw, single-frequency uv sources [24,25]. An alternative approach is to frequency-quadruple the output of a cw, solid-state laser that is pumped with high-power multimode diode lasers, as has been done using Nd:YAG and Nd:YVO_4 lasers [26,27]. We have taken this latter approach in developing an all-solid-state laser for driving the $^{199}Hg^+$ $S-D$ transition at 281.5 nm. The solid-state generation of 194 nm radiation for the cooling transition employs a diode-pumped solid-state laser in addition to sum-frequency mixing. These two laser systems are described in the following subsections.

Nd:FAP laser

For the 281.5 nm light source, we plan to replace a dye laser and its multiline Ar^+ pump laser with a frequency-doubled Nd^{3+}-doped fluorapatite (Nd:FAP) laser [28]. Nd:FAP has a lasing transition at 1.126 μm that, when frequency-quadrupled, coincides with the Hg^+ transition at 281.5 nm. The major difficulty in designing this laser is that Nd:FAP has a much stronger transition nearby at 1.063 μm [29] that must be suppressed by the laser optics. With 680 mW of diode pump light at 808 nm, the Nd:FAP output power is approximately 90 mW at 1.126 μm. Frequency doubling in $KNbO_3$ gives approximately 5 mW at 563 nm.

For the second stage of harmonic generation, we use deuterated ammonium dihydrogen phosphate (AD*P) to frequency-double 563 nm radiation to 281.5 nm [30]. Since less than 1 pW can be enough to saturate the narrow $S-D$ transition [30], we simply frequency-double the radiation at 563 nm in a single-pass configuration. approximately 25 nW is generated at 281.5 nm for 5 mW of input power at 563 nm.

Because the free-running frequency instability of this laser is dominated by low-frequency acoustical and mechanical noise, only a moderate-speed servo system is needed to lock the laser frequency tightly to the resonance of a high-finesse cavity [20,31]. In addition, since the frequency of the Nd:FAP laser is quadrupled to reach the atomic transition, this facilitates the first steps in a frequency chain from the optical to the microwave. We have demonstrated tunability of this laser through the Hg^+ transition. Poor stability of the pump-diode output mode, however, has forced us to use the original dye laser source for the experimental work described in the remainder of this paper.

Yb:YAG laser

Currently, the 194 nm cooling light is generated using a single-mode Ar^+ laser at 515 nm that is frequency-doubled in β-barium borate (BBO) to 257 nm and is then sum-frequency mixed in BBO with a diode source at

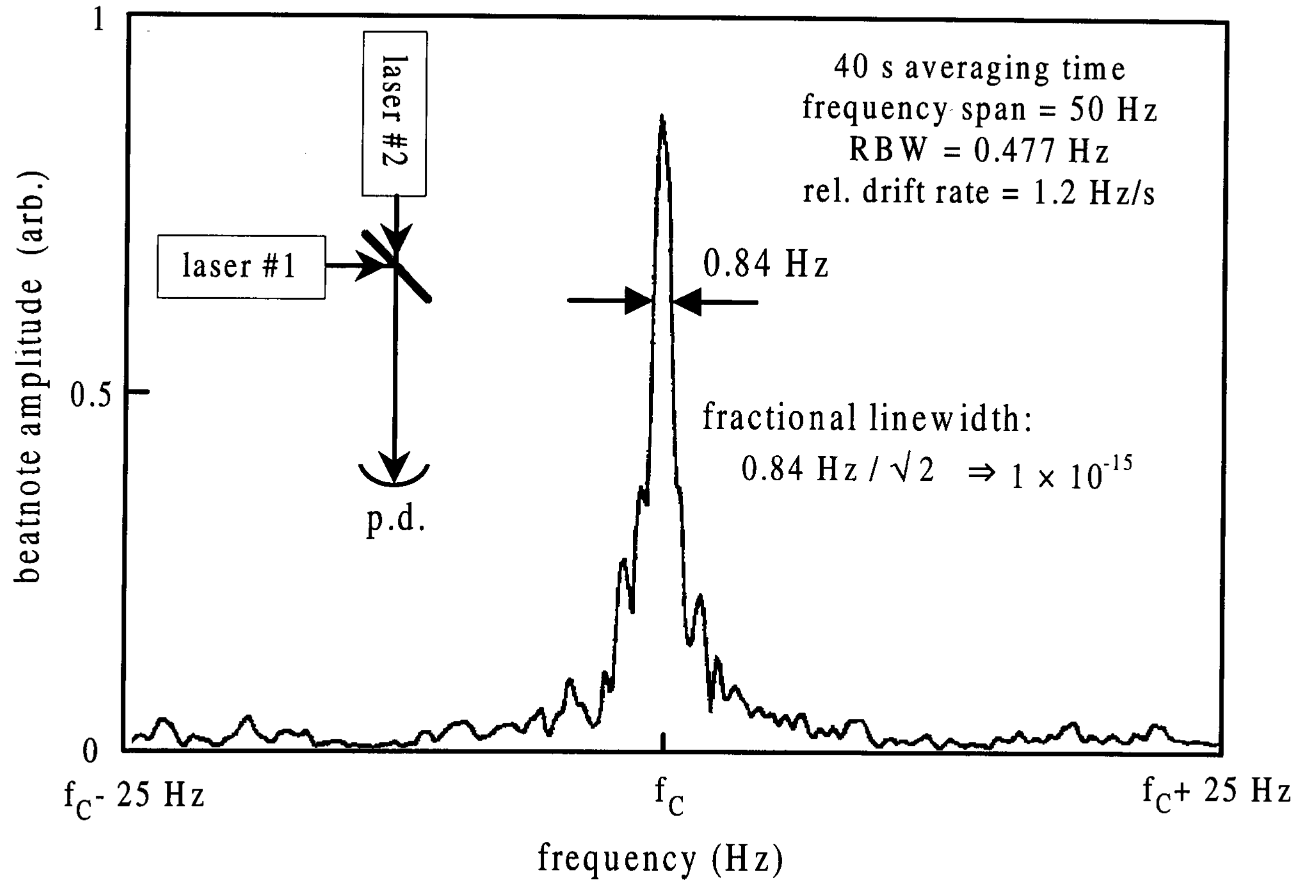

FIGURE 2. Amplitude spectrum of the beat note between two laser beams stabilized to two independent cavities. The dashed line shows the -3 dB level. The averaging time is 40 s. A nearly uniform relative cavity drift of 1.2 Hz/s is suppressed by mixing the beat note with a swept synthesizer.

792 nm to produce light at 194 nm [32]. We plan to replace the Ar^+ laser with a Yb:YAG laser at 1.03 μm [33]. With 3 W of diode pump power at 941 nm, the Yb:YAG output power is 1.2 W at 1.03 μm. We have frequency-doubled the Yb:YAG laser output in $KNbO_3$ to obtain over 400 mW at 515 nm, which should be sufficient power to replace the Ar^+ laser. At high optical powers, the frequency-doubling conversion efficiency is limited by losses from blue-light-induced infrared absorption [34]. We anticipate achieving a better conversion efficiency by frequency-doubling with lithium triborate (LBO) instead of $KNbO_3$.

HIGH-FINESSE REFERENCE CAVITY

To achieve a laser linewidth of <1 Hz for the source driving the Hg^+ ion transition, we start with a high-finesse cavity that has intrinsically low sensitivity to temperature variations, and then take great care to protect it from environmental perturbations. The separation of the cavity mirrors is set by optically contacting the mirrors to the ends of a hollow cylinder made from a low-thermal-expansion material. The mirror substrates are made of the same material as the cylinder. The cavity is supported inside an evacuated chamber by two thin wires. Keeping the cavity under vacuum both avoids pressure shifts of the cavity resonance and thermally insulates it from the environment. The temperature of the vacuum chamber is held near 30 °C, which is the point of zero coefficient of expansion for the cavity material. We protect the cavity from seismic noise by mounting it on a passively isolated optical table. The table is suspended by strands of surgical tubing approximately 3 m long. The fundamental vibrational mode of the suspension has a frequency of ≈0.3 Hz, which provides an isolation from floor noise that exceeds a factor of 50 in noise amplitude already at 3 Hz (some viscous damping is used). To prevent the coupling of acoustic noise into the cavity, we enclose the optical table in a wooden box lined internally with lead foam [35].

The intracavity light heats the mirror coatings, thereby shifting the cavity resonance. To hold this shift at a reasonable value, we couple only approximately 100 μW of 563 nm light into the cavity. Furthermore, controlling the optical power in the cavity stabilizes this power shift. Active control of the rf power driving the AOM stabilizes the output power from the cavity to $\approx$0.1%.

To characterize the cavity's short-term stability performance without referencing to Hg^+, we constructed a second cavity and isolated table similar to that described above. Figure 2 shows the spectrum of the beat note between two independent laser beams stabilized to the two cavities. A nearly uniform relative cavity drift of $\approx$1 Hz/s is suppressed by mixing the beat note with a swept synthesizer. The width of the spectrum at its half-power point is 0.8 Hz (40 s averaging time). This implies that at least one of the lasers has a frequency width less than 0.6 Hz at 563 nm, corresponding to a fractional linewidth of only 1×10^{-15}. This is roughly 40 times better than previous results with only one cavity well isolated from vibrations [20], and may represent the smallest fractional linewidth ever recorded in the optical regime.

CONCLUSIONS

We have demonstrated an optical local oscillator suitable for development of a Hg^+ optical frequency standard at 281.5 nm. The frequency source has a linewidth of less than 0.6 Hz at 563 nm (40 s averaging time), corresponding to a fractional linewidth of 1×10^{-15}. We have described work on solid-state laser replacements for gas and dye lasers presently used in the trapped Hg^+ work. Future work will involve collaboration with other researchers at NIST and JILA to develop a frequency chain for translating the frequency and the stability of our standard into the microwave regime. This work is supported by ONR and NIST.

REFERENCES

1. Dehmelt, H. G., *Bull. Am. Phys. Soc.* **18**, 1521 (1973).
2. Dehmelt, H. G., *IEEE Trans. Instrum. Meas.* **IM-31**, 83–87 (1982).
3. Wineland, D. J., *et al.*, *J. Phys. (Paris)* **42**, C8-307–C8-313 (1981).
4. Fisk, P. T. H., *Rep. Prog. Phys.* **60**, 761–817 (1997).
5. Wineland, D., and Dehmelt, H., *Bull. Am. Phys. Soc.* **20**, 637 (1975).
6. Wineland, D. J., and Itano, W. M., *Phys. Today* **40**, 34–40 (1987).
7. Bergquist, J. C., ed., *Proceedings of the Fifth Symposium on Frequency Standards and Metrology*, Singapore: World Scientific, 1996.
8. Sugiyama, K., Sasaki, K., Wakita, A., and Yoda, J., "Progress toward high-resolution spectroscopy of the $^2S_{1/2}$-$^2D_{5/2}$ transition of laser-cooled trapped Yb^+," in *International Workshop on Current Topics of Laser Technology*, 1998, p. 56.
9. Taylor, P., Roberts, M., Barwood, G. P., and Gill, P., *Opt. Lett.* **23**, 298–300 (1998).
10. Engelke, D., and Tamm, C., *Europhys. Lett.* **33**, 347–352 (1996).
11. Barwood, G. P., *et al.*, *Opt. Commun.* **151**, 50–55 (1998).
12. Bernard, J. E., Marmet, L., and Madej, A. A., *Opt. Commun.* **150**, 170–174 (1998).
13. Madej, A. A., *et al.*, "Precision absolute frequency measurements with single atoms of Ba^+ and Sr^+," in *Proceedings of the Fifth Symposium on Frequency Standards and Metrology*, 1996, pp. 165–170.
14. Urabe, S., *et al.*, *Appl. Phys. B* **67**, 223–227 (1998).
15. Knoop, M., Vedel, M., and Vedel, F., *Phys. Rev. A* **58**, 264–269 (1998).
16. Fermigier, B., *et al.*, *Opt. Commun.* **153**, 73–77 (1998).
17. Peik, E., *et al.*, "Towards an optical clock with a laser-cooled indium ion," in *International Workshop on Current Topics of Laser Technology*, 1998, pp. 23–24.
18. Nagourney, W., Burt, E., and Dehmelt, H. G., "Optical frequency standard using individual indium ions," in *Proceedings of the Fifth Symposium on Frequency Standards and Metrology*, 1996, pp. 341–346.
19. Yu, N., Dehmelt, H., and Nagourney, W., *Proc. Natl. Acad. Sci. U.S.A.* **89**, 7289 (1992).
20. Bergquist, J. C., Itano, W. M., and Wineland, D. J., "Laser stabilization to a single ion," in *Frontiers in Laser Spectroscopy*, 1994, pp. 359–376.
21. Berkeland, D. J., *et al.*, *Phys. Rev. Lett.* **80**, 2089–2092 (1998).
22. Drever, R. W. P., *et al.*, *Appl. Phys. B* **31**, 97–105 (1983).
23. Ramsey, N. F., *Phys. Rev.* **78**, 695–699 (1950).

24. Zimmermann, C., Vuletic, V., Hemmerich, A., and Hänsch, T. W., *Appl. Phys. Lett.* **66**, 2318–2320 (1995).
25. Matsubara, K., *et al.*, *Appl. Phys. B* **67**, 1–4 (1998).
26. Hollemann, G., Peik, E., and Walther, H., *Opt. Lett.* **19**, 192–194 (1994).
27. Kondo, K., *et al.*, *Opt. Lett.* **23**, 195–197 (1998).
28. Cruz, F. C., Young, B. C., and Bergquist, J. C., "Diode-pumped Nd:FAP laser at 1.126 μm: a possible local oscillator for a Hg^+ optical frequency standard," *Appl. Opt.* (to be published).
29. Ohlmann, R. C., Steinbruegge, K. B., and Mazelsky, R., *Appl. Opt.* **7**, 905–914 (1968).
30. Bergquist, J. C., Hulet, R. G., Itano, W. M., and Wineland, D. J., *Phys. Rev. Lett.* **57**, 1699–1702 (1986).
31. Zhu, M., and Hall, J. L., "Frequency stabilization of tunable lasers," in *Atomic, Molecular, and Optical Physics: Electromagnetic Radiation*, 1997, pp. 103–136.
32. Berkeland, D. J., Cruz, F. C., and Bergquist, J. C., *Appl. Opt.* **36**, 4159–4162 (1997).
33. Fan, T. Y., and Ochoa, J., *IEEE Photon. Technol. Lett.* **7**, 1137–1138 (1995).
34. Mabuchi, H., Polzik, E. S., and Kimble, H. J., *J. Opt. Soc. Am. B* **11**, 2023–2029 (1994).
35. Hils, D., Faller, J. E., and Hall, J. L., *Rev. Sci. Instrum.* **57**, 2532–2534 (1986).