Diode-pumped Nd:FAP laser at 1.126 μm: a possible local oscillator for a Hg^+ optical frequency standard

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We report the efficient operation of a continuous-wave, single-frequency, diode-pumped Nd:FAP laser at 1.126 μm. When frequency quadrupled, such a laser might be used as a local oscillator for an optical frequency standard based on the single-photon $^2S_{1/2}$–$^2D_{5/2}$ electric quadrupole transition of a trapped and laser-cooled $^{199}Hg^+$ ion. Since the frequencies of the atomic transition and the laser are harmonically related, this scheme helps to simplify the measurement of the *S–D* clock transition frequency by a phase-coherent chain to the cesium primary frequency standard.

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Trapped and laser-cooled ions have great potential for optical frequency and time standards.[1–6] High resolution is possible because perturbations can be made small[7] and interrogation times long.[8,9] In addition, laser cooling reduces first- and second-order Doppler shifts to extremely low levels.[10] Among proposed standards that use trapped and laser-cooled ions,[6] a $^{199}Hg^+$ ion standard is attractive because it offers both a microwave transition[9] and a narrow optical transition that promise high performance. The optical standard is based on the $^2S_{1/2}$–$^2D_{5/2}$, 281.5-nm electric quadrupole transition.[11] Optical frequency standards are attractive since the potential fractional frequency instability of a standard is inversely proportional to its frequency. An optical clock can have a fractional frequency instability less than 1×10^{-15} at 1 s even for a single laser-cooled ion. However, for reaching such low instabilities it is necessary to have a laser (local oscillator) whose frequency fluctuations are less than 1 Hz for times as long as a few seconds.

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 transition 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.[12,13] An alternative approach is to frequency-quadruple the output of a cw, solid-state laser that is pumped with high-power multimode diode lasers, such as has previously been done with Nd:YAG and Nd:YVO_4 lasers.[14] Here we report the development of a diode-pumped solid-state Nd:FAP laser that operates in a single spatial and single-frequency mode at 1.126 μm. The frequency of the output radiation from this laser was quadrupled and might be used as a local oscillator in an optical frequency standard based on the *S–D* quadrupole transition of trapped and laser-cooled $^{199}Hg^+$ ions at 282 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.[11,15] In addition, since the frequency of the Nd:FAP laser is quadrupled to reach the atomic transition, we have completed the first steps in a frequency chain from the optical to the microwave.

The spectral analysis and lasing performance of Nd:FAP at 1.06 μm have been reported elsewhere.[16,17] The absorption spectrum of Nd:FAP, like Nd:YAG, has a strong line near 808 nm, approximately 2.5 nm wide, which is well suited to optical pumping by a diode laser. In addition to a strong emission peak at 1.06 μm, weaker features near 1.12 and 0.93 μm are also observed. We make the Nd:

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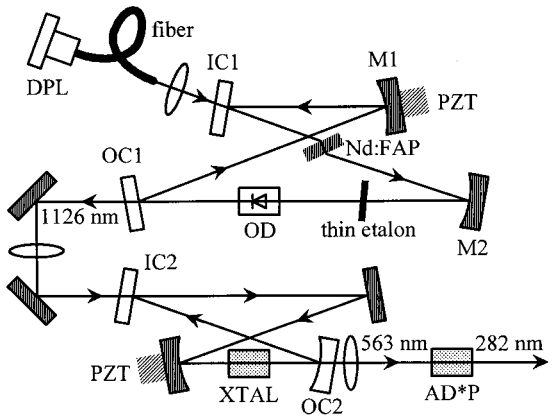

Fig. 1. Experimental setup for the frequency-quadrupled, diode-pumped Nd:FAP laser: DPL, diode pump laser at 808 nm; IC1, flat input coupler for the Nd:FAP laser (HR at 1126 nm, $T \approx 90\%$ at 808 nm); PZT, piezoelectric transducer; M1 and M2, 15-cm radius-of-curvature mirrors (HR at 1126 nm); OD, optical diode; OC1, flat output coupler ($T = 0.4\%$ at 1126 nm, $T \approx 50\%$ at 1064 nm); IC2, flat input coupler for doubling cavity ($T = 1.7\%$ at 1126 nm); XTAL, $KNbO_3$ or $LiNbO_3$ doubling crystal; OC2, flat output coupler (HR at 1126 nm, $T \approx 80\%$ at 563 nm for $KNbO_3$ or 94% for $LiNbO_3$); AD*P, deuterated ammonium dihydrogen phosphate.

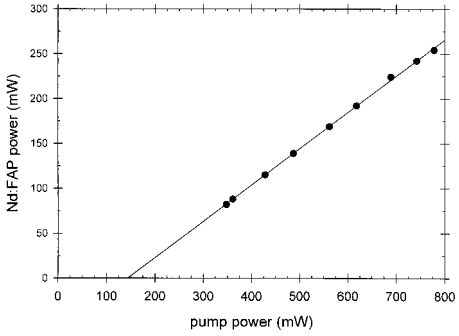

Fig. 2. Output power of the Nd:FAP laser at 1.126 μm as a function of pump power from a Ti:sapphire laser.

FAP laser oscillate at 1.126 μm by strongly suppressing the higher-gain oscillation at 1.06 μm. Our crystal is cut, polished, and oriented at Brewster's angle to avoid the need for any antireflection coatings. The *c* axis lies in a plane parallel to the polished surfaces and is oriented perpendicular to the polarization of the radiation of both the pump laser and the Nd:FAP laser. The thickness of the crystal is 2 mm, and the diameter is 6.3 mm. The Nd concentration gives approximately 80% absorption of the light from a diode laser at 808 nm. Absorption is nearly 100% when a narrow-band Ti:sapphire laser is used. We largely eliminate thermal problems for pump powers as high as 1 W by heat sinking the crystal in a tightly fitting copper support piece. Maximum output powers are achieved for cavity waists ω_0 between 80 and 100 μm. The diode laser used for pumping emits as high as 1 W at 808 nm, which is coupled into a fiber with a 100-μm core diameter. Approximately 750 mW is available after the fiber. A 3.5-cm focal-length lens collimates the beam diverging from the fiber, and a 6.7-cm lens focuses the light through the Nd:FAP crystal.

The 1.126-μm laser is built as a ring cavity formed by two flat mirrors (input and output couplers) and two 15-cm radius-of-curvature mirrors that are highly reflecting (HR) at 1.126 μm (M1 and M2 in Fig. 1). The Nd:FAP crystal is positioned midway between the 15-cm mirrors at a nearly optimum cavity waist of $\omega_0 \approx 80$ μm. The flat input coupler IC1 transmits 90% of the pump light at 808 nm and is HR at 1.126 μm. The output coupler OC1 was coated to transmit 0.4% of the radiation at 1.126 μm and 50% at 1.06 μm, which eliminates lasing on the strong line at 1.06 μm. The pump beam is focused into the crystal with a beam waist of $\omega_0 \approx 100$ μm. The astigmatism introduced by the crystal is compensated by the off-axis orientation of the 15-cm mirrors. Unidirectional oscillation is enforced by an optical diode formed by a terbium–gallium garnet Faraday rotator and a quartz compensating plate. The laser oscillates at 1.126 μm in a single-frequency TEM_{00} mode without any frequency-selective elements inside the cavity. However, a thin etalon improves the mode stability and makes it possible to tune the wavelength. Tuning of the laser from approximately 1.118 to 1.128 μm was verified by use of an interferometric wavemeter and an optical spectrum analyzer. When frequency quadrupled, the Nd:FAP laser reaches the Hg^+ *S–D* transition at 281.5 nm. The tuning range also includes the $A_2\,\Sigma(n'' = 1) \leftarrow X_2\,\Pi(n' = 0)$ transition of OH molecules at 281.93 nm, which could possibly be used for remote sensing of OH in the atmosphere.[18]

The output power of the 1.126-μm laser as a function of pump power from the Ti:sapphire laser is plotted in Fig. 2. The slope efficiency is approximately 40%. For an input power of 780 mW, an output power of 260 mW is achieved, corresponding to an overall efficiency of 33%. When pumped by the diode laser, the efficiency is reduced to approximately 20% owing to the poorer spatial mode quality of the diode laser beam and to the lower absorption of the pump power by the crystal.

We double the fundamental radiation at 1126 nm using either $KNbO_3$ or $LiNbO_3$. Noncritically phase-matched harmonic generation in $KNbO_3$, which would be highly efficient, is predicted to occur for 1126-nm light at a phase-matching temperature[19] of 268 °C. Unfortunately, this is precluded since $KNbO_3$ experiences a phase transition near 220 °C. However, less efficient type-I angle-tuned critical phase matching at room temperature is still possible for *b*-cut $KNbO_3$ at a phase-matching angle of $\theta_{pm} \approx 23.9°$. The calculated effective nonlinear coefficient d_{eff} is 11.8 pm/V and the walkoff angle is approximately 60 mrad, which implies an efficiency per unit crystal length of $2 \times 10^{-3}\ W^{-1}\ cm^{-1}$. For 2-mm-long crystals we measure single-pass efficiencies η slightly higher than $1 \times 10^{-4}\ W^{-1}$ under optimum focusing conditions, which is less than predicted. We increase the harmonic power by using

antireflection-coated $KNbO_3$ in an external build-up cavity (Fig. 1). The cavity is locked to resonance with the frequency of the laser by the Hänsch–Couillaud polarization technique.[20] The power-enhancement cavity is formed by two flat mirrors and two spherical mirrors (7.5-cm radius of curvature) that produce a waist of 14 μm, nearly optimum[21] for the 2-mm-long $KNbO_3$. The input coupler IC2 transmits 1.7% of the radiation at 1126 nm. This transmittance gives nearly 100% coupling into the cavity for 100 mW of incident radiation when light at the second harmonic is generated. The output coupler OC2 is one of the 7.5-cm mirrors, which is HR at 1126 nm and 80% transmitting at 563 nm. The other mirrors are HR at 1126 nm. The crystal is positioned in the tight focus between the two curved mirrors, and the fundamental beam is mode matched into the softer waist between the two flat mirrors. We keep the cavity angles as small as possible to minimize the astigmatism introduced by the curved mirrors. The cavity losses (excluding the input coupler) are 0.1% and the crystal losses are 0.3%, as determined from the measured finesse and build-up factors of the cavity with and without the crystal. The crystal losses are dominated by surface scattering. For an input power of 100 mW at 1126 nm and an enhancement factor of 150, we achieve an output power of 10 mW at 563 nm.

We also verified that generation of the second harmonic of 1126-nm radiation is possible in congruent $LiNbO_3$ at $T_{pm} \approx 142$ °C. At this temperature we observe no indication of optical damage. At 1126 nm, d_{eff} is 4.3 pm/V, which gives $\eta \approx 1.9 \times 10^{-3}\ W^{-1}$ for our 11.3-mm-long crystal. However, we measure a single-pass efficiency of only $8.0 \times 10^{-4}\ W^{-1}$ for optimum focusing conditions ($\omega_0 \approx 20$ μm). We again use a power-enhancement cavity, but the size of our oven restricts us to focusing mirrors with longer radii of curvature. Hence the waist in the crystal is larger than optimum, limiting η to approximately $3.7 \times 10^{-4}\ W^{-1}$. The power-enhancement factor in the presence of the crystal is only 76, largely because of scattering from the Brewster-cut crystal surfaces. Even so, we again attain an output power of approximately 10 mW at 563 nm. This corresponds to approximately 22 mW of harmonic radiation generated in the crystal when the power is corrected for the 94% transmittance of the output coupler and the 60% transmittance of the Brewster face of the crystal. Since the usable power is nearly the same in both cases, we find it easier and more practical to use $KNbO_3$.

For the second stage of harmonic generation we use deuterated ammonium dihydrogen phosphate (AD*P) to double 563 to 281.5 nm.[22] Since less than 1 pW can be enough to saturate the narrow S–D transition,[22] we simply double the radiation at 563 nm in a single-pass configuration (Fig. 1). Type-I noncritical phase matching is possible for doubling 563 nm with AD*P at $T_{pm} \approx 128$ °C. The measured single-pass efficiency is approximately $10^{-3}\ W^{-1}$ for our 3-cm-long crystal. Approximately 0.1 μW is generated at 281.5 nm for 10 mW of input power at 563 nm. If more power is needed, then another option could be β-barium borate (BBO) in an external build-up cavity. Type-I angle-tuned critical phase matching at $\theta_{pm} \approx 44°$ is possible with $d_{eff} \approx 1.64$ pm/V. The walkoff angle is 77 mrad, which gives $\eta \approx 1.3 \times 10^{-4}\ W^{-1}$ for a 6-mm-long crystal.

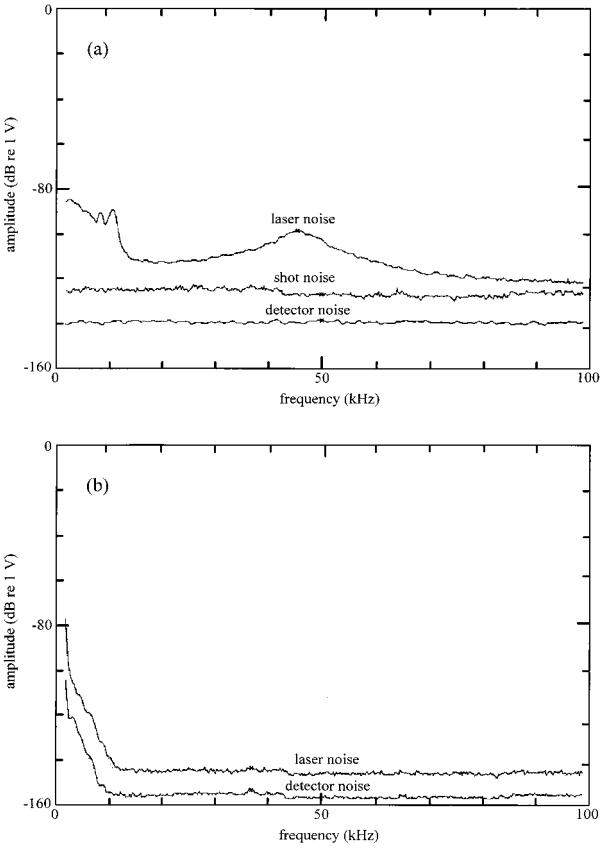

Fig. 3. (a) AM and (b) FM noise spectral densities in a 1-kHz bandwidth for the Nd:FAP laser at 563 nm. The peak at approximately 50 kHz in (a) caused by relaxation oscillations.

Figure 3 shows the power spectral densities of AM and FM noise for the radiation from the Nd:FAP laser at 563 nm. Also shown are the shot noise and detector noise. For the AM noise measurements part of the laser power is directed onto a fast photodetector. The amplitude noise spectrum of the Nd:FAP laser is obtained by a fast Fourier transform of the detected signal. The FM noise measurements are similar, except that a Fabry–Perot cavity is used as a frequency discriminator, converting frequency fluctuations into intensity fluctuations. The spectral distributions of amplitude and frequency noise of the Nd:FAP laser are dominated by contributions at low frequencies (<12 kHz). The pump diode laser, the elements in the Nd:FAP laser cavity, and the components in the external doubling cavity were all mounted in aluminum supports connected by Invar bars to improve the passive stability. A broad peak caused by relaxation oscillations at several tens of kilohertz (depending on the pump power) is observed in the spectral density of AM noise, which is shot-

noise limited above 100 kHz. The relaxation-oscillation peak is nearly absent in the FM spectral density at 563 nm. Both the AM and FM noise are lower when the Nd:FAP laser is diode pumped than when it is pumped by a Ti:sapphire laser. The free-running rms frequency excursions of the Nd:FAP laser are measured to be smaller than 100 kHz for periods of a few seconds. Therefore we anticipate that a moderate-speed servo system would be sufficient to lock its frequency to the resonance of a quiet reference cavity. When the frequency of the laser is locked to the cavity, one can measure the laser linewidth by heterodyning two independent systems[11] and by probing the narrow S–D atomic transition in $^{199}Hg^{+}$. The quadrupled Nd:FAP laser should also facilitate the implementation of a frequency chain to the cesium standard at 9.2 GHz. One can directly compare the Nd:FAP fundamental frequency to a secondary frequency standard such as the methane-stabilized He–Ne laser at 3.39 μm by using a recently demonstrated 3:1 optical divider.[23]

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References and Notes

1. H. G. Dehmelt, "Mono-ion oscillator as potential ultimate laser frequency standard," IEEE Trans. Instrum. Meas. **31,** 83–87 (1982).
2. E. Peik, G. Hollemann, and H. Walther, "Laser cooling and quantum jumps of a single indium ion," Phys. Rev. A **49,** 402–408 (1994).
3. P. Taylor, M. Roberts, G. P. Barwood, and P. Gill, "Combined optical–infrared single-ion frequency standard," Opt. Lett. **23,** 298–300 (1998).
4. L. Marmet, A. A. Madej, K. J. Siemsen, J. E. Bernard, and B. G. Whitford, "Precision frequency measurement of the $^2S_{1/2}$–$^2D_{5/2}$ transition of Sr^{+} with a 674-nm diode laser locked to an ultrastable cavity," IEEE Trans. Intrum. Meas. **46,** 169–173 (1997).
5. S. Urabe, M. Watanabe, H. Imajo, and K. Hayasaka, "Laser cooling of trapped Ca^{+} and measurement of the 3 $^2D_{5/2}$ state lifetime," Opt. Lett. **17,** 1140–1142 (1992).
6. See, for example, *Proceedings of the Fifth Symposium on Frequency Standards and Metrology*, J. C. Bergquist, ed. (World Scientific, Singapore, 1996).
7. D. J. Wineland, J. C. Bergquist, R. E. Drullinger, H. Hemmati, W. M. Itano, and F. L. Walls, "Laser cooled, stored ion experiments at NBS and possible applications to microwave and optical frequency standards," J. Phys. **42,** C8-307–C8-313 (1981).
8. J. J. Bollinger, J. D. Prestage, W. M. Itano, and D. J. Wineland, "Laser-cooled-atomic frequency standard," Phys. Rev. Lett. **54,** 1000–1003 (1985); J. J. Bollinger, D. J. Heinzen, W. M. Itano, S. L. Gilbert, and D. J. Wineland, "A 303-MHz frequency standard based on trapped Be^{+} ions," IEEE Trans. Instrum. Meas. **40,** 126–128 (1991).
9. D. J. Berkeland, J. D. Miller, J. C. Bergquist, W. M. Itano, and D. J. Wineland, "Laser-cooled mercury ion frequency standard," Phys. Rev. Lett. **80,** 2089–2092 (1998).
10. D. J. Wineland, J. C. Bergquist, W. M. Itano, F. Diedrich, and C. S. Weimer, "Frequency standards in the optical spectrum," in *The Hydrogen Atom*, G. F. Bassani, M. Inguscio, and T. W. Hänsch, eds. (Springer-Verlag, Berlin, 1989), pp. 123–133.
11. J. C. Bergquist, W. M. Itano, and D. J. Wineland, "Laser stabilization to a single ion," in *Frontiers in Laser Spectroscopy*, T. W. Hänsch and M. Inguscio, eds. (North Holland, Amsterdam, 1994), pp. 359–376.
12. C. Zimmermann, V. Vuletic, A. Hemmerich, and T. W. Hänsch, "All solid state laser source for tunable blue and ultraviolet radiation," Appl. Phys. Lett. **66,** 2318–2320 (1995).
13. K. Matsubara, U. Tanaka, H. Imajo, K. Hayasaka, R. Ohmukai, M. Watanabe, and S. Urabe, "An all-solid-state tunable 214.5-nm continuous-wave light source by using two-stage frequency doubling of a diode laser," Appl. Phys. B **67,** 1–4 (1998).
14. See, for example, G. Hollemann, E. Peik, and H. Walther, "Frequency-stabilized diode-pumped Nd:YAG laser at 946 nm with harmonics at 473 and 237 nm," Opt. Lett. **19,** 192–194 (1994); K. Kondo, M. Oka, H. Wada, T. Fukui, N. Umezu, K. Tatsuki, and S. Kubota, "Demonstration of long-term reliability of a 266-nm, continuous-wave, frequency-quadrupled solid-state laser using β-BaB_2O_4," Opt. Lett. **23,** 195–197 (1998).
15. M. Zhu and J. L. Hall, "Frequency stabilization of tunable lasers," in *Experimental Methods in the Physical Sciences*, F. B. Dunning and R. G. Hulet, eds. (Academic, San Diego, 1996), Vol. 29C, pp. 103–136, and references therein.
16. R. C. Ohlmann, K. B. Steinbruegge, and R. Mazelsky, "Spectroscopic and laser characteristics of neodymium-doped calcium fluorophosphate," Appl. Opt. **7,** 905–914 (1968).
17. X. X. Zhang, A. B. Villaverde, M. Bass, G. Lutts, and B. H. T. Chai, "Spectroscopy and lasing performance of Nd^{3+} doped $Ca_5(PO_4)_3F$," in *Growth, Characterization, and Applications of Laser Host and Nonlinear Crystals II*, B. H. T. Chai, ed., Proc. SPIE **1863,** 35–38 (1993).
18. D. D. Davis, W. S. Heaps, D. Philen, M. Rodgers, T. McGee, A. Nelson, and A. J. Moriarty, "Airborne laser induced fluorescence system for measuring OH and other trace gases in the parts-per-quadrillion to parts-per-trillion range," Rev. Sci. Instrum. **50,** 1505–1516 (1979); G. L. Vaghjiani and A. R. Ravishankara, "Kinetics and mechanism of OH reaction with CH_3OOH," J. Phys. Chem. **93,** 1948–1959 (1989).
19. I. Biaggio, P. Kerkoc, L.-S. Wu, P. Günter, and B. Zysset, "Refractive indices of orthorhombic $KNbO_3$. II. Phase-matching configurations for nonlinear-optical interactions," J. Opt. Soc. Am. B **9,** 507–517 (1992).
20. T. W. Hänsch and B. Couillaud, "Laser frequency stabilization by polarization spectroscopy of a reflecting reference cavity," Opt. Commun. **35,** 441–444 (1980).
21. G. D. Boyd and D. A. Kleinman, "Parametric interaction of focused Gaussian light beams," J. Appl. Phys. **39,** 3597–3639 (1968).
22. J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, "Observation of quantum jumps in a single atom," Phys. Rev. Lett. **57,** 1699–1702 (1986).
23. O. Pfister, M. Mürtz, J. S. Wells, L. Hollberg, and J. T. Murray, "Division by 3 of optical frequencies by use of difference-frequency generation in noncritically phase-matched RbTiO-AsO_4," Opt. Lett. **21,** 1387–1389 (1996).