Absolute frequency stabilization of diode-laser-pumped Nd:YAG lasers to hyperfine transitions in molecular iodine

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Diode-laser-pumped Nd:YAG lasers were frequency stabilized by locking their frequency-doubled output to Doppler-free absorption lines of the $^{127}I_2$ molecule. The successive two-sample deviation of the beat frequency between two independent iodine-stabilized lasers is less than 650 Hz, or 2.3×10^{-12} of the laser frequency, for averaging times between 24 and 80 s.

Owing to their inherently low-frequency noise, small size, high reliability, and potentially high power, monolithic diode-laser-pumped solid-state lasers are attractive sources for applications that require high spectral purity, such as optical frequency standards, metrology, space-based measurements,[1] and deep-space optical coherent communication.[1] However, the long-term drift of the laser frequency, typically at a rate of several megahertz per minute,[2] must be improved before those applications become possible. By externally doubling the laser frequency and locking it to the Doppler-free absorption lines of the $^{127}I_2$ molecule, we have reduced significantly the long-term frequency drift of the lasers, thus opening the possibility of using diode-laser-pumped solid-state lasers in such applications.

The spectrum of the $X \rightarrow B$ transitions in molecular iodine has more than 20,000 absorption lines in the visible, some of them near the second harmonic of Nd:YAG at 532 nm. This spectrum has been thoroughly characterized,[3] and a variety of gas lasers[4] have been frequency stabilized to iodine transitions. Several frequency-doubled flash-lamp-pumped Nd:YAG laser systems[5–7] have been used to study the hyperfine transitions near 532 nm: Kruzhalov *et al.*[5] used an intracavity-doubled Nd:YAG laser, Esherick and Owyoung[6] used a pulsed injection-seeded laser, and Schiller *et al.*[7] used an injection-locked high-power cw laser. Kruzhalov *et al.*[5] have also stabilized the laser frequency by locking to a Doppler-free line. Direct frequency stabilization of the 1064-nm radiation of a Nd:YAG laser to hyperfine transitions of molecular cesium heated to 220°C was demonstrated by Mak *et al.*,[8] and a fractional stability of 6×10^{-11} was obtained for a measurement period of 1 s. The Nd:YAG laser has also been stabilized by using a high-finesse Fabry–Perot cavity,[9,10] and a relative stability at the subhertz level was obtained between two lasers locked to a single cavity.[10] However, locking to an external cavity does not guarantee an improved long-term absolute stability, since the resonant frequency of the cavity itself may drift, and therefore stabilization to an absolute standard is required.

The experimental setup for frequency locking the laser to iodine is shown in Fig. 1. We use two Lightwave Electronics Model 122 Nd:YAG monolithic diode-laser-pumped nonplanar ring lasers,[2] emitting 300 mW of power at 1064 nm. The laser frequency can be tuned over 30 GHz by changing the laser crystal temperature between 20°C and 50°C, with a relatively slow response time (~1 s). Fast frequency tuning (a bandwidth exceeding 100 kHz) over ~100 MHz is achieved by applying a voltage to a piezoelectric transducer (PZT) that is bonded to the Nd:YAG laser crystal. Single-mode operation without mode hops can be maintained over a continuous tuning range of 8 GHz.

The laser frequency is externally doubled by using an 8-mm-long $MgO{:}LiNbO_3$ monolithic crystal, heated to its phase-matching temperature (~108°C). The triangular ring path inside the doubler is bounded by two curved ($r = 7$ mm) dielectrically coated surfaces that reflect the pump radiation while transmitting the second-harmonic radiation and by a flat total internal reflecting surface. The curved input surface has a 96.9% reflection at 1064 nm, whereas the curved output surface is a high reflector at 1064 nm and has a 89.5% transmission at 532 nm. To provide high conversion efficiency[11] with a fixed output power, the resonator is frequency locked to the pump frequency, with a slight detuning, by a servo that controls the $LiNbO_3$ resonator temperature. The high-temperature side of the doubler resonant fringe is locked to the laser frequency by keeping a fixed level of the transmitted second-harmonic power. This fringe side locking is inherently stable since increasing the doubler temperature leads to a reduced coupling of the fundamental field into the resonator, and thus to less internal heating, which counteracts the initial temperature change. Additional stability is obtained by using the doubler on the low-temperature side of the phase-matching temperature curve.[12] Thus, by operating on the stable side of the fringe and on the stable side of the phase-matching temperature curve, one can lock the resonant doubler to the laser using a relatively low-bandwidth temperature control loop. The oper-

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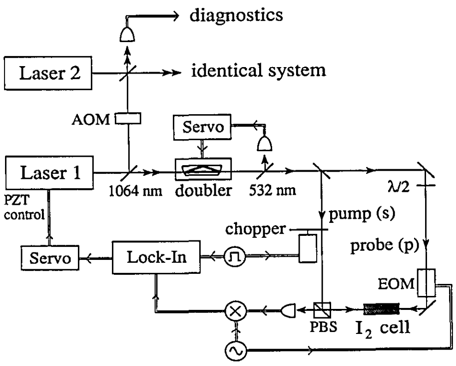

Fig. 1. Experimental setup for locking the lasers to the Doppler-free transition of iodine. PBS, polarizing beam splitter; AOM, acousto-optic modulator; EOM, electro-optic modulator.

ating output powers at 532 nm for the two systems are 25 and 5 mW (owing to a lower conversion efficiency of the second doubler).

The 532-nm output of the doubler is the source for FM Doppler-free saturation spectroscopy[13] of iodine. The phase-modulated probe beam and the pump beam counterpropagate through a 10-cm-long iodine cell. The pump/probe intensity ratio is 90/10, and the beam diameter inside the cell is ~0.2 mm. The half-wave plate and polarizing beam splitter are adjusted to maximize the pump beam power going into the iodine cell and the orthogonally polarized probe beam power impinging on the detector. Owing to partial depletion of the iodine ground state by the pump beam, the carrier and sidebands of the probe beam undergo different absorption and dispersion. This leads to the appearance of a beat signal at the modulation frequency in the vicinity of a Doppler-free absorption line. By mixing the beat signal with a local oscillator after an appropriate phase delay, we obtain the dispersion signal of this line at dc. The signal-to-noise ratio is improved by chopping the pump beam and detecting the output signal with a lock-in amplifier. The lock-in amplifier output is fed back into the PZT frequency actuator of the laser through a servo amplifier with a unity gain point near 50 Hz, which thus locks the laser frequency to a hyperfine line of the iodine.

Six relatively strong Doppler-broadened lines in the range[3] 18 787–18 789 cm^{-1}, as well as a few weaker lines, are observed within the tuning range of the laser. The lowest-frequency hyperfine component of the absorption line whose mean position is at[14] 18 788.337 1(6) cm^{-1} is used as the frequency reference (see Fig. 2). By using vibrational band head energies and rotational constants[15] this line can be identified as $R(56)32\text{-}0$ at a calculated frequency of 18 788.3345 cm^{-1}. At room temperature, we measured a hyperfine linewidth of 5.3 MHz for one system and a power-broadened linewidth of 12.4 MHz for the higher-power system.

The laser remains locked to the iodine hyperfine line as long as the frequency drift does not exceed the piezo actuator range. Typically, the system remains locked for more than 1 h. We were able to lock the laser for longer periods (up to 24 h) by occasionally readjusting the Nd:YAG temperature. An additional servo amplifier connected to the temperature frequency actuator of the laser could keep the laser locked to the iodine line for arbitrarily long periods. Two completely independent systems have been built, with each laser locked to its own iodine cell. The heterodyne beat-note signal between the lasers is measured at 1064 nm by using a photodetector followed by a frequency and time interval analyzer (Hewlett-Packard HP 5371A), with one of the lasers frequency shifted by 80 MHz using an acousto-optic modulator. The stability of the locked and free-running lasers is calculated by using the Allan variance,

$$\sigma^2(\tau) = \frac{1}{2\nu^2(M-1)} \sum_{i=1}^{M-1} (y_{i+1} - y_i)^2,$$

where τ is the time between successive measurements as well as the duration of each frequency measurement, ν is the mean optical frequency, M is the number of measurements, and y_i is the ith frequency measurement. Figure 3 shows the root Allan variance as a function of τ for free-running and iodine-

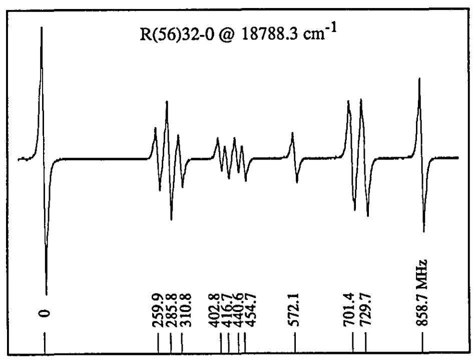

Fig. 2. FM saturated absorption spectrum of the 18 788.3-cm^{-1} line of $^{127}I_2$. The center lines are weaker owing to the Doppler envelope. The frequency spacings between the lines were determined by locking one system to the lowest-frequency hyperfine transition and locking the second system each time to a different transition. The beat frequency was measured by taking the average of one hundred 4-s measurements of the HP 5371A. The lines at 285.8, 701.4, and 729.7 MHz each contain two unresolved hyperfine transitions.

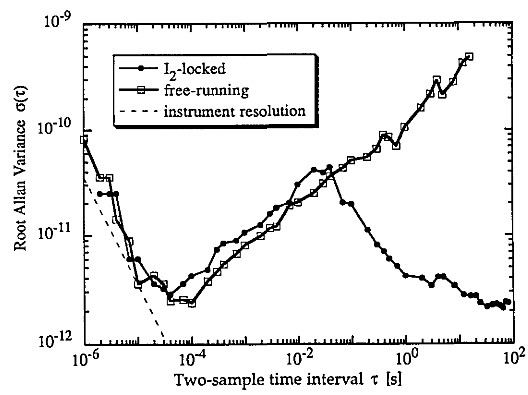

Fig. 3. Root Allan variance of the beat note between two locked lasers and two free-running lasers for $M = 100$ and $\nu = 281.63$ THz.

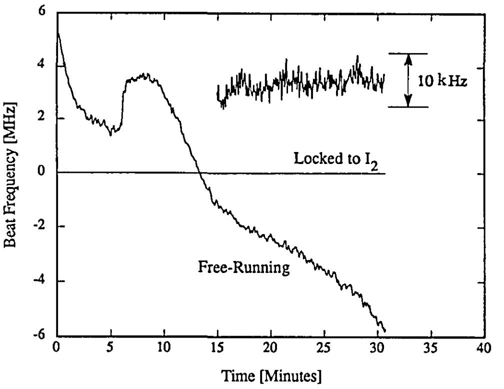

Fig. 4. Variation of beat-note frequency as a function of time. In each case 460 successive 4-s measurements of the frequency were made. The inset shows an expanded view of the beat frequency between the iodine-stabilized lasers over the last 15 min of the measurement.

stabilized lasers. As can be seen, the frequency drift becomes dominant above 100 μs in both cases. However, for $\tau > 0.03$ s the suppression of the drift by the servo controllers becomes noticeable, and the frequency stability of the locked lasers becomes much better than that of the free-running lasers and maintains a stability level of ~650 Hz over a carrier frequency of 281.63 THz (root Allan variance of 2.3×10^{-12}) between 24 and 80 s (this is the longest possible interval for 100 successive measurements with the HP 5371A). A stability of better than 1×10^{-11} is maintained for all measurements with $\tau > 0.3$ s. The drift of the free-running lasers at long time delays is quite random but is particularly high during the first hour of operation (more than 100 MHz) and when the ambient temperature changes. The data of Fig. 3 were taken more than 10 h after the lasers were turned on.

Figure 4 shows the typical variation of the beat frequency between the lasers when they are free running and when under lock over 30 min. The maximum excursion of the unlocked lasers exceeded 11 MHz, whereas for the locked lasers it is less than 15 kHz. The hyperfine lines of iodine drift by several kilohertz per kelvin, mainly owing to changes of the iodine vapor pressure,[4] hence tight control of the iodine cell temperature should reduce these line shifts and further improve the frequency stability over this time period.

In summary, we have locked for what is to our knowledge the first time diode-laser-pumped Nd:YAG lasers to hyperfine lines of iodine and essentially eliminated the long-term frequency drift of these lasers. In this system, both the fundamental and second-harmonic frequencies are stabilized. This is a favorable feature for some metrological measurements. Furthermore, the available power levels of diode-pumped solid-state lasers are sufficient to generate efficiently through nonlinear frequency conversions a series of stabilized frequencies from the UV to the near IR. Since both the laser and the doubler are monolithic and since the laser is pumped by a diode laser, technical noise is minimized, and a high level of frequency stability is obtained even without prestabilizing the lasers to a narrow-linewidth Fabry–Perot interferometer. This fact, together with the relatively strong room-temperature absorption lines of iodine near 532 nm, make the iodine-locked Nd:YAG laser a practical, reliable, and compact source for absolutely stable and reproducible radiation. We anticipate, however, that prestabilization, as well as cooling of the iodine cell temperature to reduce the pressure broadening of the linewidth and enhancement of the signal with an external resonator,[16] will further improve the present stability and extend it toward shorter averaging time intervals.

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