Absolute Frequency of the Molecular Iodine Transition R(56)32-0 Near 532 nm

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Abstract—The absolute frequency of the hyperfine component a_{10} in the transition R(56)32-0 of iodine has been measured using the D_2 line in Rb at 780 nm and an iodine-stabilized 633-nm He–Ne laser as references. This measurement provides a secondary frequency standard within the tuning range of a doubled Nd:YAG laser. The measured frequency of the a_{10} component is 563 260 223.480 MHz ±70 kHz.

I. INTRODUCTION

A FREQUENCY-DOUBLED 1.064-μm Nd:YAG MISER has recently been stabilized to iodine near 532 nm [1]–[3]. By using the Doppler-free spectroscopic technique of modulation transfer [4]–[6], excellent frequency stability was obtained (frequency reproducibility of less than 1 kHz and Allan variance at 1 s integration time $< 1 \times 10^{-13}$). Thus, the frequency-doubled Nd:YAG with iodine stabilization can serve as an accurate secondary frequency standard with several advantages over existing standards in the visible. First, the intrinsic noise in an all solid-state system can be much less than in a gas laser system. Second, due to the strong iodine transitions in the green and the higher available laser power, there is no need for an intra-cavity setup or even an external buildup cavity. This facilitates control over relevant parameters, such as optical power and iodine pressure. The S/N-ratio is also high enough (S/N = 500 in a 3 kHz BW) that it should be possible to narrow the laser linewidth to less than 20 Hz. The purpose of this paper is to report our measurement of the absolute frequency of the hyperfine component a_{10} in the green iodine transition R(56)32-0.

The most accurate way of measuring absolute frequencies in the optical domain is to use frequency chains to link the unknown frequencies to known frequency standards. There are several frequency chains that can access the iodine lines at 532 nm. One possibility is to sum two iodine-stabilized He–Ne lasers, one at 633 nm and the other at 3.39 μm. The generated sum frequency is then 1.2 THz to the red of 532 nm. However, the available intensities are very small, and it is therefore more convenient to use a chain referenced to Rb transitions $\sim$780 nm, where powerful Ti:Sapphire lasers are available. In a Rb-based chain, either the two-photon transition

$(5S_{1/2} - 5D_{3/2})$ at 778 nm or the D_2 line at 780 nm can be utilized. The principle of this chain is based on the fact that the sum frequency of the two-photon line at 778 nm and the iodine-stabilized 532-nm from a doubled Nd:YAG laser is 1.2 THz larger than the doubled frequency of an iodine-stabilized 633-nm He–Ne laser. By using as a reference the Rb-D_2 line at 780 nm, this frequency difference is only 263 GHz and should be directly measurable with a Schottky diode [7]. However, the Schottky diode is not sensitive to UV light. Thus, in our system the 263-GHz frequency difference is shifted to 780 nm by adding an auxiliary IR laser correspondingly detuned from the Rb line. By simultaneously counting the UV-beat frequency (which now is in a convenient MHz range) and the 263-GHz IR-beat frequency, the absolute frequency for the 532-nm iodine transitions can be determined. We want to stress that any drifts in the auxiliary laser will cancel, since they will appear equally in the UV- and in the IR-beat frequency.

II. EXPERIMENTAL CONSIDERATIONS

The experimental setup is shown in Fig. 1. A 0.7-mW single mode He–Ne laser is frequency doubled with an RDP crystal in a buildup cavity, producing $\sim$100 nW at 316 nm. This He–Ne laser is frequency locked to an iodine-stabilized laser. The frequency of the $^{127}I_2$-stabilized laser has recently been intercompared with two portable "transfer standard" lasers from BIPM (see below). By summing the frequencies of our iodine-stabilized, frequency-doubled Nd:YAG laser and a Ti:Sapphire laser (Ti:Sapphire #2 tuned around 780 nm) in a second RDP crystal, several μW of 316 nm are generated. The two UV beams are combined on a photo multiplier tube (PMT). The resulting beat-note is however too weak to be counted directly (S/N $\sim$20–25 dB in 30-kHz bandwidth). A tracking oscillator is formed by phase locking a voltage-controlled oscillator (VCO) to the beat-note, thus generating an exact phase/frequency replica of the beat-note. This generated sinewave is compared with the original wave to show that cycle-skipping is extremely infrequent.

The reference Ti:Sapphire laser is locked to a hyperfine component of the D_2 line in Rb. The IR beams from the two Ti:Sapphire lasers are combined on a Schottky diode. The resulting 263-GHz beat-note is mixed down by injecting a 43-GHz μ-wave directly on this diode. The 6th harmonic is tuned near our optical beat frequency and leads to a strong down-converted signal (S/N $\sim$ 45 dB in a 30-kHz bandwidth), which will be in a convenient range for a fast counter. The 43-GHz μ-wave is generated by a klystron, which is phase locked to the 12th harmonic from a frequency synthesizer. Due to the high
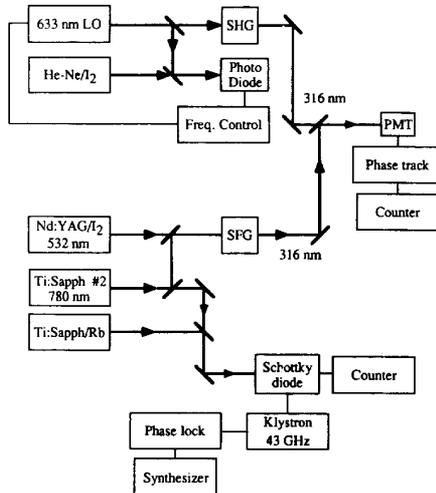
Fig. 1. The experimental setup for measuring optical frequency. Thick lines indicate optical connections while thin lines indicate electrical.

Fig. 2. Frequency scan over the hyperfine components in ^{87}Rb. Assignment of the components is indicated.

Fig. 3. Optical frequency shifts induced by laser power. The probe power used was 10 μW. One notices the strong shifts of the f-component, which arises from the closed 2-level transition $F'' = 2 \rightarrow F' = 3$, whereas the d-component ($F'' = 2 \rightarrow F' = 2$) corresponds to an open transition and has a much smaller shift. See text for details.

order of frequency multiplication, the accuracy of the beat-note will have a 26 kHz/Hz sensitivity to the timebase used in the frequency synthesizer. To achieve an optical frequency accuracy of better than 1 kHz, the synthesizer's internal timebase has to be measured (and maintained) to better than 40 mHz. Appropriate strategies for this would be referencing to a Rubidium (or Cesium) standard, although a GPS-based frequency reference may be marginally suitable[1] [8].

The saturation signals in the Rb-spectrometer were extracted by frequency modulating the laser light and using 3rd harmonic detection. The frequency dither was generated with a double-passed acoustooptic modulator (AOM). To assure a stable center frequency, the AOM was driven by an FM-modulated synthesizer. The unavoidable amplitude modulation (AM) contamination was suppressed (> 40 dB) by amplitude stabilizing the laser power going into the spectrometer. Fig. 2 shows the relevant saturation signals together with an assignment of the hyperfine components. The isotope ^{87}Rb is used due to its larger hyperfine splitting. To test the performance of the Rb-spectrometer, a second Rb-spectrometer was built. The beat-note between two Ti:Sapphire lasers, each locked to a Rb-spectrometer, provides a very accurate measure of frequency shifts due to environmental perturbations.

There are several processes that can shift the line centers. The easiest to control is the power shift. Power shifts for different lines are shown in Fig. 3. The extremely strong power dependence for the f-line is due to light pressure [9]. A closed transition will rapidly accumulate photon momentum and subsequently alter the Doppler curve. This adds a symmetric component to the antisymmetric saturation signal, which appears as a shift of the zero-crossing of the resonance. This shifting effect can also be seen on the d-f crossover, but is much weaker because this line is not completely closed. The d- (and b-) lines are relatively insensitive to the saturation power.

However, because the d-f crossover is very strong and isolated, it is still a good candidate for a reference line, provided that a low enough saturating power is used.

Magnetic fields can also shift the lines. The primary effect of weak magnetic fields is Zeeman broadening. If there is optical pumping between sublevels, a huge frequency shift is expected as well. Thus, the cell was placed perpendicular to the earth's magnetic field, and the saturation spectrometer employed appropriate linearly-polarized beams. This will favor $\Delta m = 0$ transitions, and optical pumping between sublevels is minimized. This is especially important since we use fairly large beams ($\sim$4 mm in diameter), and so the atoms are more susceptible to optical pumping effects. To measure the effect of magnetic fields, a bar magnet was placed in different positions close to the cell, and the inhomogeneous magnetic field was monitored with a Gaussmeter. The frequency shift due to the residual earth's magnetic field was thus estimated as < 5 kHz.

The Rb absorption cells were 10 cm long with windows set for near normal incidence. They were manufactured at JILA using cleaning and baking guidelines similar to those

[1] A third alternative would be direct laser based frequency transfer through the $\sim$ 1 km open air path from NIST to JILA using our new double path phase noise measuring/noise cancellation concept.

recommended for iodine cells [10]. The empty cell is heated to 350°C for two days, and the Rb was purified several times by distillation before transferring it into the cells. The pressure at tip-off was < 130 μPa. The residual background contamination can be estimated by measuring the linewidths of the Rb transitions. From the natural lifetime [11], the expected zero power linewidth is 5.9 MHz. By extrapolating to zero power, the measured linewidth was ~7 MHz. The estimated Zeeman broadening was <500 kHz. The extra 600 kHz broadening included all experimental parameters and could include a portion due to background gas contamination. However, the frequency shifts between three of these cells were measured to be less than a few kilohertz.

By overlapping the counterpropagating pump and probe beams, Doppler shifts and residual broadening became negligible. The necessary isolation from interferometric baseline instabilities is achieved by frequency shifting the pump beam with an AOM [12]. AM can occur if the pump beam has a spatial mode structure. The probe beam might then interrogate a region of the pump beam that has a huge intensity gradient. Vibrations at the detection frequency will then cause AM to be transferred to the probe beam. AM adds a symmetric part to the lineshape and thus causes asymmetry. This was avoided by cleaning up the spatial mode of the laser beam by focusing it through a 10-μm pinhole. The measured asymmetry of the lines was <10^{-3} (except for the f-line, which can have a severe asymmetry due to light pressure). Here the asymmetry is defined as the difference of the size of the "derivative" resonance above and below the baseline normalized to the total size of the resonance.

The green iodine spectrometer utilizes modulation transfer [4]–[6] to extract the transitions. More than six Doppler-broadened transitions are within the tuning range of a doubled Nd:YAG laser. Of these, the transition R(56)32–0 has a large hyperfine splitting and is fairly well isolated. Within this line, the hyperfine component a_{10} is used as the main reference line. It is isolated and near the center of the Doppler curve (see Fig. 4). The two other isolated hyperfine lines a_1 and a_{15} are both on the slope of the Doppler curve, and so the sidebands are absorbed unequally. As a result, an asymmetry is introduced, and a small frequency shift may result. The temperature of the iodine cell was stabilized to −20±0.1°C. The pump power used was 500 μW and the probe power 200 μW. A detailed description of the iodine spectrometer and stabilization of the Nd:YAG lasers is given elsewhere [13].

III. RESULTS

In a typical experimental run (run #8 on May 19 1994), the second Ti:Sapphire laser was tuned so that the UV beat-note was 21.792 MHz. In this case, the down-converted IR beat-note was 993.627 MHz, with the klystron phase-locked to the 12th harmonic of 3675.825 MHz. This gives for the frequency of the a_{10} component $f_{a_{10}} = 563\,260\,223.495$ MHz. Taking the average of approximately 50 runs performed during May and June 1994, the absolute frequency of the a_{10} line was measured to be

$$f_{a_{10}} = 563260223.480 \text{ MHz} \pm 70 \text{ kHz}$$
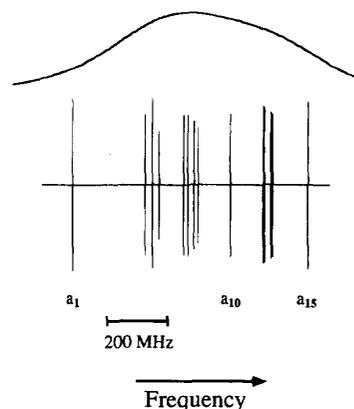
Fig. 4. The transition in R(56)32-0 in iodine. The lower trace shows the hyperfine component measured with modulation transfer. The upper trace is the Doppler curve calculated from the measured hyperfine splittings and relative intensities.

The measurement was made with a 40-μW pump power in the Rb-spectrometer, but the above result is corrected for the power shift as shown in Fig. 3. The main error contribution is from the uncertainty of the Rb-reference line, which was recently measured to ±60 kHz at NPL [14]. The standard deviation of 2000 measurements of the frequency of the component a_{10}(total measurement time ~40 min) is ±5 kHz. However, the day-to-day reproducibility of the chain is ±30 kHz, which still is well within the uncertainty of the Rb reference. The estimated error from the 633-nm iodine-stabilized He–Ne laser is ±7 kHz, which will be doubled to 14 kHz in the frequency-doubling process. This error is deduced from the intercomparison of our iodine-stabilized He–Ne laser and the two portable frequency standards from BIPM (BIPM P1, P3). These BIPM lasers were calibrated against BIPM4 both before and after the intercomparison at JILA. The frequency difference between BIPM4 and BIPM P1 differed by 6-kHz between these two calibrations. This frequency difference was 1.5 kHz for BIPM P3. In this way, the JILA He–Ne reference laser was found to be 6 ± 2 kHz red of the BIPM4. In our quoted result, we take the frequency of BIPM4 as 473 620 612.705 MHz as recommended [15] and adopt the 5 kHz uncertainty of its measurement. Experience with comparing freshly-made lasers with BIPM4 suggests that any possible drift is very small. The reproducibility of the green iodine spectrometer was less than 1 kHz [13], and is therefore negligible at the present level of accuracy. The total error (root sum of squares) is thus ±70 kHz.

All frequency sources, such as those driving the AOM's, were measured with a frequency counter recently calibrated against a rubidium standard. The long-term drift for the AOM's was found to be negligible (a few tens of hertz).

Earlier measurements by means of Fourier transform spectroscopy yielded a frequency for the center of gravity for the R(56)32-0 transition as 563 260 155 ± 60 MHz [16]. The eigenvectors of the quadrupolar hyperfine Hamiltonian were obtained by least-squares fitting of the measured hyperfine intervals [14], which leads to relative intensities for the various hyperfine lines. With this information, $f(a_{10})$ is calculated to

be 96 MHz higher than the center of gravity. Starting from the Fourier transform centroid, this gives $f(a_{10}) = 563\ 260\ 251$ MHz, which is 28 MHz higher than our results. However, it is still well within the 60 MHz uncertainty quoted for the Fourier transform results.

A second confirmation of the result was done by measuring the wavelength of the iodine transition interferometrically. We used a 1 m-long lambdameter, which was calibrated against the D_2 line in sodium [17] and the P(62)17-1 transition in iodine [15]. The achieved accuracy was ±10 MHz. The result $f(a_{10}) = 563\ 260\ 223 \pm 10$ MHz is in excellent agreement with our result based on the frequency chain.

IV. CONCLUSION AND FUTURE OUTLOOK

We have measured the absolute frequency of the R(56)32-0 transition in iodine to an accuracy of ±70 kHz. The uncertainty is mainly due to the uncertainty in the Rb-D_2 line that was used as a reference. Since the Nd:YAG laser can be stabilized to this iodine transition with a reproducibility in the sub-kHz domain, it is clearly desirable to measure these lines with an even higher accuracy. The best direct way of lowering the uncertainty is to use the two–photon transition at 778 nm in Rb as a reference. The absolute frequency of this line has been measured to ±5 kHz [18]. The two-photon lines are also very narrow (~300 kHz), so the reproducibility when stabilizing a laser to these transitions is superior to that obtained when the laser is stabilized to the D_2 line. The frequency difference of 1.2 THz can be cut in half by an auxiliary diode laser tuned between the two Ti:Sapphire lasers. Basically, this represents a realization of the Hänsch-Meschede-Telle scheme of "frequency-interval bisection" [19]. The resulting 600-GHz beat-notes can be measured with the same Schottky diode. Since 630-GHz beats have already been observed using our Schottky diode, work along these lines is already in progress.

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