

## Stability and absolute frequency of molecular iodine transitions near 532 nm

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### ABSTRACT

A frequency-doubled Nd:YAG laser has been stabilized to hyperfine transitions in  $I_2$  near 532 nm via modulation transfer spectroscopy. This technique, together with the low noise of the source, yields excellent S/N ratio (500 in a 3 kHz bandwidth). Thus an impressive frequency stability (Allan variance  $< 1 \times 10^{-13}$  for  $\tau=1s$ ) is achieved. The nearly systematic-free resonance signals obtained by modulation transfer spectroscopy give an correspondingly encouraging reproducibility, estimated to be about  $\pm 300$  Hz ( $\pm 5 \times 10^{-13}$ ). With two such stabilized lasers we found a pressure shift of only -1.3 kHz/Pa over the range 0.4 - 4.0 Pa and a power-dependent frequency shift of 2.1 kHz/mW. We have also measured the absolute frequency of the component  $a_{10}$  in the transition R(56)32-0 using the  $D_2$  line in Rb at 780 nm and an iodine stabilized 633 nm He-Ne laser as references. The measured frequency is  $563\,260\,223.471$  MHz  $\pm 40$  kHz. In turn, the absolute frequency of the  $D_2$  line was measured via the frequency difference between the  $D_2$  line and the 2-photon transition  $5S_{1/2} - 5D_{5/2}$  at 778 nm in Rb. Thus we now have realized a pure frequency measurement of this interval and of the 532 nm frequency.

### I. INTRODUCTION

Transitions in molecular iodine often provide stable references for precision spectroscopy and a half-dozen such iodine lines have been recommended in the CCDM' 92 report<sup>1</sup> for use as secondary optical frequency standards. The transitions in the green wavelength domain are typically very strong and well suitable for standard purposes. Two standards in the green are produced by gas lasers, namely 514 nm by an  $Ar^+$  laser and 543 nm by a He-Ne laser.<sup>1</sup> The transitions near 532 nm seem very attractive as frequency standards since they can be probed by frequency doubled Nd:YAG lasers. These all solid state lasers can have very low noise and be thus suitable for frequency standards. However, questions naturally arise concerning the reproducibility, stability and any possible systematic shifts of a laser source locked to one

of the hyperfine components in these transitions. In this paper we report on measurements made with two such systems, each consisting of a frequency-doubled Nd:YAG Non Planar Ring Oscillator (NPRO) with a modulation transfer iodine spectrometer. We have also measured the absolute frequency component  $a_{10}$  of R(56)32-0 with a frequency chain linking the rubidium  $D_2$ -line and the iodine-stabilized 633 nm He-Ne laser with the transition at 532 nm. The accuracy of the measurement has been improved over our previous result.<sup>2</sup> The improvement originates in a better value for the absolute frequency of the  $D_2$  line obtained by a direct frequency measurement between the  $D_2$  line and the two-photon transition  $5S_{1/2} - 5D_{5/2}$  in Rb.

The absolute frequency of these green iodine transitions can be measured in several ways. The most accurate method in the optical domain is to use frequency chains to link the unknown frequency to known 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  $\mu\text{m}$ . The generated sum frequency is then 1.3 THz to the red of 532 nm iodine transition. Another possibility is to utilize the two-photon transition ( $5S_{1/2} - 5D_{5/2}$ ) at 778 nm in Rb. The difference frequency between a laser stabilized to this two-photon transition and the iodine-stabilized 532 nm laser is 1.2 THz to the blue of a frequency-doubled methane-stabilized 3.39  $\mu\text{m}$  He-Ne laser. The accuracy of this chain is determined by the uncertainty of the frequency of the two-photon transition, which recently has been measured to  $\pm 5$  kHz.<sup>3</sup> However, in both chains described above the available intensities are very small and it is therefore more convenient to use a chain based on the  $D_2$ -line in Rb at 780 nm. The principle of this chain is based on the fact that the sum frequency of the  $D_2$ -line at 780 nm and the iodine stabilized 532 nm from a doubled Nd:YAG laser is 263 GHz larger than the doubled frequency of an iodine stabilized 633 nm He-Ne laser. This frequency difference is directly measurable with a Schottky diode.<sup>4</sup> 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 a second auxiliary 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. The accuracy of this chain is determined by the uncertainty of the Rb  $D_2$ -line. This line was measured at NPL to an uncertainty of  $\pm 60$  kHz by interferometric means.<sup>5</sup> However, since tests showed that our Schottky diode was capable of measuring  $\sim 1$  THz frequencies, the accuracy of the absolute frequency of the Rb  $D_2$ -line was improved to  $\pm 40$  kHz by a direct frequency measurement of the frequency between the  $D_2$ -line and the two-photon transition in Rb. Thus the absolute frequency measurement of the iodine transitions at 532 nm is now a pure frequency measurement.

## II. EXPERIMENTAL CONSIDERATIONS

Two  $I_2$ -stabilized, frequency-doubled Nd:YAG laser systems were built. Similar systems have been reported previously.<sup>6,7</sup> Throughout the text, we will label each  $I_2$ /doubled Nd:YAG laser system for clarity with E or W, meaning the laser system or components on the east or west side of our optical table. The source oscillator for each system is a diode pumped Nd:YAG Non-Planar Ring Oscillator (NPRO) which emits about 300 mW at 1064 nm. Second harmonic generation for each laser system is

accomplished through the use of a ring "buildup" cavity with an intracavity nonlinear crystal. These cavities are both weakly dithered at  $\sim 50$  kHz and locked to the fundamental beam at 1064 nm with a first derivative lock. In the E frequency doubler, a MgO:LiNbO<sub>3</sub> crystal is heated to its phase matching temperature of about 110 °C. The maximum generated 532 nm output is about 80 mW. In the W frequency doubler we employ a slightly different scheme. Instead of the usual 4 mirror ring cavity with one focus, we use a 6 mirror ring cavity with 2 foci. A KNbO<sub>3</sub> crystal is placed in the first focus and generates the second harmonic at 532 nm, at a phase matching temperature of about 180 °C. In the second focus a LiIO<sub>3</sub> crystal is located. It is angle-tuned so that it sums the frequencies of the fundamental and the second harmonic to produce the third harmonic at 355 nm. Thus we have produced a rather sparse comb of optical frequencies with very simple ratios. The type I phase matching in the SFG process requires an extra waveplate in the cavity which is  $\lambda$  at 1064 nm and  $\lambda/2$  at 532 nm. The output is about 35 mW of green and 200  $\mu$ W of UV-light.

The iodine spectrometers are based on the sub-Doppler technique of modulation transfer.<sup>8</sup> The beam is divided into a strong pump and a weak probe beam, with the pump beam shifted 80 MHz to avoid interference.<sup>9</sup> The pump beam is phase modulated with an EOM. In the W (E) spectrometer the modulation frequency is 500 (680) kHz and the modulation index is 2.1 (1.0). To allow efficient separation of the pump and probe beams the pump beam is rotated 90° with a half-wave plate. In the W spectrometer, the beams have crossed polarizations within the 1.2 m long iodine cell (square-cut windows). In the E spectrometer, a 45° Faraday rotator is inserted in both the pump and the probe beams. Thus the polarizations of the beams are linear and parallel within the 1.2 m long iodine cell (Brewster-cut windows). The vapor pressure in each cell is established by temperature controlling the cold finger, with  $T \sim -10$  °C (pressure  $\sim 1.4$  Pa). The probe beam is separated by a polarizing beam splitter and imaged onto a photodiode. The ac component of the signal (at the modulation frequency) is amplified and demodulated in a double balanced mixer (DBM). An analog phase shifter is inserted between the reference signal and the DBM to allow a selection of either the in-phase or quadrature component of the signal.

With both lasers independently locked to different hyperfine components, Allan Variance data was collected. By flipping the lasers back and forth between several pairs of transitions, the offsets between the two systems were determined to be  $< 300$  Hz. The pressure and power shifts could be determined by varying these parameters while keeping one laser fixed and recording the change in the beat frequency. It is however more accurate to use a second method for this task. Here one laser is treated as a master and the other as a slave laser. The master laser is locked to a hyperfine component, while the slave laser is phase locked to the master system with a precisely tunable rf offset supplied by a synthesizer. As the synthesizer is tuned, the slave laser is tuned correspondingly. Thus the slave laser can be stepped over an iodine resonance and the modulation transfer lineshape may be recorded by a computer. The produced experimental lineshape is fitted to the theoretically predicted lineshape and the parameters of interest may be determined. Here we used the line center, the amplitude and width of the resonance as well as an additive offset as adjustable parameters. By including an offset, greater accuracy is achieved in the characterization of the line center. A more detailed description of the spectrometers is given elsewhere.<sup>7</sup>

To measure the absolute frequencies of the iodine transitions the setup showed in Fig. 1 was used. A 0.7 mW single mode 633 nm He-Ne laser is frequency doubled with an RDP crystal in a buildup cavity. With a phase matching temperature of about 47 °C, about 100 nW of 316 nm was produced. This He-Ne laser is loosely phase locked to an iodine stabilized laser with an 88 MHz offset. This phase lock loop has a low bandwidth and enough phase range and incorporates a notch filter so that the 5 kHz modulation in the iodine-stabilized laser is not transferred. The frequency of our iodine-stabilized laser has recently been intercompared with two portable "transfer standard" lasers from BIPM (BIPM P1, P3) and is found to be 5 kHz to the red of the recommended value, which we assume to be the "as maintained" value of BIPM laser P4. By summing the frequencies of our iodine-stabilized frequency-doubled Nd:YAG laser and a Ti:Sapphire laser (Ti:sapph #2 properly tuned around 780 nm) in a second RDP crystal, several  $\mu$ W of 316 nm is 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  $\sim$  20 MHz beat-note, thus generating an exact phase/frequency replica of the beat-note. Effectively we have now generated a Ti:Sapphire laser locked onto  $I_2$  lines at 633 and 532 nm.

A second 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  $\mu$ -wave directly on this diode. The sixth harmonic is tuned near our optical beat frequency and leads to a strong down-converted signal ( $\sim$  45 dB in a 30 kHz bandwidth) which will be in a convenient range for a fast counter. The 43 GHz  $\mu$ -wave is generated by a klystron which is phase locked to the 12th harmonic from a frequency synthesizer. Due to the high 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 include referencing to a Rubidium (or Cesium) atomic standard, although averaging for a few hours of a GPS-referenced frequency reference also appears to be suitable.

The saturation signals in the Rb-spectrometer were extracted by frequency modulating the laser light and using third harmonic detection. The frequency dither at 5 kHz was generated with a double passed acousto-optic modulator (AOM). To assure a stable center frequency, the AOM was driven by a FM modulated synthesizer. However, a double passed AOM, even properly aligned, can produce some residual AM at the dither frequency and/or its harmonics. This unavoidable AM contamination was additionally suppressed ( $>$  40 dB for the fundamental) by amplitude stabilizing the laser power going into the spectrometer. In addition to this, the pump beam was chopped at 600 Hz to eliminate the effect of any residual AM at the third harmonic. The saturated absorption signal was obtained by synchronous detection at the third harmonic, followed by demodulation with lock-in technique at the 600 Hz chopping frequency imposed on the pump beam. This chopping action on the pump beam turns out to be very important since the amount of AM contamination is changing with time due to thermal effects in the AOM. This would produce a shifting background which in turn would show up as a frequency drift in the locked laser. When the laser was locked to the crossover between the  $F=2 \rightarrow F=3$  (f-line) and  $F=2 \rightarrow F=2$  (d-line) transitions in  $^{87}\text{Rb}$  this drift was in the order of 10 - 20 kHz without chopping the pump. The isotope  $^{87}\text{Rb}$  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 measure of frequency shifts due to environmental perturbations. The power shifts were measured to be  $-180 \text{ Hz}/\mu\text{W}$  for the d- and b-line ( $F=2 \rightarrow F=1$ ), while it was larger ( $-0.95 \text{ kHz}/\mu\text{W}$ ) for the crossover between the f- and the d-line with a pump beam diameter of 4 mm. The f-line was extremely sensitive to the pump power. In some major part, this is due to light pressure.<sup>10</sup> 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 can also be seen on the d-f crossover, but much weaker because this line is not completely closed. The Rb 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 recommended for iodine cells. The empty cell is heated to  $350 \text{ }^\circ\text{C}$  for several days and the Rb was purified several times by distillation before transferring it into the cells. The pressure at tip-off was  $< 130 \mu\text{Pa}$ .

We have also measured the frequency difference between the Rb  $D_2$  line and the two-photon transition  $5S_{1/2} - 5D_{5/2}$  ( $F=2 \rightarrow F=4$ ). Since the absolute frequency of the two-photon transition is known to  $\pm 5 \text{ kHz}$ <sup>3</sup> this yields an absolute frequency for the  $D_2$  line. The frequency difference to be measured is about 1.05 THz. To measure such a high frequency, it was mixed down by injecting a 65 mW, 91 GHz  $\mu$ -wave from a Gunn diode directly on the Schottky diode. When the two lasers are tuned apart a down-converted signal can be detected each time the frequency difference between the lasers is a multiple of the  $\mu$ -wave. In figure 2 the frequency response of our Schottky diode is shown. When a 300 mW, 45 GHz  $\mu$ -wave from a klystron was used the maximum detected beat frequency was only  $\sim 600 \text{ GHz}$ . This suggests that it is the harmonic generation of the Gunn diode (Klystron) that limits the bandwidth of the Schottky diode, at least for frequencies less than 1 THz. At 1 THz the down converted signal was only  $\sim 5 \text{ dB}$  above the noise floor in a 1 MHz bandwidth (see figure 2). However, it is still strong enough, so that a tracking oscillator can be phase locked to it for frequency counting purposes. To assure a stable frequency for the Gunn diode, it was phase locked to the 9th harmonic of a  $\sim 10.6 \text{ GHz}$  Dielectric Resonant Oscillator (DRO). The high frequency ( $>100 \text{ Hz}$ ) phase noise in such an oscillator is extremely low. This is important in frequency multiplication since the power in the noise sidebands relative to the carrier generally increases as  $n^2$  where  $n$  is the multiplication factor.<sup>11</sup> The DRO was then phase locked to a frequency synthesizer with a low ( $<100 \text{ Hz}$ ) bandwidth, so the high frequency phase noise from the synthesizer would not be transferred to the DRO. The 11th harmonic of the DRO is  $\sim 6 \text{ GHz}$  smaller than the frequency difference between the  $D_2$  line and the two-photon transition. This beat was mixed down with a frequency synthesizer to 1 GHz, filtered with a 20 MHz broad bandpass filter and then counted. The time bases for the synthesizers were generally fed from a Cesium standard, recently calibrated at NIST. All other oscillators, such as the AOM's and the oscillators in the offset phase locks were measured with a counter calibrated against the Cesium standard.

The two-photon Rb-spectrometer was constructed along the guidelines in reference 3. A collimated laser beam is retro-reflected with a plane mirror through the 5 cm long Rb-cell. The two-photon signal was recorded using a PMT which detects the fluorescence at  $\lambda = 420 \text{ nm}$ . The beam size was 0.5 mm in diameter which corresponds to  $\sim 300 \text{ kHz}$  transit time broadening. Thus the resonances resembles a Voigt profile. Computer fits of the lineshapes yield a typical linewidth of 500 kHz with a Gaussian

component of 300 kHz. To reach these linewidths the earth's magnetic field was shielded by a factor of 10 with a box made of  $\mu$ -metal. The optical power was 25 mW and the cell temperature  $\sim 110$  °C. We dithered the Ti:Sapphire laser frequency at 5 kHz with a double passed AOM (similar to the Rb D<sub>2</sub> lock) and used lock-in technique to extract the dispersive shaped signal to lock the laser to the two-photon transition. The dither depth was  $\sim 100$  kHz. The servoloop is implemented with software running real-time on a laboratory computer, achieving a locking bandwidth of  $\sim 0.5$  Hz. By using the D<sub>2</sub> line as a reference the frequency shifts for the two-photon transition were investigated. No substantial frequency shift could be detected ( $\leq 5$  kHz) when the temperature, the optical power or the modulation depth was changed. However, the stability of the laser locked to the D<sub>2</sub> line is not enough to unambiguously determine any shifts in the two-photon lock. For this purpose a diode laser locked to the two-photon line is being constructed.

### III. RESULTS

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 is very strong and has narrow hyperfine components with large splittings. To investigate the suitability of this transition as a frequency standard, the pressure and power shifts were measured from lineshape measurements mentioned earlier. Even though a small nonlinearity was detected for the pressure shift,<sup>7</sup> the slope of the frequency shift was only -1.3 kHz/Pa. This is much smaller than the measured shifts for other iodine transitions ( -10 kHz/Pa for the transition at 633 nm<sup>12</sup> and  $\sim -7$  kHz/Pa at 612 nm<sup>13</sup> ). However the power shift of 2.1 kHz/mW was of the same order as the power shift for a He-Ne/I<sub>2</sub> laser at 633 nm (-1.4 kHz/mW of estimated intracavity power<sup>1</sup>). The hyperfine splittings for the transition R(56)32-0 is summarized in table 1. The measured splittings are fit to the 4-term hyperfine Hamiltonian, which includes the nuclear electric quadrupole spin-rotation, tensor spin-spin and scalar spin-spin interactions. The standard deviation of the residuals is only about 1.3 kHz, which indicates the high precision of the locking with the I<sub>2</sub>/doubled Nd:YAG system. Within this transition the hyperfine component a<sub>10</sub> was used as the main reference line. It is isolated and near the center of the Doppler curve. The absolute frequency was measured with the temperature of the iodine cell stabilized to  $-20 \pm 0.1$  °C. The pump power was 500  $\mu$ W and the probe power 200  $\mu$ W.

Component	F'	I	Measured (MHz)	Calculated (MHz)	Meas. - Calc. (kHz)
a <sub>1</sub>	57	2	-571.546776	-571.545724	-1.052
a <sub>2</sub>	53	4	-311.848288	-311.847728	-0.560
a <sub>5</sub>	58	2	-260.176900	-260.176045	-0.855
a <sub>6</sub>	56	2	-170.066022	-170.064365	-1.657
a <sub>7</sub>	61	4	-154.551039	-154.548713	-2.326
a <sub>8</sub>	54	4	-131.915891	-131.915011	-0.880
a <sub>9</sub>	55	4	-116.199435	-116.197377	-2.058

a <sub>10</sub>	59	4	0.000000	0.000000	0.000
a <sub>11</sub>	60	4	126.513402	126.512815	0.587
a <sub>12</sub>	57	4	131.210847	131.212825	-1.978
a <sub>13</sub>	55	2	154.490700	154.489932	0.768
a <sub>15</sub>	56	4	286.410404	286.413056	-2.652

Table 1. Measured and calculated hyperfine splittings for R(56)32-0. Components a<sub>3</sub> and a<sub>4</sub> form an unresolved doublet, which is not included in the fit. No data on a<sub>14</sub>. The differences between the measured and calculated hyperfine splittings have a standard deviation of only 1.12 kHz. The reference laser E was locked to a<sub>20</sub>:P(83)33-0 which is offset 15 697.8360 MHz below a<sub>10</sub>:R(56)32-0.

The frequencies of the Rb df crossover and d-line were directly measured by using the two-photon transition 5S<sub>1/2</sub> - 5D<sub>5/2</sub> (F=2 → F=4) as reference. The absolute frequency for this line was measured in reference 3 to 385 242 216.362 MHz ± 5 kHz. In figure 3 we show a statistical representation of our results. The absolute frequencies of the 780 nm D<sub>2</sub> line were thus measured to be

$$f_{dfx} = 384\,227\,981.879 \text{ MHz} \pm 30 \text{ kHz}$$

$$f_d = 384\,227\,848.543 \text{ MHz} \pm 25 \text{ kHz}$$

The measurement was made with a 30 μW pump beam in the Rb-spectrometer. The above result is corrected for the measured Rb power shift of -0.95 kHz/μW of the df crossover. The error is estimated from the day to day reproducibility and by assuming an accuracy of ± 5 kHz for the two-photon reference. This measurement yields a ~9 kHz higher absolute frequency for the d-f crossover line than the result obtained by interferometric measurement in reference 5. This result is well within the range of experimental uncertainty.

The hyperfine splittings for the lines a, c, d and e are the same as those listed in reference 5 within a few kHz. However, the splitting between the d-f crossover and d is ~ 100 kHz higher while the splittings between d-f crossover and a, c, e lines are ~ 100 kHz lower compared with reference 5. One important factor that can shift the lines is the quality of the cell. Background gas may lead to pressure dependent frequency shifts. This was pointed out in reference 5, where a cell with substantially larger linewidths showed a frequency shift of 200 kHz to the red. To test our cells we measured the frequency shift between our cells and two others made by H. Robinson at NIST, one of which contained isotope pure <sup>87</sup>Rb. The frequency shifts between all the cells were < 10 kHz.

In reference 2 we used the interferometric measured value of d-f crossover as our reference to measure the a<sub>10</sub> component. Our present frequency-based measurement of f<sub>dfx</sub> leads to a correction for the absolute frequency for the a<sub>10</sub> component given there by - 9 kHz. The absolute frequency is thus

$$f_{a10} = 563\,260\,223.471 \text{ MHz} \pm 40 \text{ kHz}$$

The error budget is as follows:<sup>2</sup> The estimated error for the doubled He-Ne/I<sub>2</sub> system is  $\pm 15$  kHz. The standard deviation of 2000 points is  $\pm 5$  kHz. However, the day to day reproducibility of the chain was  $\pm 30$  kHz, which we believe was associated with limitations of our Rb D<sub>2</sub> spectrometer. The frequency measurement of the df crossover yields an accuracy of  $\pm 30$  kHz. Thus the total error (root sum of squares) is 40 kHz.

#### IV. CONCLUSION

We have measured the absolute frequencies of some of the hyperfine components in the D<sub>2</sub> line to an accuracy of 30 kHz. In references 2 and 5 (and in the above result) the df crossover was used as the main reference line. However, with the increased accuracy this line is no longer an ideal candidate. One branch in this crossover is the closed f-line, which is extremely sensitive to environmental perturbations. It is thus unavoidable that the f-line will have a frequency pushing/pulling effect on the df crossover. Thus we suggest to use the open d-line as a main reference line. However, even with the df-crossover as a reference the accuracy of the absolute frequency of the R(56)32-0 transition in iodine was improved to 40 kHz. The uncertainty is still due to the uncertainty in the Rb D<sub>2</sub> line. Since the Nd:YAG laser can be stabilized to its I<sub>2</sub> lines with a reproducibility in the sub-kHz domain, it is clearly desirable to measure these lines with an even higher accuracy. The most obvious way of lowering the uncertainty is to use the two-photon transition at 778 nm in Rb as reference. Not only is the absolute frequency known to  $\pm 5$  kHz but the two-photon spectrometer is generally much more robust than the spectrometer based on the D<sub>2</sub> line mainly (because optical pumping and momentum-transfer phenomena are highly suppressed) but also because the two-photon transitions are very narrow ( $\sim 300$  kHz). However, in this case the frequency difference is 1.28 THz. This can be measured in several ways. From figure 2 it is clear that even if the beat-note could be seen with our Schottky diode, it would be extremely difficult to phase lock a tracking oscillator to it. A better way is to cut the frequency in half by an auxiliary diode laser tuned between the two Ti:Sapphire lasers. This represents a realization of the Hansch-Meschede-Telle scheme of "frequency-interval-bisection".<sup>14</sup> Another possibility is to use a frequency comb generator developed by Ohtsu *et al.*<sup>15</sup> In this case the Schottky diode is no longer needed and can be replaced with an Avalanche diode or a PMT.

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## VII. FIGURE CAPTIONS

Figure 1. The experimental optical frequency chain setup for measuring the absolute frequency of molecular iodine transition R(56)32-0 near 532 nm. Thick lines indicate optical connections while thin lines indicate electrical.

Figure 2. The frequency response of our Schottky diode taken with 1 MHz resolution bandwidth. The optical powers were 15 mW in one and 20mW in another laser beam. The optical heterodyne signal was down-converted by injecting a 90 GHz, 65 mW microwave from a Gunn diode directly on the Schottky diode. The optical heterodyne signal strength decreases as the frequency increases from DC with a slope of -20 dB/100 GHz. At ~ 400 GHz, the response curve hits a plateau with a ~ 15 dB signal to noise (S/N) ratio. At 1 THz, the S/N ratio decreases to ~ 5 dB.

Figure 3. Histogram on the measurement of the heterodyne beat frequency between the  $^{87}\text{Rb}$   $D_2$  line at 780 nm and  $^{85}\text{Rb}$  two photon transitions at 778 nm. The expected beat frequency value is calculated from the frequency measurement on  $^{85}\text{Rb}$  two photon transition in reference 3 and the interferometric measurement on  $^{87}\text{Rb}$   $D_2$  df crossover line in reference 5. The hyperfine splittings of the  $^{87}\text{Rb}$   $D_2$  lines were measured in our laboratory.

# Experimental Schematics

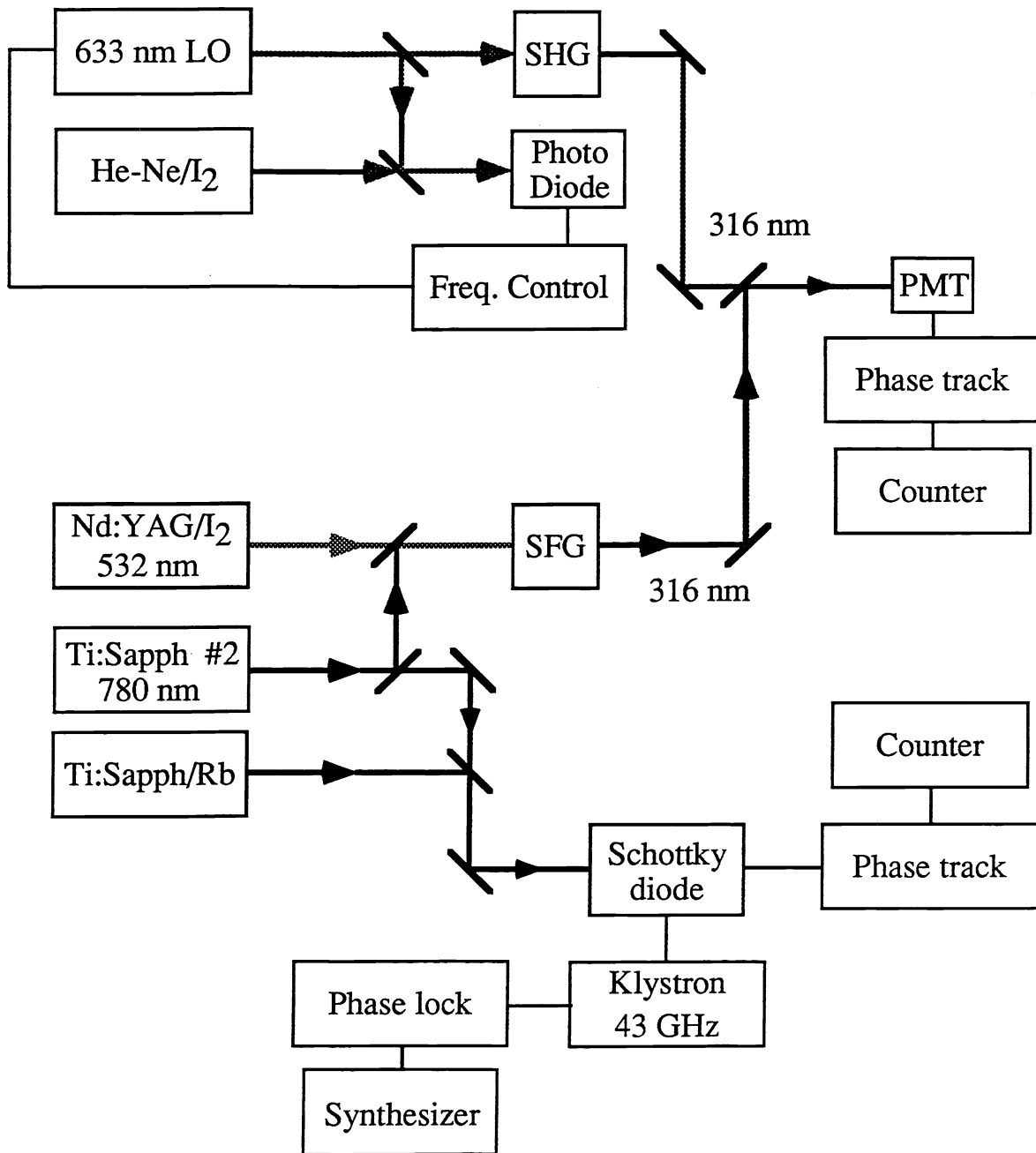


Figure 1.

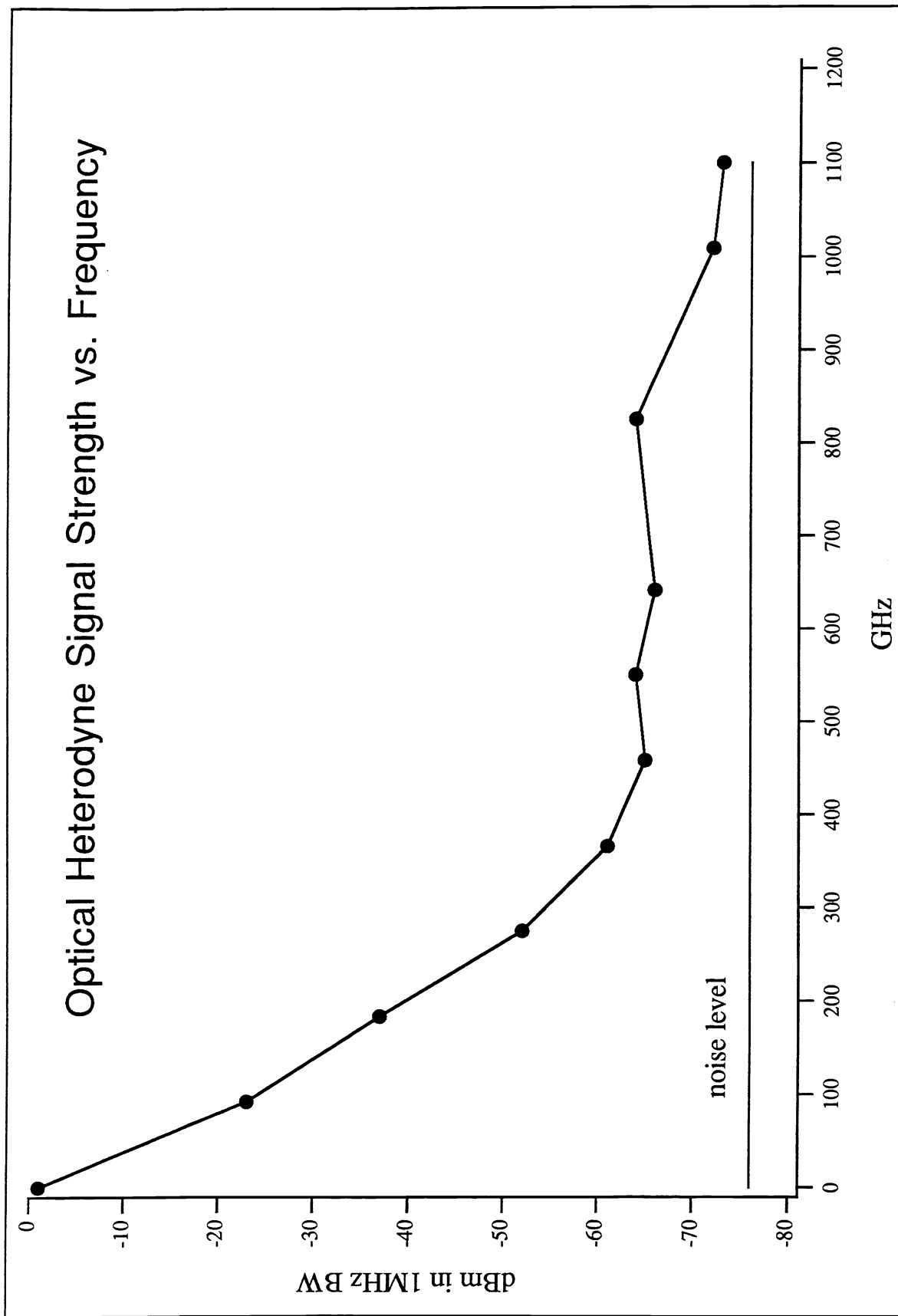


Figure 2.

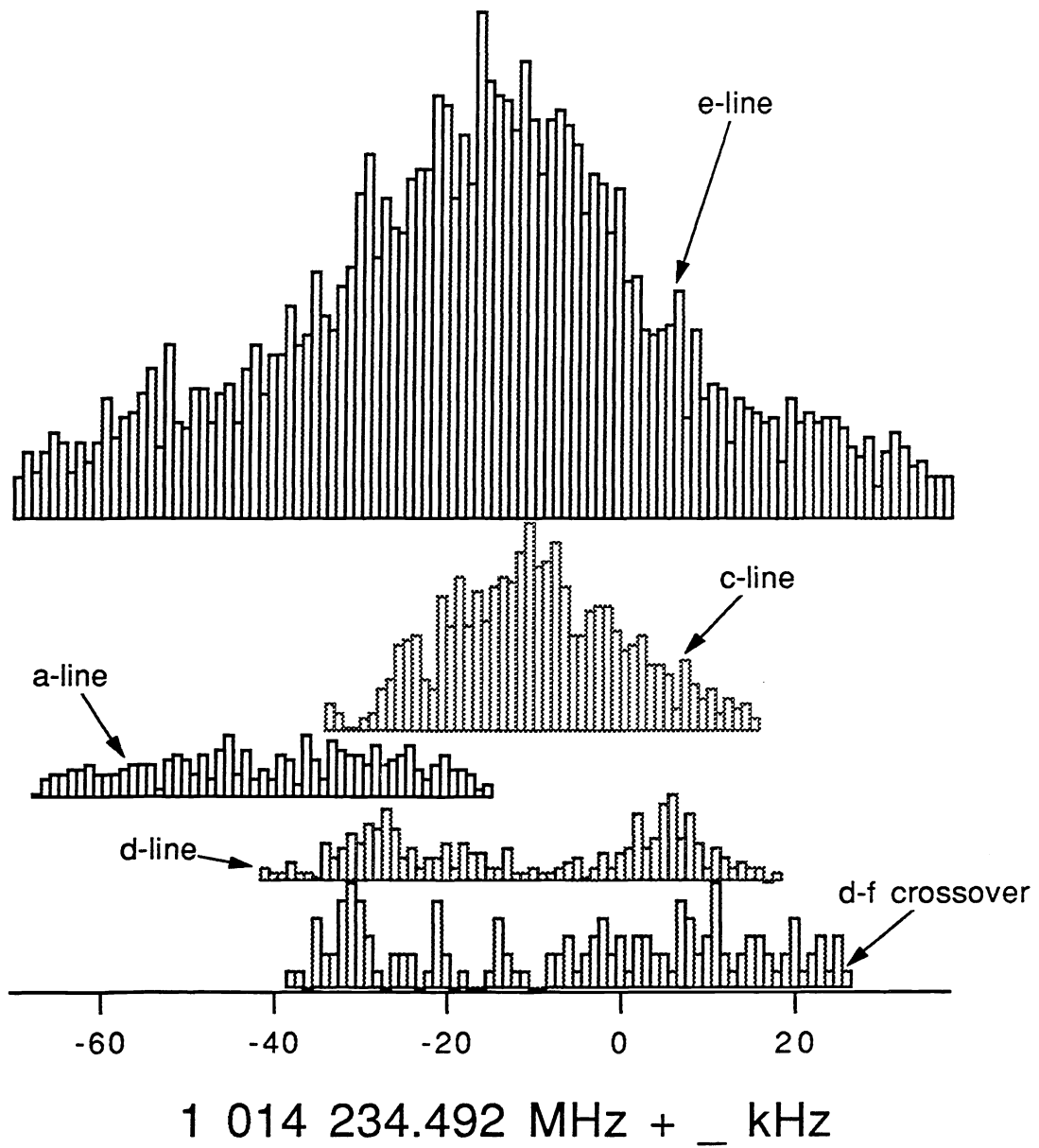


Figure 3.