

## A WIDEBAND FREQUENCY-OFFSET-LOCKED DYE LASER SPECTROMETER USING A SCHOTTKY BARRIER MIXER

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Precise frequency-controlled operation of a single-mode ring dye laser with respect to another is reported at a frequency difference of 234 GHz. This extension of the technique of laser frequency offset locking into the microwave range has been achieved by mixing the two laser lines with the harmonics of a suitable microwave frequency on a Schottky barrier mixer. The capability of this spectrometer was demonstrated by a frequency measurement of the transition  $1s_5 - 2p_8$  in  $^{20}\text{Ne}$  with an uncertainty of  $1 \times 10^{-9}$ .

### 1. Introduction

In precision spectroscopy, laser frequency metrology has several main advantages in comparison to the more commonly used interferometric methods:

1. The measurement of a frequency difference between two lasers is not affected by angle errors or wavefront problems.
2. Molecular energy differences can be directly determined by counting an offset frequency instead of calculating small differences between two wavelength values, thus making the sometimes tedious calibration procedures of interferometers unnecessary.
3. In cases where an absolute optical frequency value is needed, connection can be made to highly precise reference frequencies [1,2].
4. Finally, the well-known technique of frequency-offset locking of lasers allows very high-resolution scanning of a laser across molecular features [3].

Recent successes in using nonlinear diode mixers in the visible spectral range make it possible to measure frequency differences of up to several Terahertz by superposing two visible laser lines and microwave or FIR radiation on a Metal-Insulator-Metal (MIM)

point contact diode [4,5]. GaAs Schottky point diodes show excellent performance in mixing two laser lines with harmonics of an applied microwave frequency [6]. While being superior in terms of sensitivity and stability, the Schottky mixer exhibits a much faster frequency roll-off than MIM structures do [5,6].

We report the first successful operation of a wide-band laser spectrometer using a Schottky diode mixer for frequency spectroscopy in the visible. Radiation from two single-mode ring dye lasers, offset by about 234 GHz, was mixed with the sixth harmonic of the output from a Q-band klystron on a Schottky barrier diode and the resulting beat signal heterodyned with a suitable synthesizer frequency in another mixer. A frequency controlled scan of one dye laser with respect to the other was obtained by tuning the synthesizer output and servoing the dye laser frequency such that the beat frequency from the second mixer was kept constant. The fundamentals of this technique of frequency-offset-locking were first used by Barger and Hall in their work on high-resolution spectroscopy of  $\text{CH}_4$  [3].

**2. Experimental**

A schematical drawing of the experimental set up is given in fig. 1. It can be split into roughly four parts: the lasers, the microwave apparatus, the metastable neon atomic beam, and the mixers and frequency-offset locking loop.

*2.1. The lasers*

Two ring dye lasers (see fig. 1) pumped by a common argon ion laser were used in this experiment. The lasers had a free running linewidth of  $\lesssim 200$  kHz at a 1 ms averaging time and degraded to  $\sim 1$  MHz at 1 s due to the dye pump fluctuations. The lasers were locked to separate confocal cavities by the method of

analyzing the polarization of the beam reflected from the cavity [7]. With simple control loops to piezo-electric driven mirrors, the laser linewidths were held below 200 kHz. With faster electronic servo loops to intracavity AD\*P crystals, the error signals indicated linewidths of order 1 kHz, but the heterodyned beat frequency between the lasers indicated that the laser linewidths remained as large as 100–300 kHz! This apparent difficulty with the polarization lock scheme needs to be further investigated. As might be expected, the baseline is very sensitive to any angle variations of the light beam that is coupled into the reference cavity.

One of the ring dye lasers was used as the long-term-stable reference laser (dye laser I). This laser was locked to its reference cavity by means of the simple

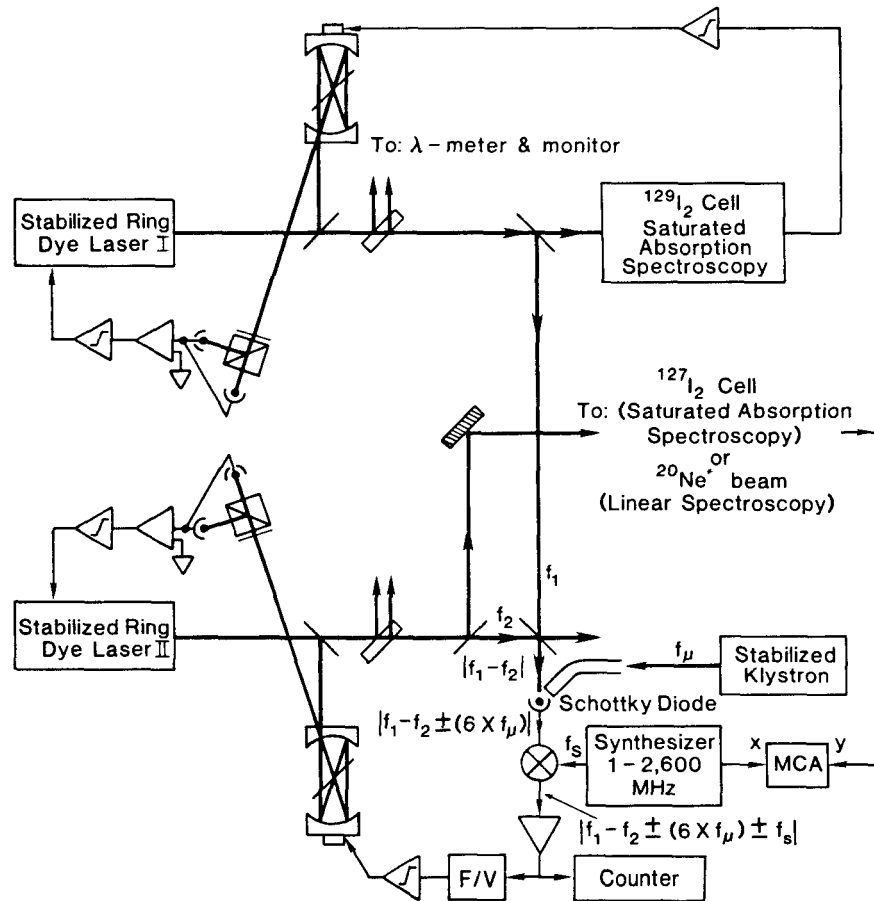


Fig. 1. Large-frequency-difference, high resolution, tunable, optical spectrometer.

servo loop. In turn, the cavity was locked to a narrow hyperfine spectral feature in  $^{129}\text{I}_2$  which provided the long term stability. The first derivative lock to this hfs component, which was located midway between the  $1s_5-2p_8$  transition in  $^{20}\text{Ne}^*$  and the  $i$ th component of the P(33) 6-3 line in  $^{127}\text{I}_2$ , was obtained by the double probe beam method which is first order insensitive to baseline effects and intensity fluctuations [8].

The second dye laser (dye laser II), or the probe laser in our high resolution spectrometer, was locked to its reference cavity also by means of the PZT driven control loop. This reference cavity was servo controlled such that the probe laser frequency was precisely, but tunably, offset from the frequency of the reference laser. This was done by means of the frequency heterodyne offset lock method [3] more fully discussed below. The offset frequency was held stable to  $\lesssim 1 \times 10^{-11}$  at a few seconds. Thus the resolving power of the laser spectrometer is of the order of  $1 \times 10^{11}$  or better and can be improved with narrower linewidth lasers [9] and phase rather than frequency offset locks [3]. We emphasize that the accuracy in our present spectroscopic measurements is limited not by the frequency offset locked laser spectrometer, but by our ability to split any line and to determine line center.

### 2.2. The microwave apparatus

To accomplish the 468 GHz difference frequency measurement with the available Q-band (38-42 GHz) klystron, a two-step procedure for nearly identical frequencies near 234 GHz was chosen. The two slightly different 234 GHz frequencies were generated in the Schottky barrier mixer as a sixth harmonic of the Q-band klystron emission which was phaselocked to the X-band radiation of another klystron at 9.7178394 (9.7739716) GHz with an offset of  $\pm 27.365$  MHz (quantities in brackets and lower signs valid for neon, otherwise for  $^{127}\text{I}_2$ ). The latter klystron was phaselocked to the 82nd (83rd) harmonic of a 118.13219 MHz quartz reference with a  $\pm 31$  MHz offset. Therefore, the two frequencies  $\nu_0$  and  $\nu'_0$  follow from

$$\nu_x(\nu'_x) = 82(83) \times 118.132189 \text{ MHz} \pm 31 \text{ MHz},$$

$$\nu_Q(\nu'_Q) = 4 \times \nu_x(\nu'_x) \pm 27.365 \text{ MHz},$$

$$\nu_0(\nu'_0) = 6 \times \nu_Q(\nu'_Q)$$

$$= 233.392338 (234.411131) \text{ GHz}.$$

The relative accuracy was better than  $10^8$ . The power output of the Q-band klystron was approximately 250 mW, the short-term linewidth about 3 kHz in 20 ms and the stability for an averaging time of 100 s better than  $10^9$ .

### 2.3. The neon metastable beam

To generate and detect neon atoms in the metastable  $1s_5$  state, a converted hydrogen beam machine with a low-energy electron bombarder and a surface ionization detector was used (similar to Lamb's apparatus in his classical experiments on hydrogen [10]). Ground state  $^{20}\text{Ne}$  atoms effused from a small glass bulb (pressure about 250 mT) through a  $8 \times 0.5$  mm rectangular orifice and a 10 mm long electron bombarder. The electron bombarder consisted of a 0.5 mm round thoriated tungsten wire at the bottom edge of a U-shaped, 10 mm high collector. At a dc heater current of 7 A, a collector voltage of 24 V was sufficient to draw a bombarder current of 14 mA. A part of the output of the probe laser was expanded to several cm, collimated, attenuated to  $\lesssim 1$  mW and di-

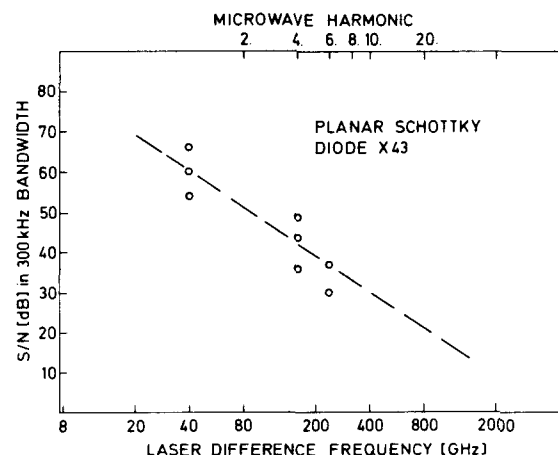


Fig. 2. Signal-to-noise ratios for beats between two visible laser lines and microwave harmonics on the planar Schottky barrier mixer used.

rected perpendicular to the metastable Ne beam about 30 cm behind the e-gun. Metastable atoms not pumped out of the  $1s_5$  state were detected by a surface ionization detector made of a small nickel plate and a copper ring anode. The anode was held at +30 V with respect to the cathode. The angular spread of the detected metastable beam was about 4 mrad determined by the effective cathode diameter of 3 mm. The metastable neon beam typically produced an electrometer current of 0.2 pA. Assuming a detector efficiency of 10%, 0.2 pA corresponds to a beam flux at the detector of about  $4 \times 10^7$  metastable atoms per second. The beam apparatus had a differentially pumped source and detector region. With the Ne beam on, the vacuum was typically  $4 \times 10^{-6}$  mbar in the source region, and  $4 \times 10^{-7}$  mbar in the detector region.

#### 2.4. Schottky mixer and frequency offset lock loop

The Schottky barrier mixer was a 2  $\mu\text{m}$  diameter AuPt anode (300 nm thick) on a 150 nm active layer of n-GaAs ( $N = 2 \times 10^{16} \text{ cm}^{-3}$ ). This epilayer was prepared by vapour-phase epitaxy on a highly doped n-GaAs substrate ( $N = 4 \times 10^{18} \text{ cm}^{-3}$ ) without a buffer layer. From the measured spreading resistance of 9  $\Omega$  and the capacitance of 3 fF a cut-off frequency near 6 THz is expected [11]. Several hundred usable diodes come on a  $0.25 \times 0.25$  mm chip. Electrical contact with any one diode was accomplished with an electrolytically sharpened tungsten tip which also served as an efficient microwave coupling antenna.

Using a microscope objective the two collinear laser beams of about 15 mW each were focussed sharply onto the contact. The laser's heat signal was optimized with the electric field vectors of both lasers lying in the plane of incidence. With 250 mW microwave output power (polarization parallel to the tungsten wire) third order beat signals of up to 70 dB above noise and eighth order mixing signals of up to 37 dB above noise in a 300 kHz bandwidth were observed. The average roll-off of 9 dB/octave compares very favorably to the average 22 dB/octave figure for the point contact diodes of [6]. Contact lifetimes between a few hours and several days were found. Application of a negative bias of -0.9 V helped only in a few cases indicating that the relatively high pressure applied by the tungsten tip led to different diode characteristics.

The laser-microwave beat signals at about 530 MHz ( $^{127}\text{I}_2$ ) and 510 MHz (neon) were broadband-amplified by 33 dB and fed into a doubly balanced mixer. With a suitable frequency from a computer controlled synthesizer, the intermediate frequency fell into the 5 MHz bandpass of a 30 MHz IF amplifier (+40 dB gain) followed by a 10 : 1 scaler. Finally, a frequency-to-voltage converter provided the input for the dye laser servo closing the control-loop.

### 3. Results and discussion

During the experiment the frequency of dye laser I was locked to a hfs feature of a transition in  $^{129}\text{I}_2$  roughly halfway between the frequencies of the neon line and the hfs components of the transition P(33) 6-3 in  $^{127}\text{I}_2$ . Dye laser II was frequency-offset locked and successively scanned over the neon and  $^{127}\text{I}_2$  transitions; fig. 3 shows typical recorder traces of those scans for a synthesizer detuning of 102.3 MHz. Curve 3a gives the saturated absorption line profiles of the hfs components j and i of the transition P(33) 6-3,  $^{127}\text{I}_2$ . This line has a 20 times better S/N ratio compared to the neighboring line R(127) 11-5 [12]. The frequency difference between P(33) 6-3, j and the reference component R(127) 11-5, i was determined in a subsequent set of measurements. While one dye laser was locked to the component r of the P(33) 6-3 line, the other was frequency-offset-locked to it and scanned across the components f through j of P(33) 6-3 and h, i, j of R(127) 11-5. The measured splitting of P(33) 6-3, j to R(127) 11-5, i is  $741.5 \pm 0.3$  MHz, in disagreement with the value of  $740.8 \pm 0.2$  MHz given by Morinaga and Tanaka [12] and  $738.1 \pm 0.35$  MHz given by Hanes et al. [13]<sup>†</sup>. While there is a general agreement between our data of the relative frequency splittings of the hyperfine components and the data of Morinaga and Tanaka, deviations of up to several MHz were obtained from the results of [13]. For instance, for the fre-

<sup>†</sup> The frequency splitting of the components P(33) 6-3, j and R(127) 11-5, i was determined by using the value of  $344.6 \pm 0.35$  MHz for the j-a P(33) 6-3 splitting given in this reference combined with the  $393.5 \pm 0.1$  MHz splitting of the components P(33) 6-3, a and R(127) 11-5, i given by Morinaga and Tanaka [12]. Hanes et al. measured the P(33) 6-3, a to R(127) 11-5, i splitting to be  $394 \pm 4$  MHz.

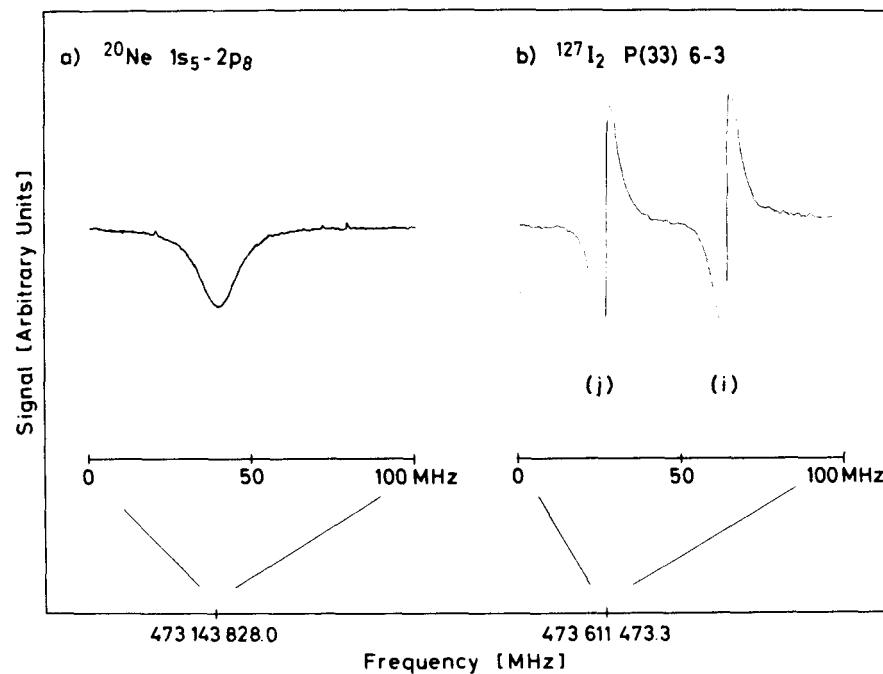


Fig. 3. Recorder traces of frequency-offset-controlled dye laser scans across a) the transition  $1s_5-2p_8$  in  $^{20}\text{Ne}$ ; b) the hfs components j and i of the transition P(33) 6-3 in  $^{127}\text{I}_2$  (see text).

quency difference between P(33) 6-3, r and R(127) 11-5, i we find  $1053.8 \pm 0.2$  MHz in good agreement with  $1053.9 \pm 0.3$  MHz given by Goldsmith et al. [14], where Hanes et al. measured  $1048.6 \pm 0.35$  MHz. (Similar deviations have been found between the result of [13] and calculations of these splittings from molecular constants [15].) In this paper we adopt the value of  $741.5 \pm 0.3$  MHz for the frequency separation between the components P(33) 6-3, j and R(127) 11-5, i. Combining this frequency with the applied microwave frequency  $\nu_0$ , the synthesizer start frequency, the offset frequency of the servoloop and the frequency to the measured line position results in the value 233 552.6 MHz for the splitting between the intermediate hfs component of  $^{129}\text{I}_2$  and the component R(127) 11-5, i in  $^{127}\text{I}_2$ .

To find the uncertainty level of this number, various sources of error have to be taken into account. Unfortunately, there is only rather limited knowledge about the influence of laser power, modulation width, iodine vapour pressure or cell temperature on the frequency of the transition P(33) 6-3 in an external cell.

Estimates of the errors have therefore to be inferred from the numerous investigation on the 632.8 nm HeNe laser stabilized to various components of R(127) 11-5,  $^{127}\text{I}_2$  [16].

In our experiments, the saturating beam of 2.2 mW into a  $\approx 200$   $\mu\text{m}$  spot corresponded to a pump intensity of  $\approx 7$  W/cm<sup>2</sup>. From the CCDM recommendation of a one-way intracavity power of 20 mW for the HeNe reference laser (meaning an intensity of 13 W/cm<sup>2</sup> in the usual 30 cm long semiconfocal cavity) together with the measurements of Layer et al. we conclude that the transition frequency cannot be shifted up by more than 100 kHz [1,17]. An estimate for the influence of both the iodine vapor pressure and the laser modulation width follows from measurements by Helmcke [18]: Under our conditions, viz., cooling finger temperature of 300 K and 3 MHz modulation width, the frequency of the Ne line would be shifted down by about 180 kHz. The combined uncertainty of the two shifts is estimated to be of the order of  $\pm 100$  kHz. Frequency shifts caused by the cell temperature and by contamination of the carefully pre-

pared iodine cell are considered negligible. Similarly, frequency shifts and errors caused by intensity fluctuations and baseline slopes are nearly eliminated by the aforementioned double probe differencing technique. Finally, a scatter of  $\pm 200$  kHz in our results and an estimated uncertainty of  $\Delta f/f = 2 \times 10^{-10}$  ( $f$ , laser frequency) for the lockpoint of dye laser I must be taken into account. Summing up all these uncertainties in quadrature the uncertainty of the iodine beat frequency measurement is roughly  $6 \times 10^{-10}$ .

Curve b in fig. 3. shows a 30% dip in the electrometer current caused by pumping neon atoms from the metastable  $1s_5$  state to the shortlived  $2p_8$ . Despite the small detector current of about 0.2 pA, the observed S/N ratios were of the order of 150/1. The observed linewidth of 14.5 MHz FWHM contains a natural linewidth contribution of about 10 MHz and a gaussian contribution of  $\approx 5$  MHz from the residual doppler spread of the atomic beam. There is another gaussian contribution of about 6 MHz, probably due to power broadening, even though the power in curve 3b was  $\leq 5 \times 10^{-4}$  W/cm<sup>2</sup>.

From the microwave frequency  $\nu_0$ , the synthesizer start frequency, the offset-frequency of the servoloop and the measured frequency of the neon line we calculate a frequency difference of 234 834.2 MHz to the  $^{129}\text{I}_2$  iodine feature. In this case, the lockpoint uncertainty of dyelaser I and all other errors can be neglected compared to the uncertainty of  $\pm 500$  kHz resulting from a possible 0.5 mrad misalignment in the orthogonality of the laser and atomic beams. The sum of the two beat frequencies gives the value of 468 386.8 MHz for the frequency difference between the transitions  $^{20}\text{Ne } 1s_5-2p_8$  and  $^{127}\text{I}_2$  R(127) 11-5, i. Thus, with the value of 473 612 214.8 MHz for the absolute frequency of the component i of the R(127) 11-5 line in  $^{127}\text{I}_2$  the resulting frequency value for the Ne  $1s_5-2p_8$  transition is 473 143 828.0  $\pm 0.6$  MHz. This compares well with a recent interferometric measurement of the same line which gave a wavelength of 633.618026 nm corresponding to a frequency of 473 143 827.8 MHz [19]. In this case, the uncertainty is claimed to reach the  $10^{-9}$  level too.

#### 4. Conclusion

We have demonstrated for the first time that high

resolution, large frequency-difference, precision measurement in the visible can be performed using the novel method of beating two dye lasers and microwave harmonics on a Schottky diode mixer. With microwave sources of higher output frequency or far-infrared lasers as IF oscillators the bandwidth of our frequency-offset-lock spectrometer can be readily extended into the Terahertz range. Finally, the achievable accuracy of this technique is ultimately limited only by the uncertainties of the reference frequencies used. For these reasons, this spectrometer appears extremely useful, for instance, in precise tests of the "calculable" spectra of H, D, He<sup>+</sup>, Li<sup>++</sup> or the like.

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