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# New Ultra-High Resolution Dye Laser Spectrometer Utilizing a Non-Tunable Reference Resonator

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**Abstract.** A new dye laser spectrometer utilizing a non-tunable reference resonator is described. The resonator consists of two Zerodur mirrors optically contacted to a Zerodur spacer. Frequency scanning of the laser is provided by acoustooptic modulation. Residual drifts of the resonator frequency – measured on line – are compensated automatically by corresponding corrections of the modulation frequency. The stability during several hours and the resettability of the dye laser frequency are $\pm 2.5$ kHz and $\pm 10$ kHz, respectively.

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Dye lasers are now widely used in many applications of laser spectroscopy. When high spectral resolution, low frequency drift and precise frequency scanning are required the laser frequency is usually stabilized by a fast servo control to a suitable eigenfrequency of a stable optical resonator. Very narrow laser line widths have been observed if the error signal of the stabilization was derived by means of a phasemodulation technique [1–5]. Long-term stability and frequency scanning can then be achieved by locking a suitable eigenfrequency of this resonator to the frequency of a He–Ne laser which is frequency-offset locked to a $I_2$-stabilized He–Ne laser. However, the use of a tunable reference resonator can increase the short term frequency noise of the laser. It would therefore be favourable to utilize a stable resonator with no tuning facilities.

In this paper, we describe a new dye-laser spectrometer using such a non-tunable resonator. In our approach, frequency scanning of the laser is provided by acoustooptic (ao) modulation. The residual drift and slow frequency fluctuations of the resonator are measured and corrected automatically by changing the ao modulation frequency. With this system, we have achieved a resettability of the laser frequency of $\pm 10$ kHz.

## 1. Concept of the Dye Laser with Non-Tunable Reference Resonator

Fast stabilization of the dye-laser frequency to a suitable fringe of a stable, high-finesse optical resonator is now widely applied in order to reduce the frequency noise and the spectral width of the laser emission. Utilizing Pound's "phase modulation technique" [3] – originally developed to stabilize the frequency of microwave oscillators – laser line widths in the subkilohertz range have been reported for short observation times below about 1 s [1, 2]. Many applications – in particular when ultra-high spectral resolution is needed – require longer observation times of several minutes up to hours depending on the signal strength. In these cases, the frequency drift and random walk of the reference resonator can not be neglected compared to the laser line width and the spectral resolution of the spectrometer is degraded as the interrogation time increases. These long-term fluctuations may be caused by aging of the resonator

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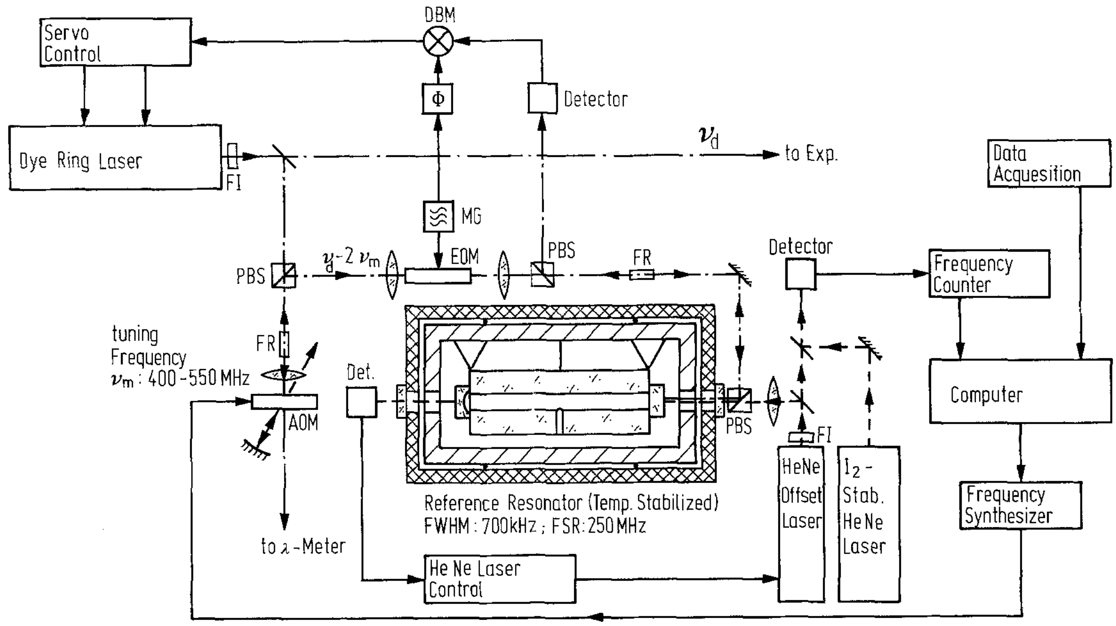


Fig. 1. Block diagram of the dye-laser spectrometer (PBS: polarizing beam splitter, FR: Faraday rotator, DBM: double balanced mixer, AOM: acoustooptic modulatir, EOM: electrooptic modulator, MG: modulation-frequency generator)

structure, imperfect temperature control, and/or resonator heating by absorbed laser power. They are usually reduced by stabilizing the cavity to a He–Ne laser which in turn is offset locked to an $I_2$-stabilized He–Ne laser standard [7, 8]. Precise tuning can then be achieved by changing the offset frequency of the tunable He–Ne laser [6].

This method of long-term stabilization and frequency scanning requires a tunable reference resonator. The incorporation of tuning means, such as PZT-mirror mounts, often degrades the free-running stability of a resonator. Furthermore, the short-term stability of the $I_2$-stabilized He–Ne laser is usually relative poor and its frequency noise may inadvertently be transferred to the resonator thereby degrading the short-term stability of the dye laser. Careful low-pass filtering of the cavity lock and critical adjustment of its unity-gain frequency would be necessary to achieve both short-term and long-term stability. Consequently, the maximum scanning rate of the dye-laser frequency is also limited.

The new dye-laser spectrometer shown in Fig. 1 reduces many of these problems, and allows independent optimization of both the long-term and short-term stability. The spectrometer is based on a very stable, non-tunable resonator mounted vibration isolated in a temperature controlled vacuum chamber. Omitting tuning elements of the resonator minimizes its intrinsic frequency noise. Tuning of the stabilized dye laser can then be performed by acoustooptic or electrooptic frequency shifting [9] the part of the laser beam which is split off from the main beam and mode matched into the resonator to stabilize the laser frequency. Residual random walk and slow fluctuations of the resonator frequency are monitored by stabilizing the frequency of a He–Ne laser to a suitable eigenfrequency of the resonator and measuring the beat frequency between this laser and an $I_2$-stabilized He–Ne laser.

The measured longterm changes of the resonator frequency can be compensated on line by corresponding changes of the modulation frequency at the frequency shifter. Moreover, provided the cavity orders to which the two lasers are frequency stabilized remain unchanged any chosen dye-laser frequency can be reset by setting the modulation frequency to the value calculated from the original pair of beat and modulation frequency and the actual measured beat frequency.

## 2. Experimental Setup

A schematic, simplified diagram of the laser spectrometer is shown in Fig. 1. The home-made single-frequency dye ring laser contains an intracavity AD*P phase modulator and a PZT-driven mirror mount serving as fast and slow tuning elements [6], respectively. Using the dye DCM, a typical useful output power of more than 100 mW is observed at 657 nm wavelength and a pump power of 4 W.

Fast frequency stabilization to a suitable fringe frequency is performed by means of Pound's sideband technique [3]: About 10% of the laser power – split off the main beam – passes through a polarizing beam splitter (PBS), a 45° Faraday polarization rotator (FR),

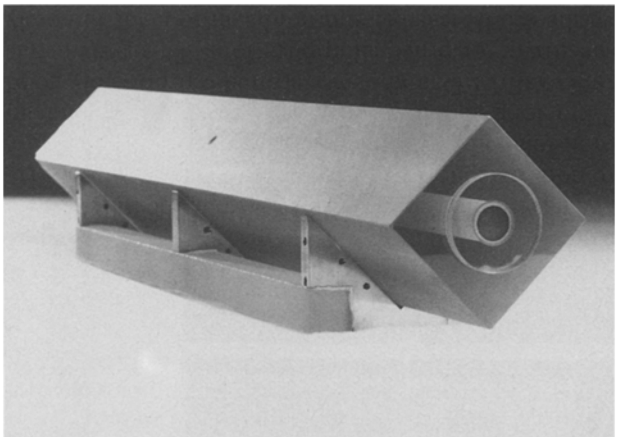

Fig. 2. Reference resonator

and is then focussed to a broad-band acoustooptic modulator (AOM). The diffracted beam is retroreflected in itself by a concave mirror, carefully focussed. This returning beam which is frequency shifted by twice the modulation frequency is deflected at the PBS, phase modulated by an electrooptic modulator (EOM) and modematched into the reference resonator. Using the "autocollimation" method of double passing through an AOM, the position and diameter of the returning beam are kept constant (to first order) when the laser frequency is scanned. Correspondingly, the mode matching condition changes only little with the modulation frequency. About 15% to 20% of the power incident to the AOM is fed to the resonator. The beam reflected at the resonator is finally focussed on a photo detector observing the amplitude modulation which is converted near the resonance from the phase modulation. The corresponding modulated photocurrent is phase sensitively detected by a double balanced mixer (DBM) and fed to the servo control which in turn locks the laser frequency to the centre of the cavity fringe. Correspondingly, the main beam is frequency offset by twice the modulation frequency driving the AOM. The frequency response of the servo loop drops to unity gain at about 2.5 MHz.

The reference resonator shown in Fig. 2 consists of two Zerodur [10] mirrors optically contacted directly to a stable Zerodur spacer. The length and the cross section of the spacer are 60 cm and 6 cm × 6 cm, respectively. It has a central, longitudinal bore of 15 mm diameter and a small side bore. The entrance mirror is flat whereas the second mirror has a curvature of 95 cm radius providing nondegeneracy of the transversal modes for $m+n \leqq 7$ ($m, n$ being the order numbers of transversal modes). Both mirrors have reflectivities of 99.2% at $\lambda = 657$ nm. Correspondingly, the measured resonance width is 700 kHz (FWHM) at a free spectral range of 249.481 MHz. Similarly, as described by Hough et al. [1] the resonator is supported by a cradle hanging on five steel wires in an aluminium tube of 21 cm diameter and 6 mm wall thickness. The aluminium tube acting as a heat shield is placed in a vacuum chamber continuously pumped by an ion getter pump. The temperature is held constant to $\pm 10$ mK at about 7 K above room temperature.

The measured thermal expansion coefficient of the resonator spacer is about $-4 \times 10^{-8}\,\mathrm{K}^{-1}$ corresponding to a drift rate of $+20\,\mathrm{MHz} \cdot \mathrm{K}^{-1}$. Therefore, a temperature stability in the range of $\pm 10^{-5}$ K is required in order to achieve sub-kHz frequency stability. At this level, heating of the resonator by absorbed laser power cannot be neglected. The adiabatic heating rate of the described resonator is already 22 nK/s corresponding to a frequency drift of +0.4 Hz/s. The thermal time constant of the resonator was measured to be in the range of 10 h. Aging of the Zerodur spacer [11] caused a frequency drift between +20 Hz/s at the beginning and less than 2.3 Hz/s about 500 days after assembling the resonator.

To monitor the resonator frequency, the beam of the He–Ne offset laser (Fig. 1) is also mode matched into the resonator and its frequency is locked to the centre of a suitable cavity fringe by means of conventional third-harmonic locking techniques. Frequency fluctuations of the resonator are then measured by counting the beat frequency $\nu_b$ between this laser and an $I_2$-stabilized He–Ne laser. In order to compensate the measured frequency drift of the reference resonator, the modulation frequency $\nu_m$ at the AOM is changed correspondingly. The relation between the frequencies $\nu_b$ and $\nu_m$ can easily be calculated: Neglecting the diffraction of the resonator and phase dispersion effects of the mirrors, the dye laser frequency $\nu_d$ may be expressed by

$$\nu_d = n \cdot c/(2 \cdot d) + 2\nu_m .$$

The frequency of the He–Ne laser $\nu_{\mathrm{HeNe}}$ is given by

$$\nu_{\mathrm{HeNe}} = \nu(\mathrm{I}_2) - \nu_b = qc/(2d) .$$

In both equations $n$ and $q$ are integers with $n/q$ being approximately the wavelength ratio $\lambda_{\mathrm{HeNe}}/\lambda_d$ of the He–Ne laser and the dye laser, $c$ is the speed of light, $d$ the length of the resonator, and $\nu(\mathrm{I}_2)$ is the frequency of the $I_2$-stabilized laser. The dye laser frequency can now be expressed by

$$\nu_d - \frac{\lambda_{\mathrm{HeNe}}}{\lambda_d} \cdot \nu(\mathrm{I}_2) = 2\nu_m - \frac{\lambda_{\mathrm{HeNe}}}{\lambda_d} \nu_b . \tag{1}$$

For a fixed dye-laser frequency, the left-hand side of (1) is constant. Correspondingly, there exists a characteristic pair of $\nu_m$ and $\nu_b$ for each dye laser frequency.

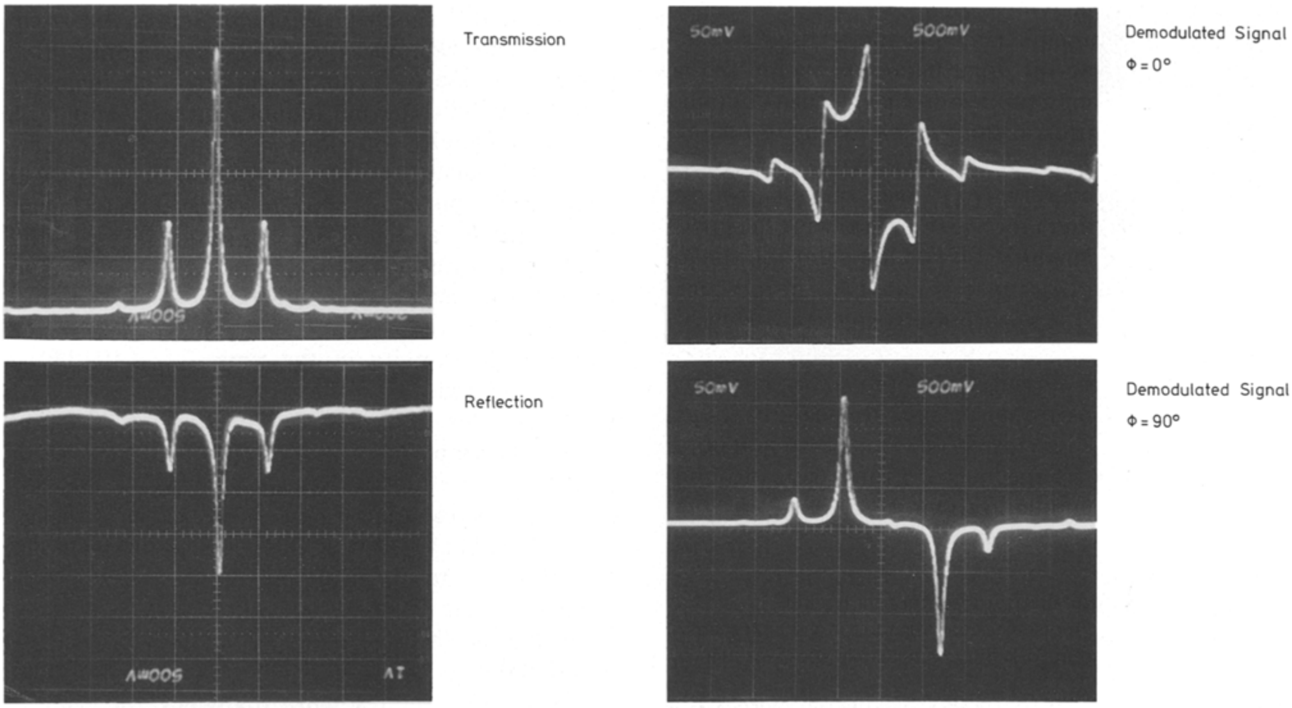


Fig. 3a–d. Photosignal vs. laser frequency observed by using the sideband technique. Photocurrent of the laser power transmitted (a) through the cavity, reflected at the cavity (b), demodulated signals (c) in phase, (d) in quadrature with the voltage at the EOM

According to (1) a selected dye-laser frequency $\nu_d$ can always be reset by measuring $\nu_b$ and setting $\nu_m$ to the value calculated from this equation. With the spectrometer in the frequency scanning mode, the beat frequency is continuously measured and the modulation frequency $\nu_m$ is corrected before each scan, automatically. Frequency scanning, drift compensation and data acquisition are performed by means of a desk computer.

## 3. Results

In order to analyze the side-band stabilization technique, we have first investigated the signals observed by using a phase-modulated dye-laser beam scanned over the resonance of the cavity. Curves a and b in Fig. 3 show the detected photosignals of the laser beam transmitted through or reflected at the resonator, respectively, as a function of the laser frequency. The side bands generated by the 15 MHz phase modulation are clearly resolved. The other two curves c and are generated by the reflected laser beam after detection and demodulation by the DBM (Fig. 1). These oscillograms show the signals in phase (c) and in quadrature (d) with the modulation voltage applied to the EOM. The central zero crossing of the dispersion like curve in Fig. 3c is used to stabilize the laser frequency.

In applications, where the band width of the servo control is higher than the width of the cavity resonance, the transient behaviour has to be taken into account. To calculate the transfer function of the discriminant curve of Fig. 3c at the line centre, we assume that the laser frequency is modulated by noise of a small modulation index $\Phi_F$ and a Fourier frequency $\nu_F$. Correspondingly, the laser beam incident to the resonator

$$E_i = E_0 \exp\{i[\omega t + \Phi_m \sin(\omega_m t) + \Phi_F \sin(\omega_F t)]\}$$

is composed of nine frequencies: $\omega$, $\omega \pm \omega_F$, $\omega \pm \omega_m$, $\omega + \omega_m \pm \omega_F$, $\omega - \omega_m \pm \omega_F$ ($\omega_m$: modulation frequency at EOM, $\Phi_m$ index of phase modulation). The amplitudes of the corresponding signals reflected at the resonator can be calculated for each sideband according to

$$\frac{E_r}{E_i} = \frac{r(1-e^{-i\Delta})}{1-r^2 e^{-i\Delta}} \tag{2}$$

with $\Delta = (2d/c) \cdot \Delta\omega$, $r$ being the amplitude reflection coefficient of each of the two mirrors and $\Delta\omega$ the detuning from the resonance. The slope of the discriminant curve at line centre (Fig. 3c) is then determined by those terms of the reflected intensity which are modulated with $\omega_m \pm \omega_F$.

As a result, the amplitude $A(\nu_F)$ and the phase $\Phi(\nu_F)$ of the transfer function at line centre are finally calculated to depend on the Fourier frequency according to

$$A(\nu_F) = A(0)(1 + \nu_F/\nu_{1/2})^{-1/2}, \quad \Phi(\nu_F) = -\arctan(\nu_F/\nu_{1/2}). \tag{3}$$

This equation describing the transient response of the discriminant curve of the side-band stabilization method corresponds to the transfer function of an integrating low pass filter were the cutoff frequency is equal to the half width at half maximum $\nu_{1/2}$ of the resonance. Consequently, the transient response of the resonator can be compensated in the electronic servo control by a simple proportional term to get a well defined integrating (or double integrating) behaviour of the total servo loop. The maximum unity gain frequency is therefore not limited by the resonance width of the cavity.

In order to investigate the performance of the stabilization we have detected the beat frequency of two lasers – a He–Ne laser ($\lambda = 633$ nm) and a dye laser tuned close to the He–Ne laser emission – both stabilized to suitable eigenfrequencies of two individual reference cavities. The side-band stabilization technique was applied. During these initial experiments conventional reference cavities with mirrors mounted on PZT transducers were used. The resonators were supported vibration damped in airtight boxes. No temperature control was provided. The frequencies of the phase modulation applied to the beams of the dye laser and the He–Ne laser were 15 and 5 MHz, respectively. Both frequencies were larger than the resonance width of 2.2 MHz (FWHM) of the cavities. The unity gain of the servo locks were 2.5 MHz for the dye laser and 80 kHz for the He–Ne laser. The beat signal had a full spectral width at half maximum of less than 5 kHz. Figure 4 shows the two sample standard deviation $\sigma(\tau)$ versus the sampling time $\tau$. Both reference resonators were free running. The PZT of one resonator was grounded whereas the other one was biased by a few hundred volts. At sampling times below about 0.1 s the beat frequency fluctuations are lower than 1 kHz. At longer integration times, however, $\sigma(\tau)$ increases according to a measured drift rate of the beat frequency of 2.5 kHz/s.

The frequency change of the new non-tunable resonator, described in Sect. 2 was measured during several months by monitoring the beat frequency $(\nu_I - \nu_R)$ of an $I_2$-stabilized He–Ne laser $\nu_I$ and a He–Ne offset laser locked to a resonance frequency $\nu_R$ of the cavity (Fig. 1). As shown in Fig. 5, the length of the resonator decreases with time. The measured drift rate 500 days after the resonator had been set up was 2.3 Hz/s averaged over several months. This value is three orders of magnitude smaller compared to that of the conventional tunable resonators described above. With the dye laser spectrometer in use, the rate of frequency change can be higher than 2.3 Hz/s due to changes of the room temperature which are not completely suppressed by the present temperature control (see below).

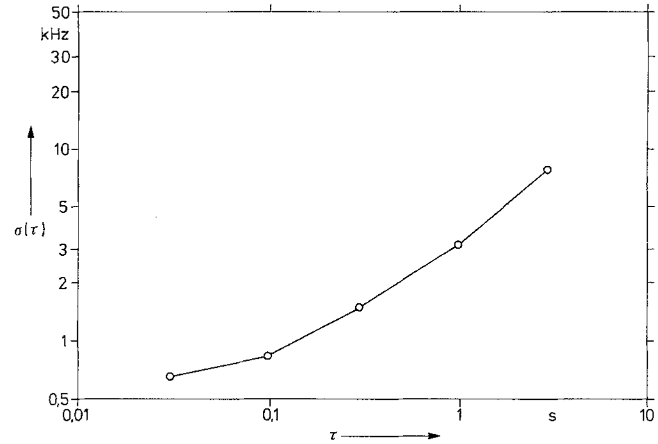


Fig. 4. Two sample standard deviation $\sigma(\tau)$ vs. sampling time $\tau$ of the beat frequency of a dye laser and a He–Ne laser stabilized to individual reference resonators comprising PZT mirror mounts

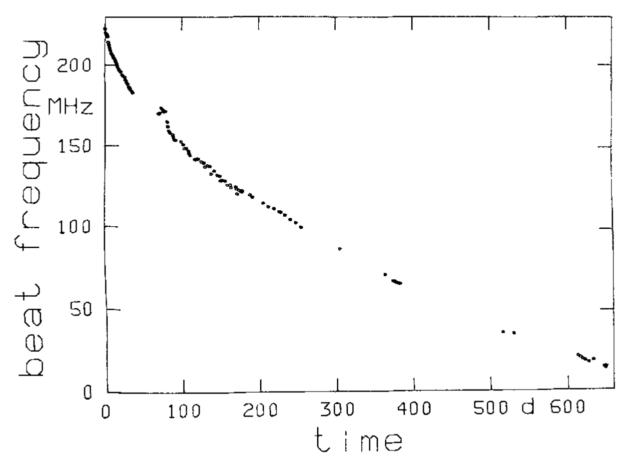


Fig. 5. Beat frequency vs. time between a He–Ne laser locked to the non-tunable resonator and an $I_2$-stabilized He–Ne laser. The drift rate of the resonator 250 days after assembling is 2.5 Hz/s (decreasing beat frequency corresponds to decreasing resonator length)

Figure 6 shows the two sample standard deviation $\sigma(\tau)$ versus the sampling time $\tau$ of the beat frequency between a He–Ne laser locked to an eigenfrequency of the reference resonator and the $I_2$-stabilized laser. $\sigma(t)$ shows a minimum of about $2 \times 10^{-12}$ ($\simeq 1$ kHz) for $10\,\mathrm{s} < \tau < 30$ s. Consequently, a sampling time of 10 s was used to compensate the fluctuations of the resonator when the spectrometer was used. The increase of $\sigma(\tau)$ at larger $\tau$ seems to represent the residual frequency changes of the resonator.

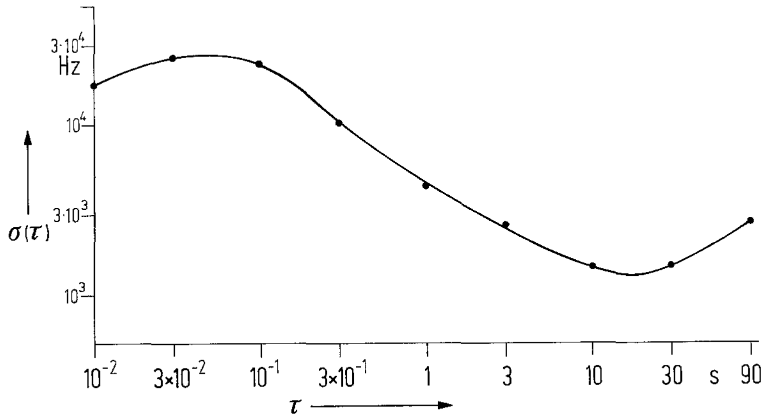


Fig. 6. Two sample standard deviation $\sigma(\tau)$ vs. sampling time $\tau$ of the beat frequency of a He–Ne laser locked to the non-tuneable resonator and an $I_2$-stabilized He–Ne laser

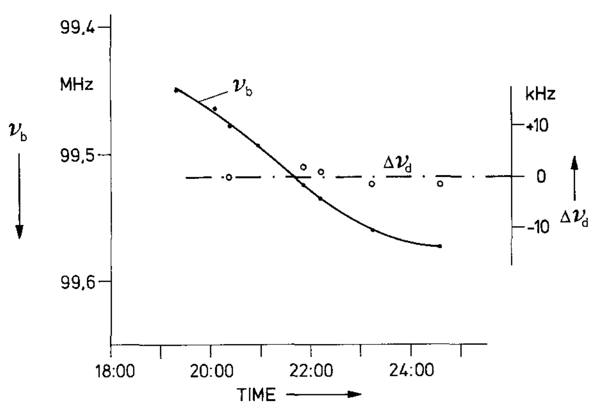


Fig. 7. Long-term frequency fluctuations of the dye laser and the resonator ($\nu_b$: beat frequency, $\Delta\nu_d$: fluctuations of the dye-laser frequency)

To investigate the frequency drift and the resetability of the complete dye laser spectrometer the $({}^3P_1 - {}^1S_0)$ Ca intercombination line at 657 nm wavelength [13] was chosen as reference frequency. Detecting either the saturated absorption signal or optical Ramsey fringes [14], the Ca line centre can be determined. Frequency changes of the dye laser are then easily measured by comparing the AOM modulation frequency $\nu_m$ needed to tune the dye laser to the line centre with the expected value determined from the beat frequency $\nu_b$ according to (1). Figure 7 shows the frequency stability of the dye laser spectrometer: During a typical measurement interval of more than five hours the frequency of the resonator measured via beat frequency $\nu_b$ changed about 120 kHz. The corresponding frequency fluctuations of the stabilized dye laser of only $\pm 2.5$ kHz clearly demonstrate the high-resolution potential of the spectrometer at long observation times.

The frequency resettability of the dye-laser spectrometer was measured during a time interval of more than three months. Figure 8 shows the drift of the reference resonator frequency in terms of $\nu_b$ and the corresponding frequency fluctuations $\nu_d$ of the stabilized dye laser. Although the resonator frequency increased by about 28 MHz the dye-laser frequency was always set to the same value within an uncertainty of about $\pm 10$ kHz.

This high resettability is of the same order as the reproducibility of the $I_2$-stabilized reference laser itself. The presented method simplifies and improves the practical use of the spectrometer extraordinarily: In order to reset the laser frequency to within $\pm 2 \times 10^{-11}$ (10 kHz), it needs to be locked to the correct cavity order, only. With its free spectral range of 250 MHz,

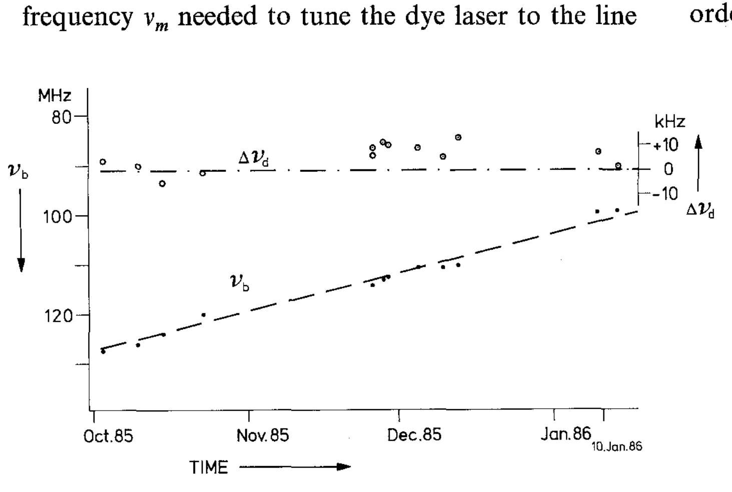


Fig. 8. Resettability of the dye-laser frequency and corresponding drift of the resonator, measured over a period of 3.5 months ($\nu_b$: beat frequency, $\Delta\nu_d$: fluctuations of the dye laser frequency)

this order can be selected by means of a lambda-meter [14] of $10^{-7}$ resolution.

## 4. Conclusion

We have described a new dye-laser spectrometer utilizing a stable, non-tunable reference resonator. Stabilization of the laser frequency to a suitable resonance frequency of this resonator is performed by means of Pound's side-band technique. Under operating conditions, the drift rate of the passive resonator is about three orders of magnitude smaller compared to conventional PZT-tuned resonators. Frequency tuning is performed by acoustooptic frequency shifting the dye-laser beam modematched into the cavity. The drift of the resonator – measured by the beat frequency between a He–Ne offset laser locked to the resonator and an $I_2$-stabilized He–Ne laser – is compensated automatically by corresponding corrections of the modulation frequency at the AOM.

A frequency stability within $\pm 2.5$ kHz was observed during a measurement time of 5.5 h. The resettability of the laser frequency of $\pm 10$ kHz is of the order of the reproducibility of the used He–Ne frequency standard. This spectrometer is very useful in applications where very weak signals have to be investigated at high resolution and long integration times. For example, it has been used to detect pulsed optical Ramsey fringes of 12 kHz FWHM in Ca [16]. Due to the poor Ramsey contrast caused by the pulsed excitation, a total measurement time in the order of 40 min was required. Even at these long times no degradation of the Ramsey contrast due to residual frequency changes was observed.

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