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# Optically stabilized tunable diode-laser system for saturation spectroscopy

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**Abstract.** We present an optically stabilized lead-salt diode-laser system which is the nucleus of a very-high-resolution instrument for sub-Doppler molecular spectroscopy in the mid-infrared. By application of external optical feedback, we have narrowed the diode-laser linewidth by two orders of magnitude, yielding a spectral width of less than 200 kHz. The diode- laser frequency is stabilized and controlled via the external reflector by variable-frequency offset-locking the diode-laser to a CO laser frequency. This substantial improvement in the spectral properties enabled us to perform a Lamb-dip experiment on a carbonyl sulfide (OCS) absorption line near 1985 $cm^{-1}$. We were able to detect a saturated dispersion signal at low pressure (5 Pa) with a signal-to-noise ratio of several thousand. The present paper describes the unique features of the optically stabilized tunable diode-laser system and its use as a spectroscopic tool for sub-Doppler applications.

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Lead-salt Tunable Diode Lasers (TDL) are one of the most important tools for high-resolution molecular spectroscopy in the mid-infrared. Depending on their composition, they cover the wavelength region from 3 to 30 $\mu$m, thus rotational-vibrational transitions of most molecules may be investigated by TDL spectrometers. In the past decade TDL spectrometers have extensively been used for conventional gas phase absorption spectroscopy using a standard frequency-modulation technique. The spectral resolution of a TDL spectrometer is generally limited by the finite spectral width of the TDL radiation. Since most lead-salt diode lasers exhibit large linewidths in the order of 10 MHz (FWHM) and more, spectroscopic applications are normally restricted to Doppler-limited studies where the TDL linewidth can be neglected in comparison with the much broader spectral width of the molecular absorption lines under observation. The spectral purity of usual lead-salt diode lasers is inadequate to resolve sub-Doppler spectroscopic features like saturated absorption signals. Experiments using a TDL as saturating laser have not been published except for one early report by Jennings in 1978 [1]. He observed a 40 MHz wide dip at the center of a strong $NH_3$ line near 888 $cm^{-1}$ which was interpreted as a saturation dip.

At our laboratory we are particularly interested in detecting very narrow and high-contrast saturated-absorption signals on transitions of calibration gases like CO, OCS, $N_2O$, etc. This permits a very accurate determination of the line position in a heterodyne frequency measurement [2]. For these experiments molecular gas lasers generally are employed as the saturating lasers. They usually exhibit a linewidth in the order of 100 kHz. Recently the generation of sidebands to the $CO_2$ laser and CO laser has also been used to perform sub-Doppler heterodyne frequency measurements [3–4]. Many spectral regions that cannot be covered by gas lasers plus sidebands are attainable only with tunable diode lasers.

In order to improve the spectral resolution capabilities of a TDL spectrometer one has to substantially improve the spectral purity of the diode lasers. In comparison to the well-studied III–V semiconductor lasers (based on GaAs and InP), lead-salt diode-laser research still is not in the mature stages. The spectral width normally reported for lead-salt lasers may vary from 1 MHz to more than 100 MHz [5–7]. One group observed linewidths less than 100 kHz for carefully selected high-power diodes [8]. The causes of this dramatic variation in the linewidths of diode lasers of the same type are not well-understood.

In recent years, several frequency stabilization schemes for TDL systems have been developed. They use *electrical* feedback servo loops to control the diode-laser frequency through the diode current. Most of them are based on the stabilization to a tunable Fabry-Perot interferometer [9–10] or on a frequency-offset locking scheme [8, 11]. These techniques provide a good wavelength or frequency calibration and are able to reduce slow frequency drifts and jitter of the laser line. This includes the jitter which is introduced by mechanical vibrations of the closed-cycle cooler. However, the fast inherent diode-laser linewidth is not affected by these methods due to the insufficient feedback-loop bandwidth (normally on the order of 10 kHz).

This situation initiated the investigations of lead-salt diode lasers with external *optical* feedback at our laboratory. This technique is often employed to reduce the linewidth of

III–V semiconductor lasers [12–14] but it was not known before if this method is applicable to improve the spectral properties of tunable diode lasers, too. We recently reported our first results on line narrowing of lead-salt lasers by optical feedback [7]. We have shown in this preceding work that under appropriate conditions it is possible to achieve considerable linewidth reduction for lead-salt diode lasers by applying controlled optical feedback. We succeded in narrowing the linewidth of 3.3 and 5.1 $\mu$m diode lasers from around 20–50 MHz down to 1–2 MHz. The object of the present study was at first the further improvement of the stabilization features. Our major interest was then focused on the question whether the optically stabilized TDL system can be used for a very-high-resolution saturation experiment. For this we chose a polarization spectroscopy scheme invented by Wieman and Hänsch in 1976 [15] which has been used in several applications in the past, but never in combination with a TDL as the saturating laser.

In the following, we report the latest results in line narrowing and frequency stabilization of our TDL system and demonstrate that this system provides a powerful instrument for very-high-resolution molecular spectroscopy.

## 1 Experiments and results

The experimental setup is schematically depicted in Fig. 1. It can be divided into two parts: the stabilized TDL system, described in Sect. 1.1, and the sub-Doppler spectroscopy arrangement (Sect. 1.2). The diode laser is line-narrowed by weak optical feedback and frequency-offset locked to a gas laser. A variable frequency offset between TDL and gas laser is defined by a sweepable microwave synthesizer. Since the wavelength region of our interest is around 5 $\mu$m we employed a CO laser as reference laser. For other wavelength regions one might alternatively use a CO overtone laser or a $CO_2$ laser. The sub-Doppler spectroscopy arrangement is based on a polarization-sensitive detection of the Lamb dip in a near-collinear beam scheme. A collinear beam geometry was preferred initially but gave rise to severe problems to be described below. In order to obtain very high sensitivity we employed a differential detection scheme. Frequency sweep and data acquisition are controlled by a computer.

### *1.1 Optically stabilized TDL system*

In this work we used a stripe-geometry, homostructure $PbS_{1-x}Se_x$ diode laser[1] which operates near 5 $\mu$m at temperatures between 20 and 40 K. The threshold current is about 180 mA. We usually operated the laser 40 to 120 mA above threshold which turned out to be a region with several single-mode operation regimes. The output power was measured by means of a power meter to be up to 2 mW single mode. The laser crystal is fixed in a special housing, developed at our laboratory, which permits bidirectional laser output beams. This rules out the possibility of uncontrolled reflections from the backside of the laser which may occur with the standard mounting (which permits an output beam in only one direction). Vibration-free cooling is realized by a temperature controlled helium-evaporation cryostat, which we modified for bidirectional beam output. The temperature stability of the laser station is estimated to be in the order of 1 mK during several minutes. Both laser beams are collimated by off-axis paraboloidal mirrors (focal length: 10.2 mm) which are placed inside the cryostat vacuum chamber. The diameter of the emerging TDL beams is about 8 mm. A small fraction of the laser light (less than $10^{-4}$) is retroreflected by an external feedback mirror and a beam splitter. Thus, the reflector and one facet of the laser crystal form an external cavity and a coupling between the diode cavity and the external cavity occurs. The distance between TDL and feedback mirror is about 40 cm; this corresponds to a free spectral range of 375 MHz. The details of our TDL cryostat as well as the appropriate optical feedback parameters have been described previously [16].

One of the beams can be directed to a 0.5 m Czerny-Turner type monochromator for a rough determination of the emission frequency. Lead-salt lasers mostly exhibit multimode operation due to their partly homogeneous and partly inhomogeneous broadened gain profile (spectral and spatial hole burning). Since the optical feedback method is not applicable if the diode laser is operating multi-mode (two or more modes with nearly the same output power), we have to find out the operating conditions where the laser runs nearly single mode[2] at the desired frequency. The proper current and temperature values were found with the help of a modechart [17].

One of the TDL beams is used to heterodyne the TDL against a gas laser. We used a flowing-gas CO laser cooled by liquid nitrogen which provides a few hundred laser lines in the wavelength region between 4.8 and 8.2 $\mu$m. The linewidth was measured earlier to be about 100 to 200 kHz. During the studies described in this paper the CO laser was line-center locked by means of a standard frequency-modulation technique. The TDL beam and the CO laser beam are focused separately, superposed by means of a $BaF_2$ beam splitter and directed to a fast Hg-doped Ge heterodyne detector/mixer[3]. This detector is cooled by liquid helium and enables us to observe difference-frequency signals (beat notes) up to a maximum frequency of 5 GHz under favorable conditions. In Fig. 2, spectrum analyzer records of the beat notes without and with optical feedback are given. The beat note obtained with the free-running TDL exhibits a spectral width of about 40 MHz (which corresponds to the TDL linewidth) whereas the beat note obtained with the stabilized TDL is dramatically narrowed to about 200 kHz. This shows that the TDL linewidth is about the same as the CO laser linewidth or even narrower. Thus we achieved a linewidth reduction by at least a factor of 200.

The resulting beat note is employed to control the length of the external resonator and consequently the TDL frequency by the feedback mirror. This was achieved by an electronic servo loop in a frequency-offset-locked (FOL)

[1] The TDL was manufactured by the Fraunhofer-Institut für Physikalische Messtechnik, Freiburg, Germany

[2] We define nearly single mode as the condition where at least 90 percent of all the power is in one mode

[3] The detector was manufactured by Santa Barbara Research Center, Goleta, California

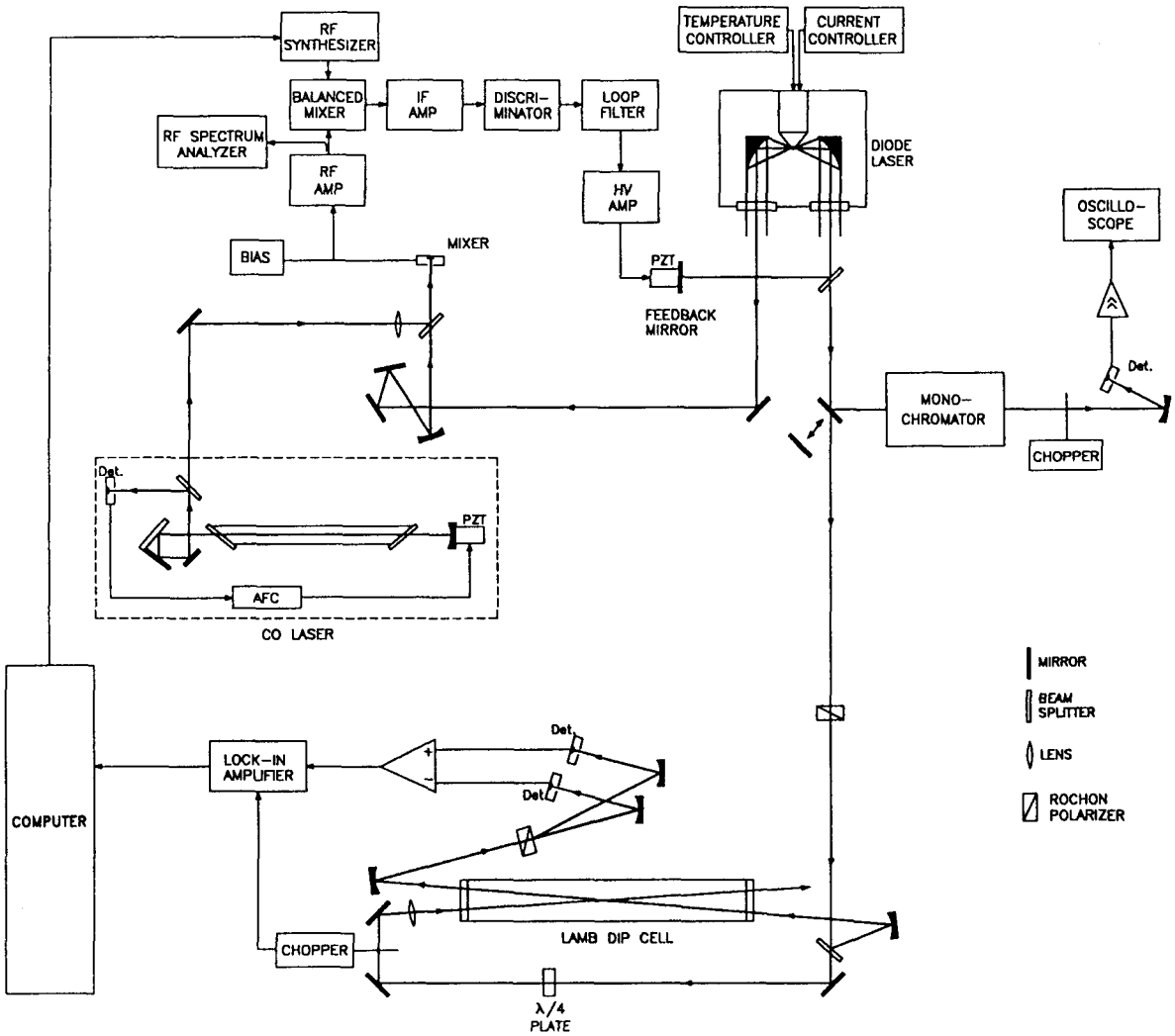


**Fig. 1.** Schematic diagram of the experimental setup. The TDL is optically line-narrowed and frequency-offset locked to the CO gas laser. One of the TDL beams is used for sub-Doppler spectroscopy of OCS in a polarization sensitive detection scheme

scheme. The beat note is down-converted (by mixing with a variable microwave frequency $\nu_{SYN}$) to an intermediate frequency of $\nu_{IF} = 160\,MHz$ and fed into a discriminator. This provides a frequency error signal which is applied to the PZT of the feedback mirror. When the servo loop is closed, the TDL frequency $\nu_{TDL}$ is locked to the CO laser frequency $\nu_{CO}$ with a variable offset[4], determined by the microwave synthesizer frequency $\nu_{SYN}$:

$$\nu_{TDL} = \nu_{CO} \pm \nu_{SYN} \pm \nu_{IF}\,. \tag{1}$$

This technique has already been utilized in [7] and some further details concerning the loop components can be found therein.

With this stabilization procedure we achieve suppression of the frequency jitter and drift of the external resonator to a large extent. The major portion of the frequency jitter is introduced by mechanical vibrations of the external resonator resulting from acoustical noise in the laboratory. Since the first report of our results, some progress on the acoustical isolation of the setup and the stability of the lock loop have substantially improved the stabilization performance. Figure 3 shows a spectrum-analyzer display, recorded with the maximum-hold option of the analyzer, over 500 consecutive scans during 1 minute. Nearly no additional broadening of the beat note becomes visible during this time which means that the frequency stability of the offset-locked diode laser is now mainly determined by the stability of the gas laser.

At the present stage of the experiment the long-term stability of the system suffers from imperfections of the evaporation cryostat. For unknown reasons the coolant flow becomes somewhat unsteady from time to time and this affects the temperature stability of the laser station. We are currently addressing this problem by constructing a novel helium-bath cryostat. In addition, a better temperature controller based on a digital data processing has been developed. The details of this system will be published elsewhere [18].

The use of a square-law photomixer allows the observation of difference-frequency signals ($\nu_{TDL} - \nu_{CO}$) between 0.1 and 5 GHz. Therefore, the TDL can be stabilized only in a restricted region of $\pm 5$ GHz next to a gas laser line. To overcome this problem, the use of a Metal-Insulator-Metal (MIM) point contact diode as mixer is required. The non-

[4] The choice of signs depends on whether $\nu_{TDL}$ is above or below $\nu_{CO}$

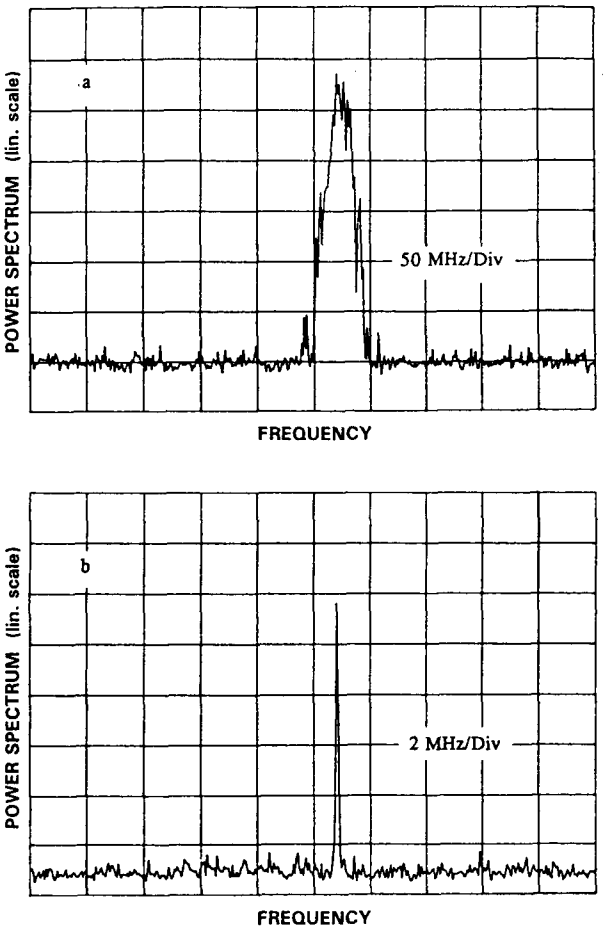


**Fig. 2a,b.** Spectrum analyzer display of the beat note between a CO laser and **a** the free-running TDL (single sweep, sweep time: 2 ms/div, resolution bandwidth: 100 kHz). The FWHM of the line is about 40 MHz. **b** The line-narrowed TDL (single sweep, sweep time: 2 ms/div, resolution bandwidth: 100 kHz). The FWHM of the line is about 200 kHz. Both records were obtained with a fast photodetector. The center frequency was near 1 GHz. It should be pointed out that the Signal-to-Noise Ratio (SNR) of the beat notes was usually observed to be more than 20 dB. This is not discernible here because we used a directional coupler in the RF signal path to monitor the beat note (Fig. 1)

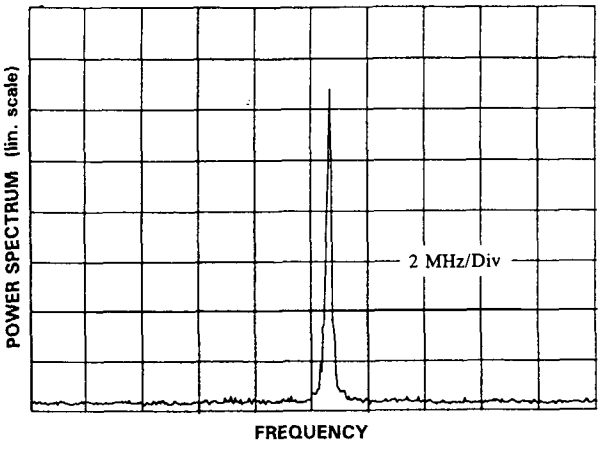


**Fig. 3.** Spectrum analyzer display of the beat note between the line-narrowed TDL and a CO laser, recorded during 1 minute by means of the maximum-hold option of the analyzer (sweep time: 2 ms/div, resolution bandwidth: 100 kHz). Compared to the single sweep of Fig. 2b, the spectral width of the beat note is nearly unchanged. This reflects the excellent stabilization performance of the offset-lock

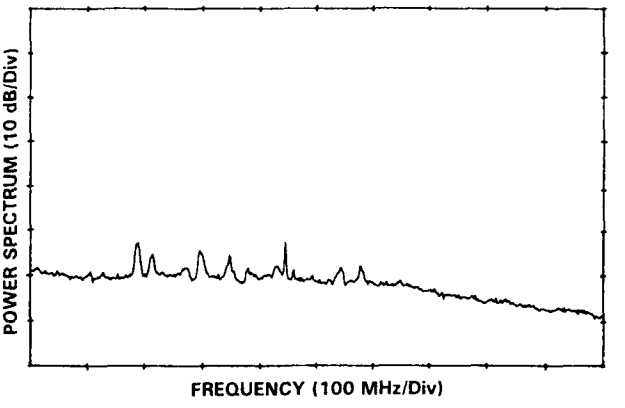


**Fig. 4.** Spectrum analyzer display of the beat note between the line-narrowed TDL and a CO laser, achieved with a MIM diode (single sweep, sweep time: 10 ms/div, resolution bandwidth: 1 MHz). The signal consists of many chaotic hopping lines; this points to uncontrolled optical feedback from the MIM diode to the TDL which de-stabilizes the diode laser

linearity of this device allows the generation of harmonics of the involved frequencies and thus enables higher mixing orders [19]. The main problem is that a MIM diode exhibits much higher noise than a LHe-cooled photodetector and yields a much lower SNR of the beat note. Up to now, no successful attempt in observing a beat note between a TDL and gas laser with a MIM diode has been reported because of the relatively low power and low spectral power density of the diode lasers. The considerably improved spectral power density of our line-narrowed diode laser encouraged us now to look for a beat note between the narrowed TDL and the CO laser by use of this device. We installed two flipping mirrors which allow us to switch both laser beams between the fast photodetector and the MIM diode. With the photodetector we achieved the narrow, well-shaped beat note as shown in Fig. 2b. After switching to the MIM diode we obtained a chaotic beat note consisting of fast jumping and fluctuating peaks (Fig. 4). This indicates a strong optical feedback from the MIM diode into the TDL which de-stabilizes the laser. Due to this difficulty we have not pursued this course and decided to defer further experiments with the MIM diode to future investigations.

Sweeps of the diode laser frequency can be performed in a well-defined linear manner by tuning the microwave synthesizer. The frequency sweep is controlled by a computer system. The procedure of a scan is as follows. First, the TDL temperature, the TDL current, and the feedback mirror position (feedback phase) have to be fine-tuned in such a way that the beat note frequency (monitored by the spectrum analyzer) is at the position of interest. Then the servo loop is closed. After defining the sweep range, the computer starts sweeping the synthesizer frequency and consequently the diode-laser frequency stepwise through the fixed frequency interval. The interval is normally divided into 400 steps. After reaching the stopping frequency the sweep direction is reversed.

Since the offset-lock loop controls only the *external* resonator, the laser frequency may be continuously swept at most by one free spectral range of the external resonator (375 MHz). After that a mode hop occurs to the next external mode. A longer continuous sweep range was obtained by a synchronous tuning of the diode-laser resonator; that is, the diode current was also scanned appropriately. In this way we were able to obtain a continuous tuning range of up to 800 MHz which was only limited by the displacement capability of the feedback-mirror PZT: we employed a piezo-ceramic stack with a maximum displacement of 6 $\mu$m. However, in the experiments described below, we were interested in about 50 MHz wide scans and thus there was no need for controlling the TDL current.

### *1.2 Sub-Doppler spectroscopy*

For the first test of the spectroscopic capabilities of our stabilized TDL system we chose carbonyl sulfide (OCS) as molecular absorber. OCS is a prominent calibration gas in the mid-infrared. It exhibits a multitude of strong absorption lines in the wavelength region of our interest (around 5 $\mu$m).

The sub-Doppler spectroscopy setup was realized in a nearly collinear beam arrangement (Fig. 1); that is, the pump beam and probe beam counterpropagate and cross each other under a small angle. The optimum beam geometry would be perfectly collinear in order to avoid residual Doppler broadening of the saturation dip but in this case isolation of the laser from the backcoming beam is essential. This is a severe problem since diode lasers are known to be extremly sensitive to small amounts of retro-reflected laser light. In a precursor experiment we tested a collinear beam arrangement utilizing a combination of a Rochon prism polarizer ($MgF_2$) and a $\lambda/4$ plate ($MgF_2$) as optical isolator. In this way we achieved an isolation ratio of about $10^4$. However, this turned out to be insufficient. With the optical isolator installed we still observed instabilities of the TDL arising from the backcoming beam.

Since the available diode-laser power to saturate the molecular transition is less than 1 mW, we have to employ a very sensitive detection method to observe a sub-Doppler feature. This is provided by the polarization spectroscopy method which is based on the observation of the nonlinear interaction of the pump and probe beam in an absorbing gas by changes in the polarization [15, 20]. We used a circularly polarized pump beam and a linearly polarized probe beam. The right-handed (or left-handed) pump beam introduces an anisotropy in the gas sample by saturating only molecular transitions with $\Delta m_J = +1$ (or $\Delta m_J = -1$); this results in different absorption coefficients and refractive indices for the two polarization components (induced dichroism and induced birefringence). If the weak linearly polarized probe beam interacts with molecules of the same velocity group, that is within the homogeneous linewidth of the absorbing molecule in the center of the Doppler profile, the polarization of the probe beam is slightly changed. This change is detected with high sensitivity.

The output of the diode laser is first directed to a $MgF_2$ Rochon prism polarizer. This improves the polarization purity of the TDL beam from less than 50:1 to more than 10000:1. The beam is then divided into two counterpropagating beams by a $CaF_2$ beam splitter. The strong pump beam is transferred into circular polarization by means of a $\lambda/4$ plate, modulated by a chopper (500 Hz) and focused to an intensity of about 100 mW/cm$^{-2}$ onto the center of the absorption cell (length: 1 m). The weak probe beam is focused onto the same location and crosses the pump beam with an angle[5] of about 30 mrad. In order to reduce noise components caused by fluctuations of the laser intensity a differential detection scheme [21–22] was used: a second Rochon prism whose axis is rotated by 45° with respect to the horizontally polarized laser beam splits the probe beam into two components of equal intensity and orthogonal polarization. Both beams are monitored by balanced $LN_2$-cooled InSb photodetectors. An induced rotation of the plane of polarization of the probe beam increases one of the detector signals while the other is decreased. A differential amplifier subtracts one signal from the other. Through the use of this detection scheme, intensity fluctuations which produce equal-sign detector signals are eliminated whereas opposite-sign signals due to the rotation of the axis of polarization are amplified. On resonance, the amplitude modulation of the pump beam is transferred to the probe beam. Phase-sensitive detection at the chopper frequency gives a dispersion-shaped profile of the Lamb dip. A detailed theoretical analysis of the signal shows that the 45° scheme detects only the induced birefringence (resulting from the saturated dispersion) whereas no contribution comes from the induced dichroism (resulting from the saturated absorption) [23]. This assumes that the polarizations of the pump and the probe beam are perfectly circular and linear, respectively. Imperfections of the polarization (for example due to anisotropy of the cell windows) lead to slight asymmetries in the signal shape.

Figure 5 shows a dispersion-shaped OCS Lamb dip, obtained with the above described arrangement. At each frequency step the phase-sensitive amplified signal is recorded by the computer. The scan was taken in a single sweep and shows an excellent S/N ratio of several thousand. The measurement was done on the P(52) transition of the asymmetric stretch ($10^00$–$00^00$ band) of the $O^{13}CS$ isotope (line strength: $0.3 \times 10^{-20}$ cm/molecule)[6]. The transition frequency of this line (1985.18218(15) cm$^{-1}$) is about 1574 MHz above the P(7), $v = 6 \rightarrow 5$ CO laser line (1985.12966(09) cm$^{-1}$). We used a commercially produced sample which contains about 1% $O^{13}CS$ in natural abundance. The absorber pressure was near 5 Pa. This corresponds to a pressure broadening of the molecular line of about 500 kHz, estimated from the broadening coefficients (92 to 105 kHz/Pa) given by Vanek et al. [11]. The observed signal width of 3 MHz is dominated by the residual Doppler width due to the finite crossing angle $\theta \simeq 30$ mrad between pump and probe beam. This geometrical broadening $\gamma_{geom}$ is given by [24]

$$\gamma_{geom} \simeq \frac{1}{2 \ln 2} \theta \gamma_D \qquad (2)$$

[5] Suitable optical components to achieve a smaller crossing angle (less than 10 mrad) were not available during these experiments

[6] For illustration of the line strength: the absorption of this OCS line is two orders of magnitude weaker than that of the strongest CO fundamental band line (under same conditions)

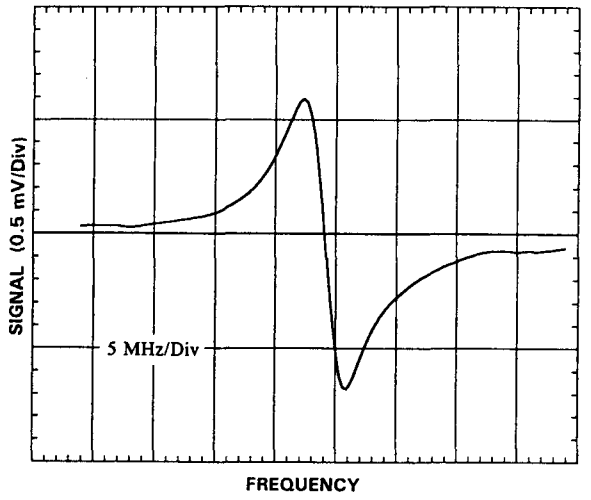


**Fig. 5.** Dispersion-shaped Lamb dip of the P(52) transition in the asymmetric stretch band of the $O^{13}CS$ isotope. The record was taken in a single sweep with a lock-in-amplifier time constant of 300 ms. The pressure in the absorber cell was near 5 Pa. The peak-to-peak linewidth is about 3 MHz, limited by the residual Doppler width due to the finite crossing angle between pump and probe beam

where $\gamma_D$ is the Doppler-width (= 94.5 MHz for $O^{13}CS$ at 300 K). This yields $\gamma_{geom} \simeq 2.2$ MHz which is the main contribution to the observed spectral width of the saturation signal.

## 2 Conclusion

The results presented in this paper demonstrate the use of tunable diode lasers for very-high-resolution spectroscopy. The most important feature of our stabilization scheme is the outstanding linewidth reduction (two orders of magnitude) and the consequent increase in the spectral power density. This enabled us to achieve a spectral resolution which has been accomplished before only with molecular gas lasers. We obtain tunable laser radiation of high spectral purity in a $\pm 5$ GHz region around any gas laser line in the mid-infrared (on condition that the gas laser power is above 50 mW). Possible gas lasers are, e.g., the $CO_2$ laser, $N_2O$ laser, CO laser, and CO overtone laser. For comparison, the side-band generation scheme provides tunable laser radiation in a 8 to 18 GHz window next to a gas laser line [3], thus both schemes complement each other in a useful way.

During this study we could not take full advantage of the resolution capability of the narrowed TDL due to the lack of appropriate optical components for the nearly collinear beam arrangement. A substantial improvement is achievable by reducing the crossing angle of the pump and the probe beam in order to reduce the residual Doppler broadening. A spectral Lamb dip width of less than 1 MHz should be attainable. The optimum would be a perfect collinear beam arrangement. For this an optical isolator with an isolation ratio of at least 60 dB would be essential. However, in practice, such a device is not available in the mid-infrared. Nevertheless, the Lamb dip of Fig. 5 is, to our knowledge, by far the narrowest spectroscopic feature observed with a lead-salt diode laser.

The frequency-offset locking of the line-narrowed diode laser allows us to sweep the TDL frequency in a well-defined and linear manner. By using computer-controlled sweeping and data-logging, we attain digitized records of molecular line shapes. Furthermore, a very accurate frequency calibration of the diode laser as well as a linear frequency scan are provided by this method. Since the frequency control is based on a heterodyne technique, coupling of the TDL frequency to the $CO_2$ frequency standard is feasible. For this, the CO laser beam is sent to the input of our $CO_2$ frequency synthesizer[7], recently described in [25]. We expect the achievable absolute TDL frequency accuracy to be in the order of $\Delta\nu = 100$ kHz ($\Delta\nu/\nu = 10^{-9}$). Compared to calibration procedures based on wavelength measurements, this frequency calibration is less prone to systematic frequency shifts. The absolute accuracy achieved with wavelength calibration procedures, e.g., used with FPI-stabilized diode laser systems, is typically one order of magnitude worse [26].

In summary, we have reported an experimental study on the application of an optically stabilized TDL system for sub-Doppler spectroscopy in the mid-infrared. The diode-laser linewidth was narrowed to less than 200 kHz. For the first time we have observed a Lamb dip in a carbonyl sulfide (OCS) absorption line near 5 $\mu$m using a polarization spectroscopy setup. These results represent a substantial improvement in the resolution capabilities of lead-salt diode lasers.

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