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Stabilization of 3.3 and 5.1 μm lead-salt diode lasers by optical feedback

M. Mürtz, M. Schaefer, M. Schneider [1], J.S. Wells [2], W. Urban
Institut für Angewandte Physik der Universität Bonn, Wegelerstr. 8, W-5300 Bonn, Germany

U. Schiessl and M. Tacke
Fraunhofer-Institut für Physikalische Meßtechnik, Heidenhofstr. 8, W-7800 Freiburg, Germany

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Optical stabilization of tunable lead-salt diode lasers (TDL) in the mid-infrared is presented for the first time. By introducing an external feedback mirror both the linewidth is substantially narrowed and the frequency of the TDL is stabilized and controlled via this mirror in a frequency-offset locked scheme. We achieved narrowing of the linewidth by 1–2 orders of magnitude or better. The TDL is offset-locked to a CO gas laser by a heterodyne technique: the beatnote between the two lasers is used to control the length of the external resonator. By this scheme we gain the capability of absolute frequency measurements with sub-Doppler accuracy. The improved spectral properties of the diode laser provide a new tool for high-resolution molecular spectroscopy in the mid-infrared.

Lead chalcogenide diode lasers are widely used as tunable light sources for IR-spectroscopy of molecules. However, the attainable spectroscopic resolution is limited by their large spectral width which is often in the order of 20 MHz. For sub-Doppler spectroscopy a linewidth reduction to at least 1 MHz or better is desirable. The object of our investigation was to find out whether the application of optical feedback can substantially improve the spectral properties of Pb-salt diode lasers; that is to prepare a tunable frequency-stabilized laser with a sub-megahertz linewidth for application in the sub-Doppler molecular spectroscopy.

In recent years investigations of different optical feedback schemes with III–V semiconductor lasers in the near-infrared (near 0.8 μm) have shown that under appropriate circumstances there is an improvement of the spectral properties [1–9]. For instance, by coupling a single-mode AlGaAs laser to an external high-finesse cavity, one can achieve a dramatic linewidth narrowing [10–12]. Different groups experimentally investigated and theoretically analyzed the proper feedback conditions (distance of reflector, reflectivity) for this diode laser type.

In comparison to this well-studied diode laser type, Pb-salt diode lasers, which are based on IV–VI compounds, are much less investigated. For this reason, their spectral behaviour is not well understood and their performance is still poor. The TDLs commonly used for IR-spectroscopy are gain-guided homostructure diodes which are operated at temperatures between 10 and 60 K. Their output power ranges from tens of microwatts up to several milliwatts and their spectral purity differs very much from laser to laser. These laser diodes mostly operate on several modes and exhibit an irregular far-field radiation pattern (spectral and spacial hole burning). These poor spectral properties and the cryogenic operating temperature considerably complicate the investigation of this diode laser type relative to the III–V semiconductor lasers.

Our first experiments were done with a $PbS_{1-x}Se_x$ homostructure diode at 5.1 μm wavelength; we op-

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[1] Now with Forschungsinstitut der Zementindustrie, D-4000 Düsseldorf, Germany.

[2] Permanent address: Time and Frequency Division, National Institute of Standards and Technology (NIST), 325 Broadway, Boulder, CO 80303, USA.

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erated then at temperatures between 20 and 40 K. The laser crystal was fixed on a special mounting. We have designed a novel laser housing which allows bidirectional laser output beams. Twice the power is then available, which turns out to be very convenient. This also 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). To ensure that we do not get any spurious optical feedback from external optical elements we use off-axis parabolic mirrors instead of lenses to collimate both laser output beams. The collimated beams emerge from the vacuum chamber through wedged and tilted CaF_2 windows. Cryogenic cooling is realized by a temperature-controlled evaporation cryostat which we operated either with liquid helium or with liquid nitrogen. In contrast to closed-cycle coolers, this cryostat cools without transmitting vibrations to the lasers which is essential for optical feedback experiments. A block diagram of the experimental arrangement is shown in fig. 1, and a comprehensive description of the experimental details is given in ref. [13].

A reliable and unambiguous way to measure the spectral linewidth of a laser is to heterodyne it against a reference laser. At 5.1 μm we used a sealed-off CO laser as local oscillator. This laser provides a few hundred laser lines in the wavelength region from 5 to 8 μm. The linewidth is in the order of 1 MHz. The mixer was a fast liquid-nitrogen-cooled photovoltaic HgCdTe crystal, which allowed us to observe beatnotes up to 5 GHz under favorable conditions. The output of the mixer was processed by a microwave amplifier (30 dB gain) and fed to a spectrum analyzer. Figure 2a shows a typical beatnote observed on the instrument. Since the linewidth of the used CO laser is about 1 MHz, the fwhm of the beatnote, which is about 70 MHz, reflects the linewidth of the diode laser. This agrees with earlier experiments in our lab where we found linewidths between 20 MHz and 100 MHz for different homostructure diode lasers. The irregular lineshape shows the insufficient spectral purity of this diode laser. Slightly different operating temperatures or currents can cause the lineshape to vary considerably. In the case of strong multimode operation we observed also further broadening of the line.

In contrast to these measurements the theoretical value for the linewidth, given by the Schawlow-Townes formula, modified by Henry [14], is about 0.1 to 5 MHz (depending on the α-value, output power, etc.). The causes for this huge discrepancy are not well understood. The spectral width normally reported for lead-salt lasers is 5 to 100 MHz. One group observed linewidths less than 100 kHz for carefully selected high power diodes [15]. This dramatic variation in the linewidths of diode lasers of the same type may be due to an additional noise generating mechanism inherent to the lead-salt diode lasers. We assume that mode competition noise and carrier density fluctuations are responsible for the linewidth in these gain-guided homostructure laser diodes [#1]. Laser action in different lasing filaments with slightly different frequencies might also contribute to the linewidth. We took care to avoid any possible external feedback to the diode, nonetheless one cannot exclude the possibility of slight optical feedback, for example, coming from the (tilted) photomixer, which might affect the lineshape.

This large spectral width prohibits use of TDLs for sub-Doppler spectroscopy up to now. With a view to narrow the TDL linewidth we applied controlled optical feedback on one side of the diode: a portion of one of the two laser beams was retro-reflected in the laser while doing the heterodyne process with the second, independent beam. In this way we could simultaneously observe the lineshape while varying the feedback conditions. We used different kinds of windows and mirrors as external reflectors at different distances from the diode laser. The effective feedback level is determined by the reflector material and a factor which describes the portion of the backcoming light that really enters the laser crystal. Since this factor depends on the geometrical position of the parabolic mirror, the incoupling properties of the laser crystal, etc. the effective feedback level is not directly accessible.

Depending on the distance of the reflector d and the reflectivity R_{ext} we observed different effects on the lineshape. The shortest distance between reflec-

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[#1] Linewidth measurements of high-performance AlGaAs laser diodes with better electron and photon confinement are in good agreement with the predictions calculated from Henry's linewidth formula [14].

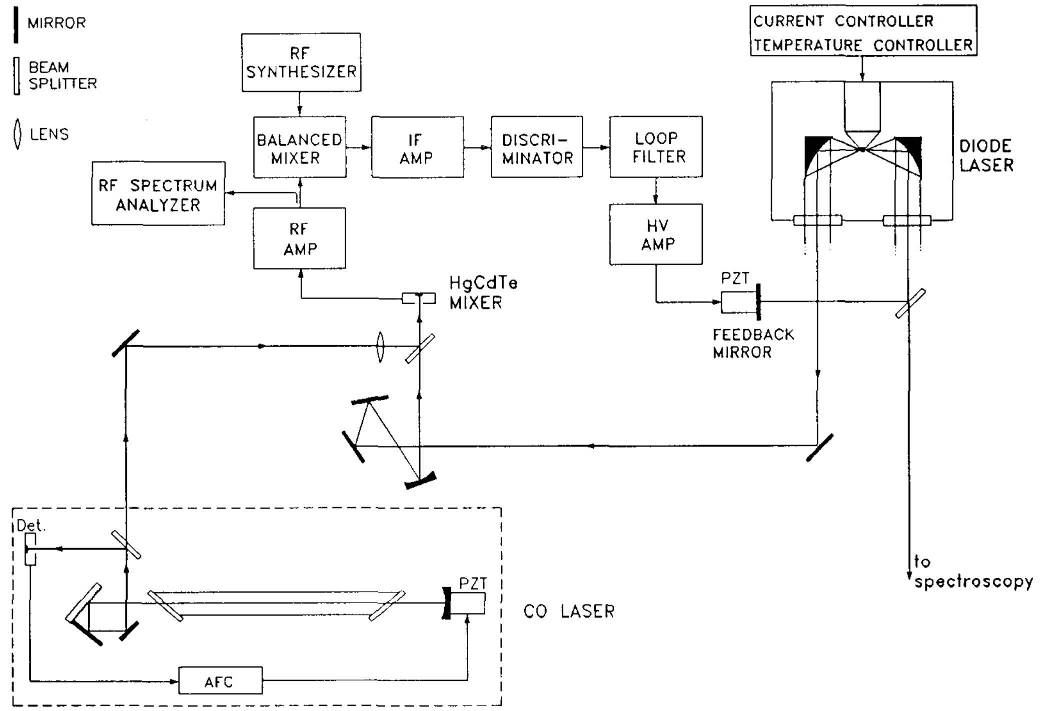

Fig. 1. Experimental diagram of the line-narrowed and offset-locked diode laser system. One of the TDL beams is used for heterodyning it with radiation from a CO laser. A fast HgCdTe photomixer is employed to achieve beatnotes up to 5 GHz. The second TDL beam is used to apply weak optical feedback from an external mirror. The length of this external resonator is controlled by the frequency of the beatnote via an electronic servo loop.

tor and diode was limited to 60 cm through the size of the vacuum chamber. At this distance we found various kinds of instabilities for strong feedback ($R_{ext} \gg 10^{-3}$) as well as line narrowing for weak feedback ($R_{ext} < 10^{-3}$). Strong feedback induced line broadening, and in the worst case, mode jumps which meant loss of the beatnote. Weak feedback, coming from a mirror via a CaF_2 beam splitter (fig. 1), results in a dramatic linewidth reduction (by two orders of magnitude) down to 1 MHz or less together with a considerable increase in the spectral power density. Figure 2b shows the narrowed beatnote. It looks much more regular and symmetric than that of the free-running diode. The peak width of 1 MHz is probably limited by the linewidth of the CO laser; that is we could not determine the exact spectral width of the TDL in this experiment. For this purpose a substantially improved reference laser will be required.

With a slightly increased feedback level we simultaneously observed one to three or even more additional narrow peaks with a spacing of about 250 MHz. These can be interpreted as modes of the external cavity formed by the reflector and one facet of the laser crystal. The free spectral range of the 60 cm long cavity corresponds to the 250 MHz spacing. A specific amount of feedback is necessary to achieve operation in exactly one external cavity mode with optimum linewidth reduction. We found this amount experimentally by adjusting the parabolic mirror which focused the reflected beam onto the laser crystal. When placing the external reflector at larger distances than 60 cm the external mode spacing decreases; furthermore the feedback level must be decreased in order to avoid rising of instabilities. In this case we also observed line narrowing but the attainable linewidth reduction becomes worse. Therefore it is desirable to go to smaller distances than 50 cm; some changes at the cryostat will render this in the near future.

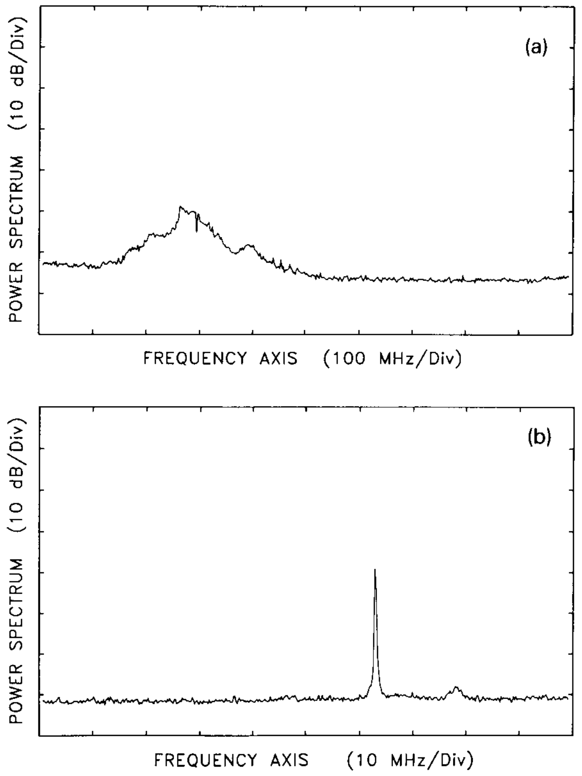

Fig. 2. Spectrum analyzer display of the beatnote between a CO laser and a homo-structure TDL at 5.1 μm. The center frequency is near 1 GHz. (a) Beatnote of the free-running diode laser (single sweep). The fwhm of the line is about 70 MHz. The irregular lineshape reflects the chaotic phase fluctuations of the TDL radiation field. It depends strongly on the operating conditions (temperature and current) of the diode laser. A bandwidth of 3 MHz and a sweep time of 10 ms/division were used. (b) Beatnote of the same diode after line narrowing by optical feedback (single sweep). The fwhm is about 1 MHz. The peak width is probably limited by the linewidth of the CO laser. Note that the dispersion is 10 MHz/division here in contrast to 100 MHz/division above. The bandwidth is 300 kHz and the sweep time is again 10 ms/division.

A consequence of the coupling between the diode cavity and the external resonator is the altered tuning behaviour of the laser. Sweeps of the laser frequency can be performed in two different ways. Either one tunes the diode laser cavity by changing the current or one tunes the external cavity by changing its length. The continuous current tuning range is up to 50 MHz. When the current is tuned further, a mode hop occurs: the laser emission jumps to the next external mode (1 FSR to higher frequency with increasing current). Following the theory of a compound cavity, the current-tuning rate of the extended cavity diode laser is reduced by a factor of

$$1+X\sqrt{1+\alpha^2}$$

compared to that of the solitary laser [5]. The feedback strength parameter, X, is defined to be $\tau_{\mathrm{ext}}R_{\mathrm{ext}}^{1/2}(1-R_{\mathrm{L}})/(\tau_{\mathrm{L}}R_{\mathrm{L}}^{1/2})$ where τ_{L}, R_{L}, τ_{ext} and R_{ext} are the roundtrip time and power reflectivity of the laser resonator, and of the external resonator. The quantity α is the linewidth broadening factor. This factor is not exactly known for our diode laser type. From calculations and estimates by other authors for similar diodes [15,16] we assume an α-value of 1 to 2. In the case of optimum linewidth reduction at a reflector distance of 60 cm we found tuning rates of 700 MHz/mA for the solitary laser and 160 MHz/mA for the laser with optical feedback. From these measurements the effective feedback level can be roughly estimated. We deduce that the feedback parameter X is about 1.5 to 2.5. This corresponds to an effective external reflectivity of about 10^{-4}.

On the other hand, we can tune the laser frequency by changing the length of the external resonator. The feedback mirror can be moved a few micrometers with a piezo-ceramic transducer (PZT). This results in a continuous tuning range of about 200 MHz, followed by a mode hop (to lower frequencies if one tunes the cavity to higher frequencies and vice versa). To achieve longer continuous tuning ranges one can either decrease the length of the external resonator in order to increase the FSR or one can tune the external cavity with the PZT and change the current synchronously. In this way one obtains continuous tuning ranges up to a few gigahertz.

Although the spectral behaviour of free-running lead-salt diode lasers are rather complicated and not well understood, the behaviour of the laser with feedback described above is in good agreement with the predictions by a rate equation model for semiconductor lasers with external optical feedback [4,5,8,12]. According to the theory we observed stable operation accompanied with linewidth reduction for weak optical feedback ($R\approx 1$) and the onset of instabilities when exceeding a critical value for the feedback strength ($R\gg 1$).

However, a unique coupling to the external resonator is not possible if the laser is working multimode, that is, if there are two or more modes with nearly the same output power. The proper current and temperature values required to run the laser nearly single mode [2] at the desired frequency were found with the help of a modechart [17].

In a different experiment we briefly tested the behaviour of a burried heterostructure MBE diode laser with optical feedback from the same arrangement as described above. This TDL which lased near 3.3 μm wavelength was commercially available [3] in a standard housing which permits beam output in only one direction. As a local oscillator we used a liquid-nitrogen-cooled, flowing gas CO laser, oscillating on $\Delta v = 2$ transitions. This laser provides more than 250 laser lines in the wavelength region from 2.7 to 4 μm [18]. The linewidth of the free-running TDL was about 20 MHz (fig. 3a). Since only one diode laser beam was accessible we had to use the transmitted part of the split beam for the heterodyne process. The first observations pointed at an insufficient effective external reflectivity. For this reason we slightly increased the feedback by exchanging the CaF_2 beam splitter for a ZnSe beam splitter. With this setup we again observed line narrowing down to a few megahertz (fig. 3b). Because collimation of the emerging beam and refocussing of the returning beam was achieved by the same parabolic mirror, we could not obtain the appropriate amount of feedback needed for optimum linewidth reduction as described for the 5 μm experiments. However, this preliminary result also demonstrates the capability of the optical feedback technique for laser diodes mounted in the standard housing. In order to improve the system performance one has to use an external mirror with variable reflectivity or an equivalent scheme.

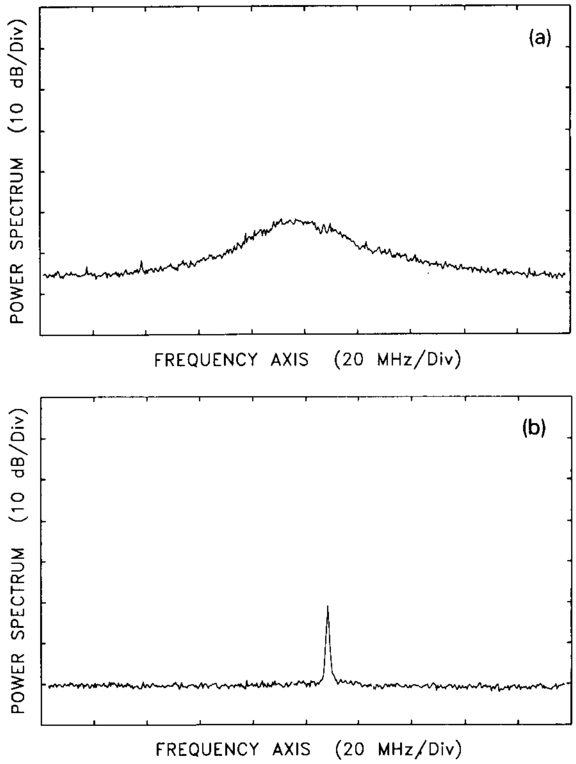

Fig. 3. Beatnotes between a CO overtone laser and a burried heterostructure MBE diode laser near 3.3 μm. The dispersion is 20 MHz/division, the bandwidth is 300 kHz and the sweep time is 10 ms/division (single sweep) for both (a) and (b). The center frequency is again near 1 GHz. (a) The diode is free running, the linewidth is about 20 MHz (fwhm). (b) Beatnote of the same diode after application of weak optical feedback.

The application of optical feedback from an external mirror does not provide stabilization of the center frequency of the narrowed line. Due to the acoustical noise in the laboratory, vibrations of the feedback mirror and consequently a jitter of the center frequency of the narrowed line are introduced. The insufficient acoustical isolation of the external resonator leads to a jitter of ±15 MHz which becomes visible during consecutive scans of the spectrum analyzer. Moreover temperature variations of the external apparatus cause slow drifts of the frequency. With a view to the planned precision spectroscopy, stabilization and control of the center frequency of the narrowed line is essential. This is accomplished by controlling the length of the external resonator. We installed a servo loop which offset-locks the TDL to a reference laser with a variable frequency offset defined by a frequency synthesizer (fig. 1). This frequency-offset holding technique was used in a different approach by Freed et al. [19].

For this experiment we again used the 5 μm TDL. The beatnote is down-converted by means of a double-balanced mixer and an rf synthesizer as local os-

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[2] That is, the power level of any other mode was less than 10% of the dominant mode power level.

[3] The TDL was manufactured by Laser Photonics, Analytics Div. Bedford, MA in the USA.

cillator. The intermediate frequency (IF) equals 160 MHz. The IF signal is amplified by 70 dB and fed to a discriminator which gives an error signal with a sensitivity of 100 mV/MHz. The loop filter contains an amplifier with good low-pass characteristics and an integrator. The signal is finally amplified to a high voltage and fed to the PZT of the feedback mirror. When the servo loop is closed, the frequency jitter is reduced to 1 MHz or less. The TDL frequency is locked to the reference CO laser frequency with a variable offset of $\nu_{\mathrm{syn}} \pm 160$ MHz[#4] (provided that there is no additional offset in the loop). This means we can scan the stabilized diode laser frequency within 0.3–5 GHz above and below a CO laser line. This frequency band is limited through the bandwidth of the photomixer. The CO laser was line-center locked by means of a standard frequency modulation technique. Optimization of the locking scheme is still in progress, and some improvements in the stability of the lock appear possible.

In summary, our experiments show that it is possible to achieve considerable linewidth reduction for a lead-salt diode laser by applying controlled optical feedback. After optimizing the feedback features we expect linewidths on the order of 100 kHz. We combined this method with frequency stabilization and control by offset-holding the TDL to a CO laser. This tunable narrowband light source in the mid-infrared provides a new tool for very high resolution spectroscopy with sub-Doppler accuracy. Since the frequency control is based on a heterodyne technique, coupling of the TDL frequency to a secondary frequency standard is easily feasible. By introducing one or two Lamb dip stabilized CO_2 lasers, we gain the capability to make very accurate frequency measurements [20]. We will extend our investigations to longer wavelengths (10 μm) and also to different diode laser structures (DH-, BH-laser, etc.) at 5 μm. Further experiments with other optical feedback elements like a grating or an external high-finesse cavity are in preparation.

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[#4] The sign depends on whether we choose ν_{syn} either above or below the beatnote frequency.

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