

DIODE LASERS AND METROLOGY

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Introduction

At NIST in Boulder we have been pursuing an active research program developing diode-laser technology for scientific applications. Commercial diode lasers are readily available in a few wavelength bands in the red and near IR region of the spectrum. Our work has focused on the AlGaAs, InGaAlP, and InGaAsP lasers that operate at room temperature in the red and near IR region of the spectrum between 650 nm and 1.5 microns. These lasers have a number of recognized advantages, including: high efficiency, low cost, tunability, and moderate power levels (~1 to 100 mW). Increasing interest in applying diode lasers to science in general and spectroscopy in particular has stimulated a number of recent reviews on the subject.¹⁻⁴

Unfortunately the distribution of wavelengths of commercial diode lasers only incompletely covers the potential spectral region. The available lasers are grouped into bands that meet specific commercial applications (such as laser printers, communication systems, CD players etc). In addition to the wavelength limitation the tuning range of any specific laser with temperature and/or injection current is discontinuous. Attempts to tune a diode laser to a specific wavelength are often frustrated by the tendency of these lasers to avoid the desired wavelength by jumping modes. The tuning characteristic of any given mode for a specific laser is somewhat reproducible but shows hysteresis and is easily perturbed by optical feedback. Usually once the laser reaches the desired wavelength it is very stable as long as the temperature, injection current and optical feedback are stable. However on a longer time scale there is some evidence of wavelength change as the laser ages.

Wavelength Tuning and Anti-Reflection Coatings

Wavelength selective optical feedback may be used to almost eliminate diode laser tuning problems. The most popular technique is to use an optical grating to feedback to the output of the laser. This is an extended cavity configuration and will work to some degree with almost any laser, but it works best if the laser is single-mode (both spatial and longitudinal) and with a reduced reflectivity coating on the facet towards the grating⁴⁻⁶. Fortunately many of the higher power commercial diode lasers have a high reflectivity coating on the back facet and reduced reflectivity coating on the output facet. Obviously, feedback from the grating enhances the gain of this coupled cavity system at the feedback wavelength. The gain at other wavelengths is suppressed by the antireflection coating on the output facet. The lower the reflectivity the farther we can hope to tune the extended

780 nm Laser AR Coated with $\lambda/2$ Al_2O_3 and $\lambda/4$ HfO_2

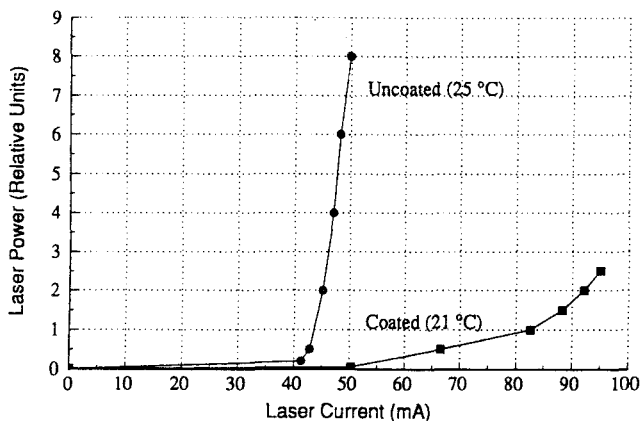


Fig. 1 Laser output power versus injection current for an AlGaAs diode laser as purchased, and with an anti-reflection coating on one facet of the laser.

cavity laser away from the gain peak of the solitary laser. We do not know that the optimum reflectivity is necessarily zero. Other factors such as linewidth, single mode operation or dynamical instabilities may turn out to be worse if the reflectivity is very low. However our experience suggests that the lower the reflectivity the better the extended cavity laser works. Reflectivities that we currently work with range from about 30 to 0.1 percent.

With these systems we can tune to the desired wavelength by tuning the grating and changing laser chip's temperature when necessary. Synchronously changing the laser cavity length while turning the grating will result in larger continuous tuning sweeps. However for finite facet reflectivities, the continuous tuning range near any given solitary chip mode depends on the type of semiconductor laser, the temperature, the grating dispersion, feedback power, injection current relative to threshold, tuning relative to the semiconductor gain curve, and the level of other parasitic optical feedback. For a typical AlGaAs system

Table I. Diode laser antireflection coating results. I_{th}/I_{th0} is the ratio of the laser's threshold current after coating to threshold before coating. The last column gives the calculated after-coating output facet reflectivity as a percentage.

Lasers and Coating Materials				
Laser Type ¹⁰	Half-Wave Correction	Coating Material	I_{th}/I_{th0}	% Front Facet Reflectivity
LTO26	YES	Y ₂ O ₃	1.4	6.9
LTO26	YES	HfO ₂	1.9	1.9
LTO26	YES	SiO	2.1	0.6
LTO26	NO	HfO ₂	1.5	5.4
HLP1400	NO	Ta ₂ O ₅	1.8	1.9
HLP1400	NO	Al ₂ O ₃	1.4	9.2
ML2701	NO	SiO	1.6	3.8
ML4405	YES	HfO ₂	1.4	1.0
TOLD9215	NO	HfO ₂	1.7	2.0
TOLD9215	YES	HfO ₂	2.1	0.6
TOLD9220	NO	HfO ₂	1.7	2.0
TOLD9220	YES	HfO ₂	2.0	0.1

we might have a continuous tuning range of ~50 GHz while the spacing between the solitary chip modes is about 150 GHz. The intermediate frequencies between the modes are accessible by changing the temperature of the laser chip. There is a great deal of published literature^{7,8} on coating diode lasers and some of the results demonstrate⁹ that if the output facet reflectivity is low enough the grating can control the laser wavelength without any mode jumps associated with the solitary chip modes.

In order to improve the operation of our extended cavity diode lasers we have been putting anti-reflection coatings on commercial diode lasers. Usually we do not have access to information on the actual semiconductor materials or geometry of these devices which makes it very difficult to design the appropriate coating. With each new laser type we are forced to make an educated guess and then empirically determine which coatings work best. While coating the laser we usually monitor the output power (from the back and/or front facet) as a function of the injection current and coating thickness. A typical power versus injection current plot for a laser both before and after coating is shown in figure 1.

An additional complication in coating diode lasers is that we are usually dealing with commercial lasers that already have some unknown optical/passivation coating on their facets. This coating makes it more difficult to achieve very low reflectance. Table I is a

compilation of some of our anti-reflection coating results on commercial diode lasers. Our present best results are reflectance of $\sim 0.1\%$, and an increase in laser threshold of ~ 2.1 times.

Ultra-sensitive Detection

Now switching our discussion to applications, we can consider the prospects for using diode lasers for ultra-sensitive detection or other analytic applications.³ The first thing we note is that the amplitude noise on diode lasers is very small which means we should be able to do direct absorption detection of absorbing species with very high sensitivity. Our first effort in this direction was to look at using diode lasers for laser enhanced ionization (LEI) in flames.¹¹ This work was done as a joint effort between NIST Boulder and NIST Gaithersburg. The very simple experimental system consisted of an

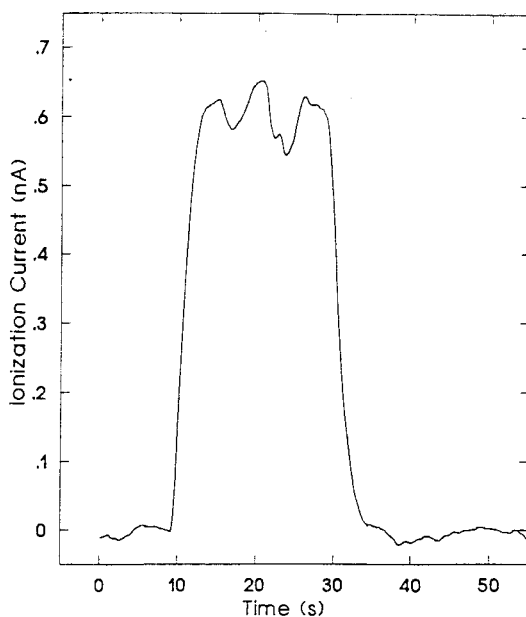


Fig. 2 10 ppb rubidium in water detected using laser-enhanced-ionization in a flame with a diode laser.

atmospheric pressure air-hydrogen (or acetylene flame) and water samples that contained the atoms of interest at low concentrations. We first measured cesium and rubidium detection limits because we could reach their resonance lines with convenient diode lasers. The water samples were aspirated into the flame where the diode laser light excited the atoms. The atoms were subsequently ionized by the flame and the electric field from a high voltage (~ -500 V) electrode. The laser beam was chopped and the current was detected synchronously with a lock-in amplifier. Experimental data for rubidium is shown in fig. 2 where the LEI signal is shown for a 10 ppb (parts/billion) concentration of rubidium in water. With just 2 mW of laser power this data gives a detection limit of 300 ppt (parts/trillion) atom concentration of rubidium in water.

These are just preliminary experimental results for a very simple, practical system that has not been optimized for ultimate detection sensitivity. This first demonstration does show that the technology is workable but it is limited by the present availability of diode laser wavelengths in the near-IR spectral region. However with blue light from frequency doubled diode lasers we could reach transitions in many more atomic species. This should greatly enhance the usefulness of diode laser LEI and other ultra-sensitive detection techniques.

Diode Laser fluctuations

One of the usual limitations to using lasers for high sensitivity detection of atomic species is the amplitude fluctuation of the detected laser light. This noise comes from fluctuations in the laser's output power, direction, and frequency. Fortunately the intrinsic fluctuation in the laser's output power is very low (roughly 10^{-6} for frequencies above 1 MHz). These fluctuations can be reduced by the use of electronic feedback.¹² Within the electronic servo bandwidth, the amplitude noise can be reduced to ~ 3 dB above the shot noise level with a 50% expenditure of power for the feedback loop. However even with smaller fractions of the laser power used for the servo, significant reduction of the amplitude noise is possible.

But unfortunately with diode lasers we have the additional problem that when the frequency of the laser is scanned there is usually a relatively large systematic change in the output power. This is in addition to the usual etalon and other multi-path effects that often interfere with low-level absorption signals. The affect that this residual amplitude modulation has upon detection limits can be greatly diminished by suitable demodulation techniques.

Fast detectors - frequency difference measurements

Using optical locking¹³⁻¹⁵ and/or electronic feedback techniques it is possible achieve very narrow linewidths with diode lasers. To effectively use the resolution that is available from narrow linewidth lasers it is necessary to be able to first measure, and second control the laser's center frequency with a precision that is comparable with the linewidth. For example, an optically-locked diode laser with a linewidth of ~ 3 kHz has a potential resolution of ~ 1 part in 10^{11} . It takes a very high accuracy frequency reference to control the center frequency with that level of precision. This can be achieved with the best quality optically-contacted Fabry-Perot reference cavities or with standards-quality atomic/molecular resonances. Even with one of these good references to lock to we often want to measure or even scan the laser's frequency relative to the reference. This can be done with RF frequency-offset-locking techniques as pioneered by Hall and collaborators.¹⁶

We have been exploring the use of high speed Schottky diodes with diode lasers for measuring large optical frequency intervals and for controlling the laser frequency with high precision. The use of very small area Schottky diodes to detect very high frequency laser beat-notes in the visible has been demonstrated by Daniel et al.^{17,18} In our present near-infrared experiments we optically-lock two 830 nm diode lasers to the same confocal reference cavity. The beat note between these two lasers is then detected with the fast Schottky diode. By increasing the difference frequency between the two lasers we can measure the useful signal-to-noise ratio and bandwidth of the Schottky diodes. In our initial experiments we measure a signal-to-noise ratio of ~ 60 dB with a resolution bandwidth of 10 kHz for beat-notes in the "base-band" region from DC to about 25 GHz. The detector's DC optical responsivity at 830 nm is about 0.2 mA/mW. Based on the Schottky's capacitance and series resistance the detectors intrinsic millimeter-wave bandwidth should be about 2 THz. In order to look for the very high speed response we

used the Schottky as a harmonic mixer to generate the harmonics of a 47 GHz klystron. The signal-to-noise of the laser beat note signal as detected relative to harmonics of the klystron is plotted in fig 3. This data is plotted as a function of the difference frequency between the two lasers and the data points correspond to the various klystron harmonics.

The signal-to-noise ratio at low frequencies in this data is presently limited by a frequency noise pedestal that remains on the two optically locked diode lasers. We believe that the apparent roll-off towards higher frequencies is a combination of the Schottky diode's harmonic generating efficiency and some optical roll-off, but this has not been confirmed yet. With our preliminary results we know we can see beat notes between diode lasers with good signal-to-noise out to at least 400 GHz and with appropriate local oscillators we are hopeful that this can go much higher.

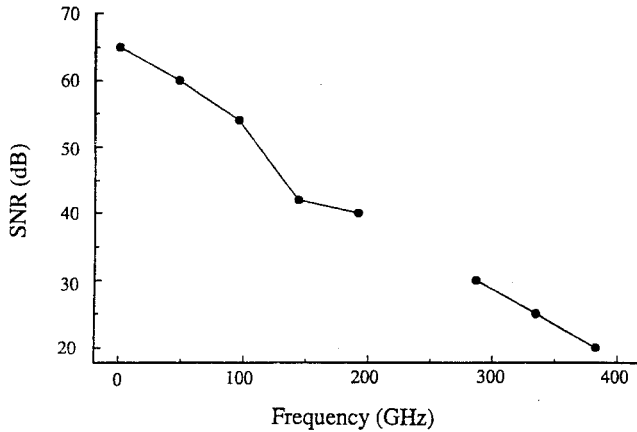


Fig. 3 Signal-to-noise ratio (SNR) of the beat-note between two optically-locked diode lasers and the klystron harmonics, as a function of the laser-laser difference frequency. The klystron was operated at about 47 GHz. The data point corresponding to the 5th klystron harmonic is not shown.

Visible Wavelength / Frequency References

The relatively new red diode lasers (with wavelength bands between 630 and 690 nm) used in conjunction with narrow transitions in the alkalis and alkaline-earths atoms provide us with the opportunity for significantly improve visible wavelength/frequency references. Very nice results have already been achieved with Ba (791 nm)¹⁹, and Sr (689 nm)²⁰, while we have been pursuing Ca (657 nm)¹². Calcium is attractive because of its very narrow (400 Hz natural width) transition, because it can be readily laser cooled, and because it is well established as a reference wavelength.²¹⁻²⁴ There is a growing need for an improved visible wavelength reference as evidenced by recent precision measurements of atomic hydrogen transitions.²⁵ Because of the relative simplicity of the diode lasers there is a realistic hope for a portable length/frequency transfer standard based on calcium with

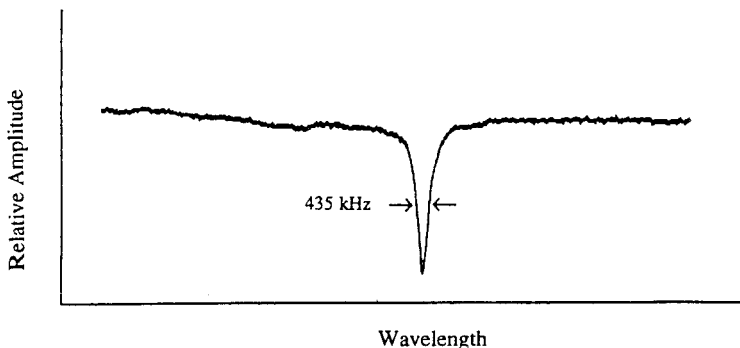


Fig. 4. Calcium Saturated absorption spectrum of the 657 nm line taken with a diode laser. The laser was locked to a cavity and the cavity scanned over the line. A precise frequency scale for the horizontal axis was provided by modulation sidebands of the laser that are not shown in this scan.

very high accuracy. Figure 4 shows a recent saturated absorption signal that we have obtained from a hot calcium cell and 500 μW of diode laser power. Similar calcium linewidths have been obtained with this same diode laser and the PTB calcium beam (in collaboration with V. Velichansky, F. Riehle, and J. Helmcke). The narrowest saturated absorption line-widths that we have observed with our diode laser and a hot calcium cell are less than 150 kHz. These widths are presently limited by transit broadening and wave-front curvature effects due to the inhomogeneities in the laser's spatial mode.

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References

1. J. Camparo, The diode laser in atomic physics, *Contemp. Phys.* **26**, 443, (1985).
2. M. Ohtsu and T. Tako, Coherence in semiconductor lasers, in: "Progress in Optics XXV," E. Wolf ed., Elsevier, p. 193, (1988).
3. J. Lawrenz, K. Niemax, A semiconductor diode laser spectrometer for laser spectrochemistry, *Spectrochimica Acta.* **44B**, 155, (1989).
4. C. Wieman and L. Hollberg, Using diode lasers for atomic physics, *Rev. Sci. Inst.* **62**, 1, (1991).
5. E.M. Belenov, V.L. Velichansky, A.S. Zibrov, V.V. Nikitin, V.A. Sautenkov, V.A. Uskov, Methods for narrowing the emission line of an injection laser, *Sov. J. Quant. Elect.* **13**, 792, (1983).
6. A. Akul'shin, V. Bazhenov, V. Velichansky, M. Zverkov, A. Zibrov, V. Nikitin, O. Okhotnikov, V. Sautenkov, N. Senkov, E. Yurkin, *Sov. J. Quant. Elect.* **16**, 912, (1986).
7. I.P. Kaminow, G. Eisenstein, and L.W. Stulz, Measurement of the Modal Reflectivity of an Antireflection Coating on a Superluminescent Diode, *IEEE J. Quant. Elect.* **19**, No. 4, 493, (1983).
8. H. Ukita, K. Mise, and Y. Katagiri, Simple Measurement of the Reflectivity of Antireflection-Coated Laser Diode Facets, *Jap. J. Appl. Phys.* **27**, L1128, (1988).

9. P. Zorabedian, W.R. Trutna Jr., and L.S. Cutler, Bistability in Grating-Tuned External Cavity Semiconductor Lasers, *IEEE J. Quant. Elect.* 23, 1855 (1987).
10. Mention of specific commercial laser products does not constitute an endorsement but is made to clarify the particulars of our experiments. Other devices may be better suited to this type of application.
11. G.C. Turk, J.C. Travis, J.R. DeVoe and T.C. O'Haver, Laser Enhanced Ionization Spectrometry in Analytical Flames, *Anal. Chem.* 51, No. 12, 1890 (1979).
12. L. Hollberg, R. Fox, N. Mackie, A.S. Zibrov, V.L. Velichansky, R. Ellingsen, and H.G. Robinson, Diode Lasers and Spectroscopic Applications, in: "Tenth International conference on Laser Spectroscopy," M. Ducloy, E. Giacobino and G. Camy, p. 347, World Scientific, (1992).
13. B. Dahmani, L. Hollberg, R. Drullinger, Frequency stabilization of semiconductor lasers by resonant optical feedback, *Opt. Lett.* 12, 876, (1987).
14. Ph. Laurent, A. Clairon and Ch. Breant, Frequency Noise Analysis of Optically Self-Locked Diode Lasers, *IEEE J. Quant. Elect.* 25, No. 6, p. 1131, (1989).
15. H. Li and H.R. Telle, Efficient Frequency Noise Reduction of GaAlAs Semiconductor Lasers by Optical Feedback from an External High-Finesse Resonator, *IEEE J. Quant. Elect.* 25, No. 3, 257, (1989).
16. J. Hough, D. Hils, M. D. Rayman, Ma L.-S., L. Hollberg, and J. L. Hall, Dye-Laser Frequency Stabilization Using Optical Resonators, *Appl. Phys. B* 33, 179, (1984).
17. H.-U. Daniel, B. Maurer and M. Steiner, A broadband Schottky Point contact mixer for visible laser light and microwave harmonics, *Appl. Phys. B* 30, 189, (1983).
18. J.C. Bergquist, H.-U. Daniel, A wideband frequency-offset-locked dye laser spectrometer using a Schottky barrier mixer, *Optics Comm.* 48, 327, 1984.
19. A.M. Akulshin, A.A. Celikov and V.L. Velichansky, Nonlinear Doppler-free spectroscopy of the 6^1S_0 - 6^3P_1 intercombination transition in barium, *Optics Comm.* 93, 54 (1992).
20. G.M. Tino, M. Barsanti, M. de Angelis, L. Gianfrani and M. Inguscio, Spectroscopy of the 689nm intercombination line of strontium using and extended-cavity InGaP/InGaAlP diode laser, *Appl. Phys. B* 55, 397, (1992).
21. J.C. Bergquist, R.L. Barger and D.J. Glaze, in: "Laser Spectroscopy IV," H. Walther and K.W. Rothe eds., Springer-Verlag, p. 120, (1979).
22. J. Helmcke, A. Morinaga, J. Ishikawa and F. Riehle, Optical Frequency Standards, *IEEE Trans. Inst. Meas.* 38, 524, (1989).
23. N. Beverini, F. Giammanco, E. Maccioni, F. Strumia, and G. Vissani, Measurement of the calcium 1P_1 - 1D_2 transition rate in a laser-cooled atomic beam, *J. Opt. Soc. Am. B* 6, No. 11, p. 2188, (1989).
24. T. Kurosu, F. Shimizu, Laser Cooling and Trapping of Calcium and Strontium, *Jap. J. Appl. Phys.* 29, L2127, (1990).
25. T. Andrae, W. König, R. Wynands, D. Leibfried, F. Schmidt-Kaler, C. Zimmermann, D. Meschede, and T.W. Hansch, Absolute Frequency Measurement of the Hydrogen 1S-2S Transition and a New Value for the Rydberg Constant, *Phys. Rev. Lett.* 69, 1923, (1992).