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GaN blue diode lasers: a spectroscopist's view

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Abstract. We have characterized the spectroscopic properties of one of the first samples of blue-emitting diode lasers based on GaN. With such a laser diode operated inside a standard extended cavity arrangement we find a mode-hop free tuning range of more than 20 GHz and a linewidth of 10 MHz. Doppler-free spectroscopy on an indium atomic beam reveals the isotope shift between the two major indium isotopes as well as efficient optical pumping.

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Laser diodes have caused a revolution in communications and in optical data storage. Particularly in the latter field a light-weight, robust, and energy-saving laser source is desirable in order to facilitate mobile applications. Initially, in the consumer market (audio-CD, CD-ROM) 780-nm wavelength diode lasers were employed. With the introduction of DVD drives a much higher data storage capacity and therefore longer playing times were achieved through several improvements in the overall system, among them the switch to visible red-emitting diode lasers with their smaller diffraction-limited spot size. Over the last decade tremendous efforts have been made towards the development of blue-emitting diode lasers because these promise even higher storage densities. Laser powers of a few milliwatts in cw operation are required with a laser lifetime of at least 10 000 hours. Last year this goal was reached by the Japanese company Nichia Chemical Industries, and for the past few months the first trial samples have been available commercially.

The diode lasers developed for telecommunications and the consumer market have also had a large impact in laser spectroscopy, because the same advantages that make them interesting for commercial applications are also very welcome in spectroscopic experiments. However, not all desirable wavelengths have been available. The deep blue and near-UV range, a particularly interesting region of the spectrum because of the electronic absorption frequencies of many atoms and molecules, has been accessible only by indirect methods. For instance, frequency doubling of IR lasers, while routinely used, nonetheless requires sophisticated setups if stable output power and frequency are desired. Even then it is very hard to obtain a cw output power of more than several hundred microwatts in the blue, although many milliwatts have been demonstrated in fully optimized systems.

The availability of blue-emitting diode lasers offers a chance to extend diode laser spectroscopy also into the blue region of the spectrum. As a spectroscopist one can hope that the GaN diode lasers will turn out to be as useful as the IR diode lasers have been to near-IR spectroscopy. The purpose of this work is to examine the spectroscopic characteristics of this new type of laser diode. Quantities of interest to the spectroscopist are the mode structure, both spectrally and spatially, the tuning behavior, and the linewidth. We will address these issues in turn, using a 410-nm transition in an indium atomic beam as an example.

1 GaN diode lasers

It is beyond the scope of this paper to describe details of the manufacturing process because the average spectroscopist will certainly buy the diodes ready-made. Many details have been given in a particular publication [1], and an overview for the non-specialist has been published in the scientific literature [2]. The manufacturer will not start to sell its samples directly before the end of this year. We have therefore obtained our blue diode laser through an OEM manufacturer who incorporated the diode into his standard system [3], consisting of a collimation lens and an external diffraction grating in Littrow configuration providing feedback into the laser diode. This is the standard setup routinely used in many laboratories all over the world and has been described by several authors who adapted the technique, developed for dye lasers, independently (see, for example, [4, 5]).

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2 Spatial mode structure

The most visible characteristic of the blue diode laser beam, apart from its color, is its spatial mode profile. In our prototype this is rather disappointing because it does not look like a TEM_{00} at all, but more like a TEM_{40} mode. In order to clean the mode we used an optical fiber as a spatial filter. Out of the 3.4-mW blue power at the fiber input a total of about 300 μW was available in a clean Gaussian TEM_{00} mode. However, due to strong optical feedback from the input facet of the fiber, the laser frequency was extremely unstable. Therefore, the fiber coupler was intentionally misaligned in order to avoid strong feedback, which reduced the available TEM_{00} power to 250 μW.

According to TuiOptics [3], the complicated mode structure is not intrinsic to the laser diode itself but is due to aberrations in the collimating lens. In our system this was still an aspheric lens designed for a 780-nm wavelength because diode laser collimators for the blue spectral region were not yet available. However, blue-optimized collimators are expected to be on the market soon, and it is hoped that this problem will vanish.

3 Tuning characteristics

For all spectroscopic applications a wide mode hop-free tunability of the laser output frequency is desirable: the wider, the better. A diode laser with an external grating can be tuned by several means: diode temperature, diode injection current, grating distance, and grating angle. The diode temperature is a useful parameter only for the initial tuning of the diode to the vicinity of the spectral lines of interest because it can be adjusted only slowly.

Rapid scanning across the spectral features is performed by tilting the grating with the help of a piezo element. In the TuiOptics setup this changes both the distance between grating and laser diode (i.e., the length of the external cavity) and the grating angle. Because these two changes are not matched to each other there will eventually be mode hops, in the present case typically after about 6 GHz, the free spectral range of the external cavity.

This range can be expanded by employing a feed-forward loop: an attenuated version of the grating piezo voltage is added to the DC injection current of the diode. Fine adjustment of the attenuation factor and the DC operating point enable the mode hop-free scan range to expand to more than 20 GHz. Over this wide scan range there is a strong laser output power decrease when scanning towards shorter wavelengths. This power variation is a well-known characteristic of non-antireflection coated extended-cavity diode lasers and various ways to reduce it using more sophisticated setups have been published (see, for example, [6–8])

As an example we consider the fluorescence spectrum of indium atoms in an atomic beam. The $6\,^2S_{1/2}$ excited state of indium can be reached from the $5\,^2P_{1/2}$ ground state by absorption of 410 nm laser light (Fig. 1a). The excited state can decay into either the $5\,^2P_{3/2}$ or back into the $5\,^2P_{1/2}$ state, while emitting 451 nm or 410 nm light in the process. Both naturally occurring isotopes (^{115}In: 95.7% relative abundance, ^{113}In: 4.3%) have a nuclear spin quantum number of 9/2, almost identical nuclear magnetic moment, and a hyperfine splitting of 11.4 GHz in the ground state and 8.4 GHz in the excited state. All excitation lines are therefore expected to be asymmetric doublets: a strong ^{115}In peak accompanied by a weak ^{113}In peak, separated by the isotope shift of 258.2 MHz for this transition.

In our experimental setup the In beam emerges from an effusive cell heated to 1265 °C and is collimated by slits to a 1:2150 ratio. It is illuminated by the collimated blue laser beam at 90°. A photomultiplier tube detects the fluorescence light emitted into the third perpendicular direction. Figure 1b shows the spectrum of the total fluorescence intensity, normalized to the laser intensity which changes from 20 mW/cm^2 at 0 GHz relative detuning to 6 mW/cm^2 for 20 GHz detuning due to the feed-forward loop. All four hyperfine components for transitions from $F = 4, 5$ to $F' = 4, 5$ are resolved within a single mode hop-free scan.

The fluorescence intensity plotted in Fig. 1b consists of two components, one at 410 nm and another one at 451 nm wavelength (see Fig. 1a). The former can be prevented from reaching the photodetector by a narrow-band interference filter with 50% maximum transmission centered at 450 nm. The resulting spectrum (Fig. 1c) is much less noisy because the residual stray light (mostly due to laser light scattered by the windows of the vacuum vessel) is blocked as well.

The relative areas under the Lorentzian peaks in Fig. 1c are found as 100:71:39:117 (from left to right), in good agreement with the ratios calculated from the angular momenta (100:67:37:100). The fact that the $F = 4 \mapsto F' = 5$ transition is stronger than expected is probably not due to hyperfine optical pumping because the branching ratio from the excited state back to the $5\,^2P_{1/2}$ states is only 0.36 so that most atoms absorb just one 410-nm photon before being pumped into the non-interacting states $5\,^2P_{3/2}$ or the other hyperfine component of the $5\,^2P_{1/2}$ state. Instead, it is more likely a combination of the effects of changing laser power and therefore laser linewidth (via the Schawlow–Townes formula [9]) during the laser scan: at 20 GHz detuning the laser is operating barely above threshold while at 0 GHz it is close to its maximum allowed output power.

At higher resolution one can distinguish the contributions by the two isotopes ^{113}In and ^{115}In in the fluorescence spectrum: each fluorescence peak in Figs. 1b,c is actually a doublet (Fig. 1d). The known isotope shift was used to define the scale of the frequency axis. The relative height of the two peaks in Fig. 1d corresponds to the natural abundance ratio of the two isotopes.

4 Laser linewidth

A feature of special interest is the spectral width of the laser emission. From a spectroscopist's point of view the so-called spectroscopic linewidth is the relevant quantity, i.e., the minimum width of spectral features that can be observed in an experimental spectrum caused by the width of the laser spectrum itself. This quantity is some mixture of the intrinsic quantum noise limited laser linewidth and the contributions made by technical noise and drifts.

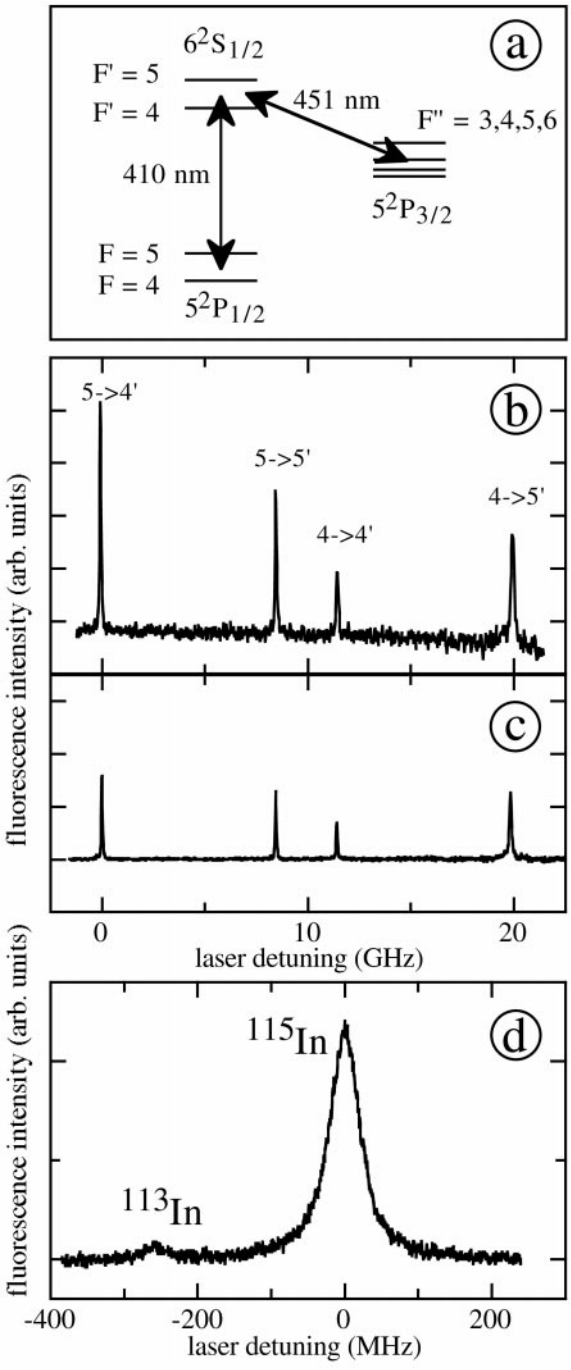

Fig. 1. a Excerpt from the indium level scheme. **b** Hyperfine resolved total fluorescence spectrum of the $5\,^2P_{1/2} \mapsto 6\,^2S_{1/2}$ transition in an indium atomic beam. **c** Spectrum taken with a bandpass filter for 450 nm [same scale as in **b**]. **d** The lowest-frequency peak measured with better resolution: the contribution of the ^{113}In isotope becomes noticeable

4.1 Spectroscopic linewidth

The width of the fluorescence peaks in Figs. 1b,c is mostly limited by power broadening, as can be seen from Fig. 2 where the full width at half maximum (FWHM) of the $F = 5 \mapsto F' = 4$ component (far left peak in the spectrum) is plotted as a function of laser intensity. Even for the smallest intensities the minimum width of about 35 MHz is still larger than the natural width $\gamma/2\pi = 25.1$ MHz. It was checked experimentally that this is not due to a residual Doppler shift caused by non-perpendicular laser and atomic beams or by beam divergence. The remaining explanation is a rather large spectroscopic laser linewidth. If we assume a Lorentzian shape for both the laser line and the "intrinsic" atomic line the measured line shape is their convolution, i.e., another Lorentzian with a width equal to the sum of the two original Lorentzians. One can therefore estimate a spectroscopic linewidth $\Delta\nu_s \approx$ 10 MHz which is about a factor of 5–10 larger than for a typical near-IR external cavity laser.

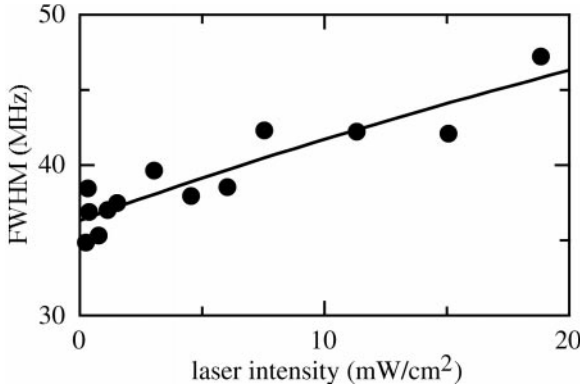

Fig. 2. Width of a Doppler-free absorption line as a function of laser power. The natural width of the line is 25.1 MHz. The *solid line* is a numerical fit of a saturation broadening curve

While the data in Fig. 2 is certainly consistent with a linear dependence, the solid line shows the result of a fit of the standard model function for power broadening, $\mathrm{FWHM}(I_{\mathrm{laser}}) = \Delta\nu_s + (\gamma/2\pi)\sqrt{1 + I_{\mathrm{laser}}/I_{\mathrm{sat}}}$, where I_{sat} is the effective saturation intensity of the transition. From the fit one obtains $\Delta\nu_s = (11.2 \pm 0.6)$ MHz and $I_{\mathrm{sat}} = (21.0 \pm 3.2)$ mW/cm^2. This value is not the same as the two-level saturation intensity $I_s = \pi hc\gamma/3\lambda^3 \approx 75.7$ mW/cm^2 because of optical pumping to the $5\,^2P_{3/2}$ state.

4.2 Beat signal

In order to obtain an independent estimate of the diode laser linewidth one can look at the width of the beat signal between two identical systems. Unfortunately we have only one such diode laser system available. Instead, we have used an 820-nm extended cavity diode laser and frequency-doubled it in an external ring resonator using a $LiIO_3$ crystal. The linewidth of the IR laser itself is about 1 MHz. In order to stabilize the 410-nm output power from the ring resonator the polarization of the fundamental light reflected at the input was used as an error signal [10] for an electronic servo loop that kept the ring resonant with the laser light at all times. Still, the blue output power fluctuated by about 25% around its mean value of about 90 μW.

The frequency-doubled beam and the beam from the blue diode laser (after spatial filtering by the fiber) were superposed on a fast photodiode and the resulting beat signal was amplified and displayed on a radio-frequency spectrum analyzer. Figure 3 shows a typical beat note for a sweep time of 60 ms. Its width of about 8.3 MHz, taken from a numerical fit of a Lorentzian, is a combination of the "fast", quantum limited linewidths of the laser diodes and their

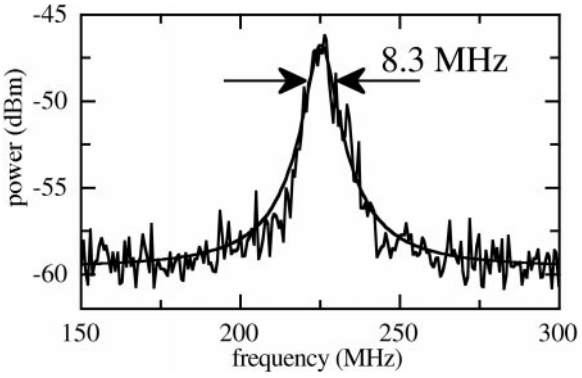

Fig. 3. Typical beat note between the frequency doubled 820-nm extended cavity diode laser and the 410-nm extended cavity GaN diode laser (resolution bandwidth 300 kHz, sweep time 60 ms). The *smooth line* is a Lorentzian fit to the beat note

"slow" linewidths, corresponding to jumps in laser center frequency due to acoustic and other perturbations. Because of the 100 kHz bandwidth of the current driver, a small electronic noise contribution to the fast linewidth cannot be excluded. A linewidth of the GaN laser diode of around 10 MHz, obtained by the two independent methods, is certainly good enough for all but the most demanding spectroscopic applications.

Nevertheless it is tempting to speculate about the origin of the large laser linewidth. For a free-running laser diode the quantum-limited linewidth is given by the modified Schawlow–Townes formula [9]

$$\Delta\nu_{\mathrm{MST}} = \frac{\pi h\nu \Delta f_{\mathrm{res}}^2}{P} \frac{N_{\mathrm{C}}}{N_{\mathrm{C}} - N_{\mathrm{V}}}(1+\alpha^2) \qquad (1)$$

where P is the laser output power, $h\nu$ its photon energy, Δf_{res} the linewidth of the resonator formed by the two diode facets, N_{C} and N_{V} the carrier concentrations in conduction and valence band, and α is the linewidth enhancement factor [11]. With facet reflectivities of 95% and 14% and a chip length of 650 μm [12] one obtains $\Delta f_{\mathrm{res}} = 80$ GHz which is comparable to the situation for typical near-IR diodes. On the other hand, $h\nu$ is about twice as large as in the near-IR. With $N_{\mathrm{C}} \gg N_{\mathrm{V}}$ for simplicity and at $P = 5$ mW, this results in $\Delta\nu_{\mathrm{MST}} = 1.9\,\mathrm{MHz} \times (1+\alpha^2)$, giving an estimate of $\alpha = 2.0$ if the influence of the feedback from the grating is ignored for now. This is well within the range of traditional quantum well diode lasers [13]. Clearly, a full characterization of the noise properties of a GaN laser diode will be needed for the determination of a reliable value for α.

5 Conclusion

It has been shown that the behavior of the new blue-emitting GaN laser diodes is very similar to that of the better-known red and IR-emitting diodes. The non-antireflection coated diodes allow a tuning range of more than 20 GHz without mode hops with a linewidth of about 10 MHz.

It would be interesting to study the modulation characteristics of GaN diode lasers when operated in an extended-cavity configuration. From the recently published data sheet of the manufacturer [14] one can infer a relaxation oscillation frequency of 1.5 GHz for the solitary laser diode. This is therefore the practical upper limit for fast diode current modulation. Instead of soldering a bias tee network directly to the blue laser diode in order to check the characteristics of the combined system we have used an open SMA cable as an antenna to transmit a radio frequency modulation directly onto the pins of the plastic socket holding the laser diode. With this crude setup modulation sidebands at 600 MHz and more could be seen. This indicates that an important tool in sensitive spectroscopy, high-frequency modulation spectroscopy [15], would also be possible with GaN extended-cavity diode lasers.

Our own interest in the blue laser diode lies in its application to atom lithography with indium. Here a laser-collimated atomic beam is deposited onto a substrate after focussing by a pattern of standing light waves. Suitable variation of this pattern during indium deposition should allow the growth of III-V semiconductors from the GaAlInP family with a three-dimensional modulation of chemical composition and therefore of refractive index on a nanometer scale. This method could provide a new way to produce photonic bandgap structures in the optical domain. However, efficient atomic beam collimation by laser cooling requires many absorption-emission cycles which cannot be reached by interaction on the 410-nm transition alone because of the optical pumping discussed above. Instead, we plan to use a combination of 410-nm and 451-nm light to pump all atoms into the $5\,^2P_{3/2}$, $F = 6$ state and from there use the cycling transition at 326 nm to the $5\,^2D_{5/2}$, $F = 7$ state for laser collimation. The availability of an easy-to-use blue light source brightens the prospects for the success of this longer-term project.

Another interesting application might be the selective removal of one of the In isotopes from the atomic beam by resonant laser light pressure. This would increase the contrast of the atomic patterns written by the atom lithography process because it removes the background provided by the isotope which is not interacting with the lithography lasers. On a broader scale, it would allow the growth of isotopically pure compound semiconductors even when using an indium oven loaded with the natural isotope mixture.

Currently the price of a single GaN laser diode of $2000 is still rather high; however, a quantity price of $8 for diodes sold to optical disk drive manufacturers was estimated some time ago [16]. If one considers the rapid progress of GaN laser diodes (commercialization within two years of their first laboratory demonstration) as compared to the two decades or so it took for the near-IR diodes, one can certainly expect a rosy – or more appropriately, blue – future for diode laser spectroscopy around 400 nm wavelength.

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References

1. S. Nakamura, G. Fasol: *The Blue Laser Diode* (Springer-Verlag Heidelberg 1997)
2. S. Nakamura: Science **281**, 956 (1998)
3. TuiOptics GmbH, 82152 Martinsried, Germany
4. C.E. Wieman, L. Hollberg: Rev. Sci. Instrum. **62**, 1 (1991)
5. K.B. MacAdam, A. Steinbach, C. Wieman: Am. J. Phys. **60**, 1098 (1992)

6. F. Favre, D. Le Guen, J. C. Simon, B. Landousies: Electron. Lett. **20**, 795 (1986)
7. M. de Labachelerie, G. Passedat: Appl. Opt. **32**, 269 (1993)
8. D. Wandt, M. Laschek, A. Tünnermann, H. Welling: Opt. Lett. **22**, 390 (1997)
9. A. Yariv: *Quantum Electronics* (Wiley, New York 1987) Chapt. 21
10. T.W. Hänsch, B. Couillaud: Opt. Commun. **35**, 441 (1980)
11. C.H. Henry: IEEE J. Quantum Electron. **18**, 259 (1982)
12. S. Nakamura, Nichia Chemical Industries: private communication
13. M. Osinski, J. Buus: IEEE J. Quantum Electron. **QE-23**, 9 (1987)
14. Nichia Chemical Industries: Violet Laser Diode NLHV500A, data sheet (September 1999)
15. W. Lenth: IEEE J. Quantum Electron. **QE-20**, 1045 (1984)
16. C.T. Whipple: Photonics Spectra, May 1998, p. 118