Probing the ultrafast gain and refractive index dynamics of a VECSEL

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Typically, strong gain saturation and gain dynamics play a crucial role in semiconductor laser mode-locking. While there have been several investigations of the ultrafast gain dynamics in vertical-external-cavity surface-emitting lasers (VECSELs), little is known about the associated refractive index changes. Yet, such refractive index changes do not only have a profound impact on the pulse formation process leading to self-phase modulation, which needs to be compensated by dispersion, but they are also of particular relevance for assessing the feasibility of Kerr-lens mode-locking of VECSELs. Here, we measure both refractive index as well as gain dynamics of a VECSEL chip using the ultrafast beam deflection method. We find that, in contrast to the gain dynamics, the refractive index dynamics is dominated by an instantaneous (∼100 fs) and a very slow component (∼100 ps). The time-resolved measurement of nonlinear refraction allows us to predict a pulse-length dependent, effective nonlinear refractive index $n_{2,\text{eff}}$, which is shown to be negative and in the order of $10^{-15} m^2/W$ for short pulse lengths (∼100 fs). It becomes positive for large excitation fluences and long pulse lengths (few ps). These results agree with some previous reports of self-mode-locked VECSELs for which the cavity design and pulse properties determine sign and strength of the nonlinear refractive index when assuming Kerr-lens mode-locking.

Ultrashort-pulse mode-locking and frequency combs have emerged as a major topic in semiconductor laser research, and state-of-the-art semiconductor disk laser systems (or VECSELs) are under development that utilize passive mode-locking with various applications in mind such as spectroscopy, biomedicine, nonlinear optics and many more. Mostly optically-pumped, VECSELs combine flexible wavelength design of the gain chip, high optical output powers and excellent beam quality. The external cavity allows to further functionlize the laser emission, for example to achieve single-frequency generation or nonlinear frequency conversion by inserting optical filters or nonlinear crystals into the cavity, respectively.

Mode-locking can routinely be achieved by inserting semiconductor-saturable absorber mirrors (SESAMs) into the cavity with the possibility of obtaining ultrashort pulses in the sub-100 fs regime as well as peak powers of several kilowatts. In this context, recently also saturable-absorber-free mode-locking, usually referred to as "self-mode-locking", has received considerable attention and has been demonstrated by several groups. These results remain subject of ongoing debate within the community concerning what the driving mechanisms behind this phenomenon are, and whether those truly lead to a mode-locked state of the laser. Both, mode-locking by a four-wave-mixing nonlinearity in the gain chip and Kerr-lens mode-locking have been discussed as possible explanations. In particular, the latter hypothesis triggered considerable efforts to characterize the nonlinear refractive index of the gain chip under realistic conditions, i.e. using probe irradiances and excitation fluences comparable to how they occur in a mode-locked VECSEL. However, most of these investigations have been performed using pulse lengths of only a few hundreds of femtoseconds. Yet, pulses generated by self-mode-locked VECSELs usually are longer, that is in the few-ps regime down to sub-ps pulse durations. Therefore, considering the strong gain dynamics of semiconductor lasers, which intrinsically affects the refractive index of the gain chip, such nonlinear lensing investigations so far have not provided a very accurate picture of the nonlinear refractive index of the gain chip. Time-resolved measurements of the refractive index dynamics of a VECSEL would therefore allow us to obtain a more realistic estimate of the strength of nonlinear lensing in VECSELs. Beyond this, it would allow us to obtain insight into the phase dynamics that affects a short pulse in VECSEL mode-locking. In phenomenological modeling of pulse formation, this is usually taken into account by introducing a constant, the linewidth enhancement factor, which relates gain changes to phase changes. However, this is a strongly simplifying assumption and therefore the full knowledge of the refractive index dynamics might improve modeling significantly.

In this work, we measure the time-resolved nonlinear optical response of a gain chip using the recently developed ultrafast beam-deflection technique. This uniquely enables us to simultaneously acquire gain as well as refractive index dynamics. Moreover, we validate the results by additional Z-scan measurements well known from effective nonlinear lensing characterization of materials without temporal resolution. Our pump-probe setup, including a prepulse for excitation

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of the sample, is displayed in Fig. 1(a). Here, the probe spot is overlapped with the pump spot on the sample in a way that it sits exactly on the position of the largest slope of the Gaussian-shaped intensity profile of the pump as shown in Fig. 1(b) (which requires that the probe spot diameter is considerably smaller than the pump at the position of the sample). Consequently, the refractive index changes induced by the pump in the sample will lead to a deflection of the probe beam that will be detected by a non-zero difference signal between the upper and lower half of the segmented detector.

In the following, we investigate beam deflection measurements for different excitation fluences ranging from 1.1 µJ to 21.2 µJ and a fixed pump peak irradiance (1 GW/cm²). Figure 2(a) and (b) show the normalized pump-induced transmission changes \( \Delta T / T \) and the deflection changes \( \Delta \epsilon / E \) as a function of the delay between pump and probe pulse, respectively. Transmission changes \( \Delta T \) are normalized with respect to the total signal of the probe in the absence of the pump, \( T \), and deflection changes \( \Delta \epsilon \) are normalized with respect to \( E = T + \Delta T \), which also takes into account the transmission changes of the probe during the deflection measurement.

When the sample is only weakly excited, the probe pulse experiences first an increase of transmission before decaying back to a transmission change of close to zero. The deflection signal experiences a very fast negative response and subsequently a rather constant negative deflection signal over the few-ps time scale. Negative deflection corresponds to a negative refractive index change.

When the sample is strongly excited, the probe experiences first a strong decrease in transmission and subsequently a recovery to a slightly negative transmission change. This offset from zero transmission change at longer times indicates that the pump beam experiences net gain. Note that the
and one obtains a time constant of 300-400 fs for carrier cooling, i.e. when the sample is in the absorption regime and only weakly excited. This corresponds to values measured in Ref. 33. When the sample is strongly excited, this time constant increases slightly to 400-500 fs, corresponding to the time the carrier-distribution heats up to the lattice temperature. In comparison to the measurement of Ref. 33, the ultrafast gain recovery also contains a pronounced instantaneous component (∼100 fs) for large excitation fluences.

Interestingly, the effect of carrier cooling/heating, i.e. the component with a sub-ps time constant, is not very strong in the measurement of the deflection signal (Fig. 2b), as that signal mostly consists of an ultrafast (∼100 fs) and a very slow component (∼100 ps). In contrast to measurements of semiconductor optical amplifiers, the instantaneous negative decrease of the refractive index reduces significantly with increased excitation fluence, while otherwise the trend is similar.33,36

The time-resolved deflection measurement can be mapped to the refractive index change averaged over the length of the probe pulse, <Δn(t)>, where τ represents the delay between pump and probe pulse. This is done by comparing the deflection signal of the VECSEL to the deflection signal of a reference sample (SiC) with a known nonlinear refractive index n2 (see the Supplementary Material for more details of this procedure). The right axis of Fig. 2b displays the corresponding <Δn(t)> for the measurement of the VECSEL sample.

The response function, obtained by fitting <n(t)> to Eq. 2 can be used to calculate the effective nonlinear refractive index n2,eff. This quantity describes the nonlinear refractive index that would be seen by a single beam propagating through the sample, for example when performing a Z-scan measurement, and depends on the pulse length. It relates to the total refractive index change by Δn = n2,effI, with I being the irradiance of the single beam. It can be calculated by

\[ n_{2,\text{eff}} = \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R(t-t')I(t')dt'dt}{\int_{-\infty}^{\infty} I^2(t)dt}. \]

Here, I(t) is the intensity pulse envelope of the single beam. Fig. 3a shows the excitation-dependent n2,eff calculated from various deflection measurements with different pump irradiances for a pulse length of 1 ps. One can see that n2,eff is similar for all pump irradiances, being around -6·10^{-16} m^2/W for no excitation and increasing to slightly positive values of around +2·10^{-16} m^2/W for large excitation irradiances.
In addition to the beam deflection measurements, Z-scan measurements were performed on the unexcited sample, which yield an $n_{2,\text{eff}}$ of $-1.2 \times 10^{-16}$ m$^2$/W. This corresponds in sign and order of magnitude to the $n_{2,\text{eff}}$ of around $-3 \times 10^{-16}$ m$^2$/W calculated from the beam deflection measurements with the Z-scan pulse length of 121 fs. Furthermore, the trend of the refractive index change with increasing excitation was measured by Z-scan and is compared in Fig. 3(b) to the beam deflection measurement with relatively good agreement of both measurements. For details of the measurement of the excitation-induced refractive index change $\delta n$, we refer to the Supplementary Material. Also, we note that both the order of magnitude of $n_{2,\text{eff}}$ and the trend with increasing excitation correspond to previous measurements of nonlinear refraction in VECSELs as well as theoretical investigations.\textsuperscript{21-24,38}

We proceed to calculate the pulse-length dependent $n_{2,\text{eff}}$ for several excitation fluences as shown in Fig. 3(c). It can be seen that, for low excitation fluences, $n_{2,\text{eff}}$ is negative for all pulse lengths and increases strongly in magnitude for pulse lengths larger than 1 ps. This is a consequence of the nearly constant refractive index change at longer delays as shown in Fig. 2(b). In contrast, for large excitation fluences, $n_{2,\text{eff}}$ changes sign from negative to positive when going to pulse lengths beyond 1000 fs.

It is interesting to compare the trend of the pulse-width-dependent, as well as the excitation-dependent, effective nonlinear refractive index with previous observations of self-mode-locking in VECSELs. In Ref.\textsuperscript{14}, self-mode-locking in a V-cavity was shown to possibly originate from a negative nonlinear refractive index in the order of $10^{-16}$ m$^2$/W. Interestingly, when increasing the pump power, the measured pulse width decreased from $>1$ ps to $<500$ fs which probably guarantees a negative effective nonlinear refractive index over the whole excitation range as shown by our investigations.

In Ref.\textsuperscript{39}, self-mode-locking was reported for a linear cavity. The insertion of a slit in front of the outcoupling mirror allows only assuming Kerr-lens mode-locking with a nonlinear lens of positive focal length. As the pulse length obtained in this experiment was 3.5 ps, a positive Kerr lens is indeed expected and thus the assumption of Kerr-lens mode-locking caused by a nonlinear refractive index in the order of $10^{-16}$ cm$^2$/W is justified.

Beyond assessing the feasibility of Kerr-lens mode-locking of VECSELs, transient nonlinear refractive index changes generally play a crucial role in modeling pulse formation of VECSELs as it causes chirped pulses. In the semi-classical approach used in Refs.\textsuperscript{25,26} to model SESAM-mode-locking of VECSELs, the refractive index change induced by the changes in carrier occupation is modeled by the so-called linewidth-enhancement factor $\alpha$, that relates transient pulse phase changes $\Delta \phi(t)$ to the time-dependent gain $g(t)$\textsuperscript{25}.

$$\Delta \phi(t) = -\alpha g(t)/2. \quad (4)$$

However, our measurements show that the refractive index dynamics, which directly translates to the pulse phase changes via $\Delta \phi(t) = k\Delta n(t)$, with $k$ being the wave vector, differs significantly in shape and relative contributions of components with different time scales from the gain dynamics. Therefore, it appears useful to incorporate the response function of the refractive index measured here into pulse simulations for more realistic modeling instead of using the phenomenological parameter $\alpha$.

In conclusion, we have probed both the gain and refractive index dynamics of a VECSEL chip under conditions similar to laser operation. Our results allow us to retrieve the response function of the refractive index change and to predict a pulse-length dependent effective nonlinear refractive index, which is negative and in the order of $10^{-16}$ m$^2$/W for sub-ps pulse lengths but becomes positive for large excitation

![Figure 3](image_url)

**FIG. 3.** (a) Effective, excitation-dependent, nonlinear refractive index $n_{2,\text{eff}}$ calculated for a pulse length (FWHM) of 1 ps and several pump peak irradiances. (b) Excitation-induced refractive index change $\delta n$ obtained from a Z-scan measurement conducted with a peak irradiance of 3.3 GW/cm$^2$ and a pulse length (FWHM) of 121 fs and the beam deflection measurement performed with 2.4 GW/cm$^2$ pump peak irradiance. (c) Calculated $n_{2,\text{eff}}$ as a function of the pulse length (FWHM) for different excitation fluences and a pump peak irradiance of 1 GW/cm$^2$. The calculation of $n_{2,\text{eff}}$ (Eq. 3) is based on the fitted response function of the nonlinear refractive index change.
fluences and ps pulse lengths. These findings support the assumption of Kerr-lens mode-locking for some self-mode-locking results obtained with VECSELs. Additionally, our results might improve modeling of pulse formation in VECSELs by providing the time-resolved refractive index response of a VECSEL in the gain regime that could be directly incorporated into pulse-shaping simulations instead of a constant linewidth-enhancement factor.

See the Supplementary Material for further information about the VECSEL characterization and additional beam deflection as well as Z-scan measurements.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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