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Large inherent optical gain from the direct gap transition of Ge thin films

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The recent demonstration of Ge-on-Si diode lasers renews the interest in the unique carrier dynamics of Ge involving both direct (Γ) and indirect (L) valleys. Here, we report a large inherent direct gap optical gain $\geq 1300\,cm^{-1}$ at room temperature from both tensile-strained n^+ Ge-on-Si films and intrinsic Ge-on-insulator using femtosecond transmittance spectroscopy captured before direct-to-indirect valley scattering. This inherent direct gap gain is comparable to III-V semiconductors. For n^+ Ge, this transient gain is $\sim 25\times$ larger than its steady state gain, suggesting that reducing Γ→L or enhancing L→Γ intervalley scattering may significantly increase the optical gain of Ge lasers. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4800015]

Ge-on-Si is a particularly interesting candidate for monolithic lasers on Si platform due to its compatibility with CMOS transistors and pseudo-direct gap behavior suitable for active photonic devices.[1,2] In 2007, we proposed the concept of band-engineered Ge lasers, where tensile strain and n-type doping were combined to compensate the 136 meV energy difference between the direct (Γ) and indirect (L) conduction valleys.[3] Rapid progresses have been made in this field since then.[4] Especially, optical gain and lasing from band-engineered Ge-on-Si have been demonstrated under both optical pumping and electrical pumping.[5–7]

A unique feature of the Ge laser is that it is the only semiconductor laser involving carrier transitions in both direct (Γ) and indirect (L) conduction valleys so far. Therefore, it is important to understand the intervalley carrier dynamics between the Γ and the L valleys. Considering that the optical gain in Ge is dominated by the direct gap transition, it is assumed that increasing the electronic occupation of the direct Γ valley by reducing Γ→L or enhancing L→ Γ intervalley scattering will lead to higher optical gain and better lasing performance. If this assumption is true, one would expect to observe a higher optical gain under ultrafast pumping compared to steady-state pumping since ultrafast spectroscopy captures the optical gain before the injected electrons in the Γ valley fully relax to the L valleys. However, previous studies on the ultrafast spectroscopy of Ge showed inconsistent results.[8–13] For example, instead of observing optical gain, in Refs. 8 and 9, a significant *decrease* in transmittance at 1550 nm was observed upon ultrafast pulse pumping with photon energy well above the direct gap of Ge (~360 meV higher). Contrarily, optical bleaching, or increase in transmittance, was reported in Refs. 10 and 11 with the ultrafast pumping photon energy within 70 meV above the direct band gap of Ge. In a recent study on Ge/SiGe quantum wells (QWs) with an increased direct gap of 0.87 eV due to quantum confinement and compressive strain, a transient gain is only observed at pumping photon energies <0.91 eV, i.e., within ~40 meV of the direct band gap.[13] These results suggest that pumping photon energy may significantly affect the hot carrier relaxation and band filling processes, and subsequently, the gain/bleaching spectra in Ge due to the unique dual-valley transitions. Since carriers are mostly injected near the band edges under electrical pumping, ultrafast spectroscopy using pumping photon energies close to the direct gap is more relevant to electrically pumped Ge lasers. On the other hand, the probing time window also plays a critical role. The lifetime of Γ→L intervalley electron scattering is reported to be ~230 ± 25 femtoseconds (fs) in bulk Ge[10,11] and 185 fs in Ge/SiGe QWs under reverse bias.[12] If the probing time window is less than the intervalley scattering time, the *inherent* optical gain from the direct gap transition of Ge, without being diluted by Γ→L intervalley scattering, can be obtained since all the injected electrons occupy the direct Γ valley within this probing window. This inherent gain of the direct gap transition also reflects the ultimate potential of Ge lasers, and it can be approached by engineering the intervalley scattering to enhance the electronic occupation of the Γ valley.

To elucidate the effect of Γ→L intervalley scattering on the optical gain and single out the *inherent* gain behavior from the direct gap transition of Ge, in this Letter, we report injection-level-dependent broad-band fs transmittance spectra captured *before* direct-to-indirect valley scattering, using photon energies close to the direct gap to resemble the electrical injection of carriers near the band edge. We reveal a large inherent direct gap optical gain $\geq 1300\,cm^{-1}$ at an injected carrier density of $\sim 10^{19}\,cm^{-3}$ for both intrinsic Ge-on-insulator (GeOI) and tensile-strained n^+ Ge-on-Si films at room temperature. This inherent direct gap gain is comparable to III-V semiconductors, and 1–2 orders greater than the steady-state gain in Ge at the same injection level. Our result confirms that reducing Γ→L intervalley scattering may significantly increase the optical gain in Ge and decrease the lasing threshold. Together with our recent observation of enhanced L→Γ intervalley scattering in tensile-strained n^+ Ge,[14] these results indicate that the performance of Ge lasers can be further improved by engineering the

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intervalley scattering for enhanced electronic occupation of the Γ valley. Additionally, the result on injection dependence of fs transmittance spectra is directly relevant to the saturation absorption behavior of Ge, which can be applied to saturable absorbers and other important nonlinear photonic devices.[15]

The injection-dependent ultrafast transmittance spectroscopy in this study utilizes broad-band fs incident pulses at $\lambda = 1500$–1700 nm (Fig. 1) and measures the transmittance spectra at different incident pulse intensities/injection levels, similar to the technique described in Ref. 15. Since a notable fraction of the incident photons have energies greater than the direct band gap of the Ge samples, the increase in incident pulse intensity can induce population inversion, which modifies the transmittance spectra due to saturation of absorption and optical gain. Compared to the conventional pump-probe method, this experimental setup is more suitable for probing the transient gain with minimal delay after the excitation since most of the photons in the broad-band incident pulse arrive at the sample within the duration of the fs pulse width. A Newport Tsunami Ti-sapphire fs laser with a pulse width of ~50 fs and a tunable central wavelength around 800 nm is used as the seed laser in the ultrafast spectroscopy.[16] All the pulse widths reported in this study are directly measured by a Spectra-Physics autocorrelator. The laser pulses are amplified by a Spitfire amplifier to reach an output pulse energy of 1 mJ with a pulse width <60 fs. These high energy pulses are applied to drive an OPA-800 C optical parametric amplifier, where an input photon is "split" into two lower energy photons, known as "signal" and "idler," respectively. The signals and idlers are combined as incident pulses to cover the wavelength range of 1500–1700 nm (Fig. 1) for investigating the direct gap transition of Ge. The pulse energy is ~20 μJ, which can be attenuated by a variable attenuator wheel for measuring fs transmittance spectra at different injection levels. The broadband fs pulses are focused on the surface of the Ge samples, and an optical spectrometer is used to measure the transmittance spectra. Even with the dispersion in glass lenses in the optical path, the measured pulse width is still less than 80 fs when focused on the Ge samples.

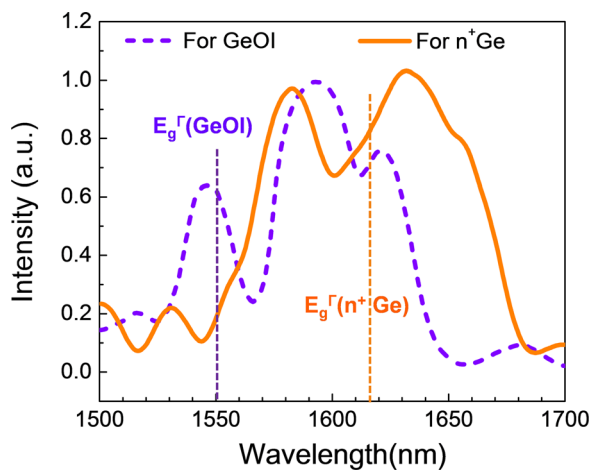

FIG. 1. Spectra of incident pulses for the injection-level-dependent fs transmittance spectroscopy of the tensile strained n^+ Ge and GeOI samples. The spectra are tuned such that there is a notable amount of incident photons with energies greater than the direct gap of Ge thin films for carrier injection in each case.

The GeOI sample from Umicore consists of a 3.58 μm thick intrinsic Ge layer and a 1 μm thick buried SiO_2 on a double-side polished Si substrate with a 1.5 μm thick backside oxide. The epitaxial n^+ Ge film with a thickness of 0.74 μm and a uniform phosphorus doping concentration of 1×10^{19} cm^{-3} is grown on a single-side polished (SSP) Si (001) substrate by ultrahigh vacuum chemical vapor deposition (UHVCVD).[5] The thermally induced tensile strain in the Ge film is 0.23% according to X-ray diffraction (XRD) analysis. Correspondingly, the direct band gap of the tensile strained n^+ Ge film is reduced from 0.8 eV to 0.767 eV.[5] The doping-induced band gap narrowing (BGN) at $n = 1 \times 10^{19}$ cm^{-3} is negligible.[5,17] As shown in Fig. 1, the spectra of the incident pulses are tuned such that there is a notable amount of incident photons with energies greater than the direct band gap of GeOI and n^+ Ge-on-Si, respectively. These photons are strongly absorbed by the Ge films and, at a high enough pulse intensity, could induce population inversion and lead to optical bleaching, saturation of absorption, and gain. Considering that our pulse width is <80 fs, much smaller than the Γ→L electron intervalley scattering lifetime of ~230 ± 25 fs reported in bulk Ge,[10,11] our measurement can capture the *inherent* direct gap optical gain of Ge films *before* the Γ→L intervalley scattering. Injected carrier densities were initially estimated from the pulse energy, pulse spectra, focal spot size, and optical absorption of Ge thin films.[5,6,18] For a refined estimation of the injection level, we use the onset energy of the optical gain spectrum to determine the direct band gap,[19] then apply the universal formula describing the direct band gap renormalization and shrinkage in highly excited semiconductors to determine the injected carrier density.[20] This method will be discussed in more detail later.

Fig. 2(a) shows the fs transmittance spectra of the GeOI sample measured at two different injection levels/pulse intensities. A calculated transmittance spectrum using the refractive index of bulk Ge[18] is also shown for comparison, neglecting the effect of injection. In the calculated transmittance curve, there are three interference peaks at $\lambda = 1500 \sim 1750$ nm. The interference peak intensity increases with wavelength due to the decreasing absorption coefficient beyond the direct band gap ($\lambda > 1550$ nm) of GeOI. At a relatively low injection level (hollow circles in Fig. 2(a)), the transmittance spectrum is largely consistent with the calculated curve. The slight blueshift at $\lambda > 1600$ nm is due to the change in refractive indices upon injection. As the injection level further increases (solid circles in Fig. 2(a)), the spectrum shows an additional transmittance peak right at the interference valley of the calculated curve in the wavelength range of 1580–1605 nm, suggesting the presence of optical gain. Using the transfer matrix analysis and Kramers-Kronig (K-K) relation, we derive the real part of the refractive index and the absorption coefficient of GeOI upon high excitation deterministically from the transmittance data via an iterative self-consistent regression approach.[5] As shown in Fig. 2(b), negative absorption coefficients corresponding to net gain are observed in the wavelength range of 1580–1605 nm, and the shape of the gain spectrum is similar to that of III-V semiconductors.[19] Accordingly, the real part of the refractive

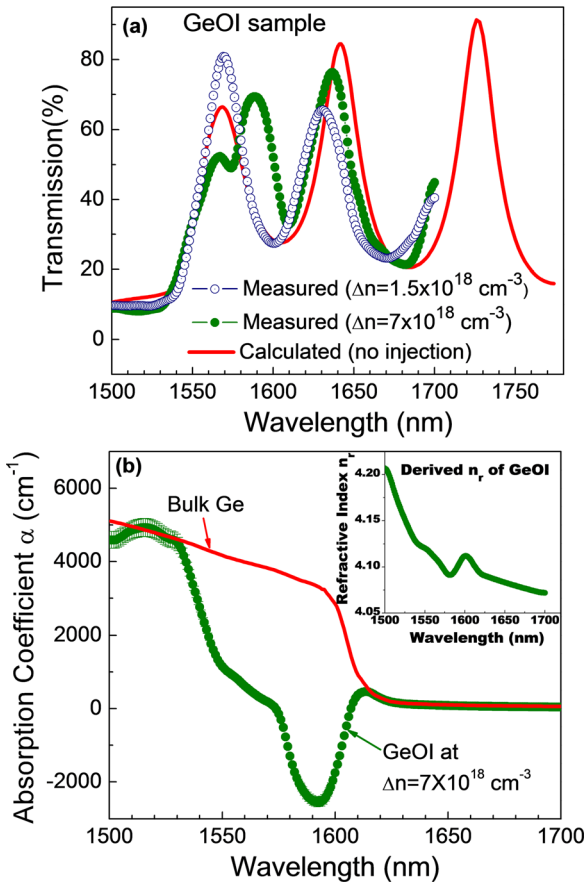

FIG. 2. (a) Comparison of the measured transmittance spectra of the GeOI sample under a low injection ($1.5 \times 10^{18}\,cm^{-3}$, hollow circles) and a high injection ($7 \times 10^{18}\,cm^{-3}$, solid circles) with the theoretical calculation (solid curve, without considering the injection effect); (b) Derived absorption spectrum of the GeOI sample under $7 \times 10^{18}\,cm^{-3}$ carrier injection in comparison with the absorption spectrum of bulk Ge.[18] The latter has been redshifted by 30 meV to accomodate band gap narrowing in the former for a direct comparison. The inset shows the derived real part of the refractive index (n_r) of GeOI at $\Delta n = 7 \times 10^{18}\,cm^{-3}$.

index shows a kink in this wavelength range due to K-K relation. The onset wavelength of optical gain at 1605 nm corresponds to a direct band gap of 0.77 eV,[19] which indicates band gap renormalization and a 30 meV BGN of the direct gap transition due to many-body effect at a high injection level. To verify the validity of the derived absorption spectrum of GeOI under the high injection level, we also show the absorption spectrum of bulk Ge[18] with the direct band gap shifted from 0.8 eV to 0.77 eV for a direct comparison. One would expect that the absorption coefficients remain the same at wavelengths away from the direct band edge where no bleaching or gain occurs. Indeed, the direct gap absorption at $\lambda = 1500 \sim 1530$ nm and the indirect band gap absorption at $\lambda = 1610 \sim 1700$ nm remain the same, indicating the validity of our self-consistency iteration method. Based on the universal formula on BGN described in Ref. 20, we further determined that the 30 meV BGN corresponds to an injected carrier density of $\Delta n = 7 \times 10^{18}\,cm^{-3}$ using the following material parameters of Ge: dielectric constant $\varepsilon_{Ge} = 16$, electron effective mass at Γ valley $m_e^{\Gamma} = 0.038 m_0$ (m_0 is the electron mass), light hole effective mass $m_{lh} = 0.043 m_0$, and heavy hole effective mass $m_{hh} = 0.284\, m_0$.[21] The maximum net gain of GeOI is $\sim 2500\,cm^{-1}$ at $\lambda = 1593$ nm, comparable to III-V semiconductor QWs under a similar injection level of $\Delta n \sim 1 \times 10^{19}\,cm^{-3}$.[22] In addition to the optical gain at 1580–1605 nm, the optical bleaching spans from 1530 to 1605 nm, as shown in Fig. 2(b). With a similar approach of analysis, we found that the relatively low injection level (hollow circles) in Fig. 2(a) corresponds to $\Delta n = 1.5 \times 10^{18}\,cm^{-3}$. A net gain is achieved in the wavelength range of $1564 \sim 1585$ nm, and the maximum gain is $\sim 1000\,cm^{-1}$ at 1570 nm. Note that although optical bleaching has been observed in GeOI under steady-state optical pumping,[23] no gain was observed in that case since most of the injected electrons relax to the indirect L valleys. It is only with fs transmittance spectroscopy captured before $\Gamma \rightarrow L$ intervalley scattering that we can observe such a large inherent optical gain from the direct gap transition of Ge.

For the n^+ Ge-on-Si sample, we compare the transmittance of n^+ Ge-on-Si to that of the Si substrate directly in order to eliminate the influence of backside roughness of the SSP Si wafer. As shown in Fig. 3(a), at the low injection level, there is hardly any population inversion, so the transmittance starts to decrease significantly at $\lambda < 1625$ nm due to the onset of direct gap absorption. At the high injection level, the transmittance spectrum shows obvious optical bleaching at $\lambda = 1500$–1700 nm. Remarkably, the transmittance through the tensile strained n^+ Ge-on-Si sample at $\lambda = 1610$–1693 nm is even greater than the Si substrate itself.

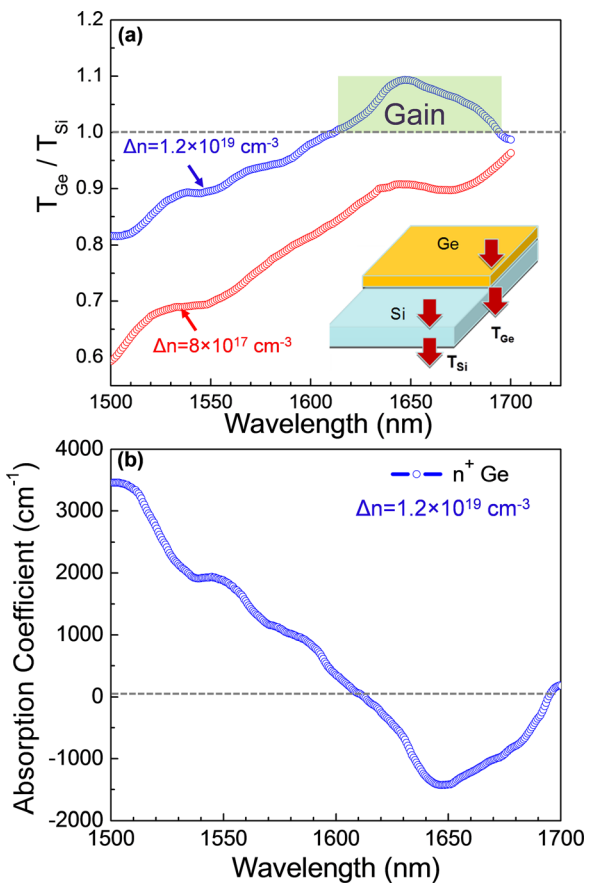

FIG. 3. (a) Transmittance spectra through the n^+ Ge-on-Si sample in reference to the Si substrate under the injection levels of $1.2 \times 10^{19}\,cm^{-3}$ and $8 \times 10^{17}\,cm^{-3}$. T_{Ge} and T_{Si} refer to the transmittance through the n^+ Ge-on-Si sample and the Si substrate alone, respectively. (b) Derived absorption spectrum of the n^+ Ge sample under $1.2 \times 10^{19}\,cm^{-3}$ carrier injection.

Since Ge has a higher refractive index than Si (4.2 vs. 3.5 at 1640 nm), the reflection on the Ge surface can only be higher than that of Si (37.4% vs. 30.5%). Consequently, this enhanced transmittance through the n^+ Ge film cannot be explained by anti-reflection coating effect. The entire incident photon spectrum is well below the band gap of Si, and we confirmed that the optical pulses did not generate any non-linear effects (such as two-photon absorption) to reduce the transmittance of the Si substrate. Therefore, the higher transmittance at $\lambda = 1610$–1693 nm through the Ge-on-Si sample compared to the Si substrate can only be attributed to optical gain. The absorption coefficient, α, can be estimated from the relative transmittance ratio $T_{Ge}/T_{Si} = \exp(-\alpha L)$, where L is the thickness of the Ge film (0.74 μm). The derived absorption spectrum at the higher injection level is shown in Fig. 3(b). The onset of enhanced transmittance at 1693 nm corresponds to 35 meV direct band gap shrinkage from the original direct band gap of 0.767 eV,[5] indicating an injected carrier density of $1.2 \times 10^{19}\,cm^{-3}$ using the same BGN model mentioned earlier. Accordingly, the lower injection level is estimated to be $\sim 8 \times 10^{17}\,cm^{-3}$ based on the ratio of pulse intensities. Note that the injected carrier density in tensile-strained n^+ Ge is a bit higher than that of GeOI ($1.2 \times 10^{19}\,cm^{-3}$ vs. $7 \times 10^{18}\,cm^{-3}$) under a similar incident power density because the tensile strain leads to a smaller band gap, and consequently, a larger fraction of photons absorption in the incident pulses. Based on the band gaps and incident pulse spectra in Fig. 1, 38% of the incident photons are absorbed by the tensile-strained n^+ Ge sample vs. 20% by GeOI. The ratio of absorbed photons is 1.9, in good agreement with the ratio of our estimated injected carrier density ($\sim$1.7) from BGN analysis.

It is also interesting to note that, compared to GeOI, tensile-strained n^+ Ge has a much broader spectral width for optical bleaching (200 nm vs. 75 nm) and gain (83 nm vs. 25 nm), while its peak gain of $\sim 1300\,cm^{-1}$ at $\lambda = 1645$ nm is smaller than that of GeOI ($2500\,cm^{-1}$ at $\lambda = 1593$ nm). In addition to the larger wavelength range of absorbed photons by tensile strained n^+ Ge than GeOI (Fig. 1), it is also possibly related to the enhanced electron-electron scattering in n^+ Ge compared to GeOI. Both factors lead to a broader energy distribution of injected electrons in Γ valley in <100 fs time frame. The broader energy distribution of non-equilibrium electrons can lead to a broader gain spectrum at the cost of a lower peak gain according to nonequilibrium statistical mechanics.[24] For n^+ Ge, the inherent direct gap gain is 25$\times$ greater than the steady-state optical gain with similar injected carrier density since only a few percent of injected electrons occupy the direct Γ in the latter case.[5] An implication of this study is that if we could reduce the $\Gamma \rightarrow$L (direct-to-indirect) intervalley scattering rate, or conversely, enhance the L$\rightarrow\Gamma$ (indirect-to-direct) intervalley scattering, then a much higher steady-state optical gain can be achieved for more efficient Ge CW lasers. For example, tensile strain can reduce L-Γ energy separation and lead to a strong $L \rightarrow \Gamma$ intervalley scattering absorption, which has been observed recently in the infrared absorption spectra of 0.25% tensile strained n^+ Ge films[14] yet never reported in n^+ bulk Ge.[8] On the other hand, pressure-dependent *ab initio* calculation shows that the contributions of various phonons to the intervalley scattering are quite dependent on the conduction band energy separation.[25] Further combination of tensile strain and phonon engineering with nanostructures[26] may help to achieve this goal.

In conclusion, we report a large *inherent* direct gap optical gain of $\geq 1300\,cm^{-1}$ from both intrinsic and n^+ Ge-on-Si films at room temperature based on injection-level-dependent femtosecond transmittance spectroscopy captured *before* direct-to-indirect valley scattering. This inherent direct gap gain is comparable to III-V semiconductors, indicating that reducing $\Gamma \rightarrow$L or enhancing L$\rightarrow\Gamma$ intervalley scattering may significantly enhance the optical gain of Ge CW lasers. These results offer fundamental understanding of inherent direct gap optical gain of Ge thin films for further improvement of Ge lasers.

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