which becomes much more negative in the confined plasma ($V_P = -1.55$ V and $V_F = -11.9$ V) than in the unconfined one ($V_P = 0.3$ V and $V_F = -1.5$ V). The ion bombardment energy of an isolated substrate is $e(V_F - V_P)$. In the confined plasma, a negative V_F therefore favors a moderate bombardment energy (≈ 10 eV). Both the ion bombardment and high density of electrons in the 10-eV range produce a high surface reactivity and contribute to a dense a-Si:H film growth.[13]

Finally, in the electrostatically confined plasma, a high a-Si:H deposition rate has been obtained. At pressures as low as 6 μbar, the deposition rate is 0.33 nm/s. Both the very reproducible low-pressure plasma properties of the electrostatically confined dc glow discharge and the high deposition rate are interesting features for the deposition of a-Si:H by this glow discharge technique.

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Spectral hole-burning and gain saturation in semiconductor lasers: Strong-signal theory

Govind P. Agrawal
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

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The nonlinear susceptibility of semiconductor gain media is obtained by including the effect of intraband carrier relaxation within the density-matrix formalism. The result is used to obtain the approximate analytic expressions showing how the optical gain and the refractive index change with an increase in the laser power. The index change is generally small but can be positive or negative depending on which side of the gain peak the lasing mode is located. By contrast, the gain is always reduced because of spectral hole-burning. The implications of gain saturation on the dynamic response of semiconductor lasers are discussed together with the possibility of experimental verification.

Spectral hole-burning, a phenomenon responsible for a power-dependent reduction of the optical gain,[1–4] has been found to significantly affect the performance of semiconductor lasers.[5–13] In particular, it increases the damping rate of relaxation oscillations and affects the modulation response. The effect of spectral hole-burning is generally included in the single-mode rate equations by writing the gain in the form[9,10] $g = g_L(1 - P/P_s)$, where g_L is the mode gain at threshold, P is the output power, and P_s is the saturation power. Typical values of P_s are in the range 0.1–1 W depending on the laser wavelength and material.[4] The linear form of the saturated gain can be derived using the third-order perturbation theory within the density-matrix approach,[1–4] and is valid for $P \lesssim 10$ mW. However, at higher operating powers it is necessary to modify the functional form of the nonlinear gain. This is particularly the case for InGaAsP lasers for which P_s is relatively small ($P_s \sim 100$ mW).

An alternative form of the gain, $g = g_L/(1 + P/P_s)$ has been used in the single-mode rate equations[5,11] to go beyond the third-order perturbation theory. This form is based on the theory of gain saturation in homogeneously broadened two-level systems[14,15] and is not generally valid for semiconductors lasers. In this communication we use the density-matrix approach to obtain an expression for the optical gain in semiconductor lasers that includes the saturation effects. In this approach the semiconductor-gain medium is modeled as an ensemble of two-level systems with different transition frequencies distributed in accordance with the joint density of states.[1–4,15–17] Although in general a numerical approach is necessary,[16,17] we show that an approximate analytic expression for the nonlinear part of the optical gain can be obtained. We discuss the implications of the nonlinear gain on the dynamic and modulation performance of semiconductor lasers. In particular, we show that gain saturation leads to a sublinear increase of the damping rate of relaxation oscillations with an increase in the optical power.

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If we assume that the laser oscillates in a single longitudinal and transverse mode, the electric field inside the laser cavity can be written as

$$\mathbf{E}(\mathbf{r},t) = \tfrac{1}{2}\hat{x}[E_0 U(\mathbf{r})\exp(-i\omega_0 t) + \text{c.c.}], \tag{1}$$

where ω_0 is the optical frequency, $\hat{x}$ is the polarization unit vector, and $U(\mathbf{r})$ is the spatial distribution of the fundamental mode supported by the laser waveguide. The induced polarization $\mathbf{P}(\mathbf{r},t)$ is calculated by considering the dipole response of a single two-level system with the transition frequency ω_T and then summing over all possible band-to-band transitions, i.e.,

$$\mathbf{P}(\mathbf{r},t) = \int_{\omega_g}^{\infty} \mu(\omega_T) D(\omega_T)(\rho_{12} + \rho_{21})\, d\omega_T, \tag{2}$$

where ω_g is the band-gap frequency, μ is the dipole moment, D is the joint density of states, and ρ_{12} and ρ_{21} are the off-diagonal density-matrix elements. For a two-level system, the density-matrix equations can be solved exactly[14,15] with the following nonperturbative solution for ρ_{12}:

$$\rho_{12} = \frac{\mu\tau_{\text{in}}}{i\hbar}\,\frac{(\bar\rho_{11} - \bar\rho_{22})}{1 + i\Delta}\,\frac{1+\Delta^2}{1+\Delta^2+I}\,U E_0 \exp(-i\omega_0 t), \tag{3}$$

where $\bar\rho_{11}$ and $\bar\rho_{22}$ are the occupation probabilities for electrons and holes in thermal equilibrium, Δ is the normalized detuning parameter, and I is the normalized intensity. I and Δ are defined by

$$I = |E_0|^2/I_s, \tag{4}$$

$$\Delta = (\omega_T - \omega_0)\tau_{\text{in}}, \tag{5}$$

where the saturation intensity

$$I_s = \{(\mu/\hbar)^2 \langle |U(\mathbf{r})|^2\rangle \tau_{\text{in}}(\tau_c + \tau_v)\}^{-1} \tag{6}$$

and τ_c, τ_v, and τ_{in} are the intraband relaxation times for electrons, holes, and polarization, respectively. The angular brackets in Eq. (6) denote average over the active volume. As a result, I in Eq. (3) is an average intracavity intensity. This procedure, although not exact, leads to considerable simplification in the following analysis. In the axial direction, it amounts to averaging over the spatial holes burnt by the counterpropagating waves in the carrier population. This is justified for semiconductor lasers since carrier diffusion tends to wash out the spatial holes.

The susceptibility χ that determines the medium response to an applied field is obtained by substituting Eq. (3) in Eq. (2) and expressing the result in the form

$$P(\mathbf{r},t) = (\epsilon_0/2)[\chi E_0 U(\mathbf{r})\exp(-i\omega_0 t) + \text{c.c.}]. \tag{7}$$

The result is

$$\chi = -\int_a^{\infty} \frac{1+\Delta^2}{1+\Delta^2+I}\,\frac{f(\Delta)d\Delta}{\Delta - i}, \tag{8}$$

where

$$f(\Delta) = \Gamma\mu^2 D(\bar\rho_{11} - \bar\rho_{22})/(\epsilon_0\hbar). \tag{9}$$

The confinement factor Γ appears because $\bar\rho_{11} - \bar\rho_{22} = 0$ outside the active region. The lower limit of integration $a = (\omega_g - \omega_0)\tau_{\text{in}} < 0$, since generally ω_0 exceeds the band-gap frequency ω_g.

In general, the susceptibility χ given by Eq. (8) should be evaluated numerically.[16,17] Here we adopt an analytic approach that enables us to obtain an approximate functional form of the nonlinear gain. For this purpose we separate χ into linear and nonlinear parts defined by

$$\chi = \chi_{\text{L}} + \chi_{\text{NL}}, \tag{10}$$

where

$$\chi_{\text{L}} = -\int_a^{\infty} \frac{f(\Delta)d\Delta}{\Delta - i}, \tag{11}$$

$$\chi_{\text{NL}} = I\int_a^{\infty} \frac{f(\Delta)d\Delta}{(\Delta - i)(1+\Delta^2+I)}. \tag{12}$$

The evaluation of χ_{L} still requires numerical integration. However, χ_{NL} can be evaluated approximately using the method of contour integration. The approximation is based on the observation that the main contribution to the integral comes from a region $|\Delta| < 1$. We assume the $f(\Delta)$ varies slowly in this region and can be approximated by

$$f(\Delta) \simeq f(0) + f'\Delta, \tag{13}$$

where $f' = df/d\Delta$ is evaluated at $\Delta = 0$. The lower limit of integration in Eq. (12) can be replaced by $-\infty$ for the same reason. By closing the contour in the lower-half plane, we find that a single pole at $\Delta = -i\sqrt{1+I}$ contributes to the integral. Evaluating the residue at this pole, we obtain

$$\chi_{\text{NL}} \simeq \frac{i\pi f(0)(1 - i\beta\sqrt{1+I})I}{\sqrt{1+I}\,(1+\sqrt{1+I})}, \tag{14}$$

where $\beta = f'/f(0)$ is a dimensionless parameter.

The parameter $f(0)$ can be related to the linear gain as follows. The induced susceptibility changes the dielectric constant by $\Delta\epsilon$ which manifests as a change in the refractive index and the optical gain according to the relation[13]

$$\chi = \Delta\epsilon = 2n(\Delta n - ig/2k_0), \tag{15}$$

where n is the background index, $k_0 = \omega_0/c$, g is the optical gain, and Δn is the index change that accompanies the gain. Similar to Eq. (10), both Δn and g can be decomposed into linear and nonlinear parts. In particular, the linear parts from Eq. (11) are given by

$$\Delta n_{\text{L}} = -\frac{1}{2n}\int_a^{\infty} \frac{\Delta f(\Delta)d\Delta}{1+\Delta^2} \simeq -\frac{\beta_c g_L}{2k_0}, \tag{16}$$

$$g_L = \frac{k_0}{n}\int_a^{\infty} \frac{f(\Delta)d\Delta}{1+\Delta^2} \simeq \frac{\pi k_0}{n} f(0). \tag{17}$$

The approximate form of the linear gain g_{L} is obtained by using the same approximations as those used in the derivation of Eq. (14). The integral in Eq. (16) generally requires a numerical approach.[16] The last term relates Δn_{L} to g_{L} in a phenomenological manner. The proportionality constant β_c is often referred to as the linewidth broadening factor.[13,18]

Using Eqs. (14), (16), and (17) and a relation similar to Eq. (15) for χ_{NL}, we obtain the nonlinear index and gain:

$$\Delta n_{\text{NL}} \simeq -\frac{\beta\Delta n_{\text{L}}}{\beta_c}\,\frac{I}{1+\sqrt{1+I}}, \tag{18}$$

$$g_{\text{NL}} \simeq -\frac{g_{\text{L}} I}{\sqrt{1+I}\,(1+\sqrt{1+I})}. \tag{19}$$

Equations (18) and (19) are our main results. They show how the refractive index and the optical gain in the active region of a semiconductor laser change with an increase in the optical intensity. The index change depends on the parameter β that can be related to the slope of the linear-gain profile at the laser frequency, i.e.,

$$\beta = \frac{f'}{f(0)} = \frac{1}{g_L(\omega_0)\tau_{in}} \left(\frac{dg_L}{d\omega}\right)_{\omega_0}. \qquad (20)$$

This parameter $\beta = 0$ if the lasing occurs exactly at the gain peak. However, for distributed feedback semiconductor lasers it can be positive or negative depending on which side of the gain peak the lasing mode is situated. Using typical parameter values ($\tau_{in} = 0.1$ ps), we estimate that $\beta = \Delta\lambda/20$, where $\Delta\lambda$ is the wavelength deviation in nanometers from the gain peak.[4] Since generally $\beta < 1$ and $\beta_c = 2$–8 depending on the laser wavelength,[18] Δn_{NL} is small compared to Δn_L particularly for InGaAsP lasers for which β_c is relatively large. It is interesting to note that for $I \gg 1$, Δn_{NL} increases with I as $\sqrt{I}$.

The nonlinear-gain expression (19) shows that the linear gain is reduced by an amount g_{NL} with an increase in the optical intensity ($I = |E_0|^2/I_s$). This effect is similar to that occurring in gas lasers[14] and is often referred to as spectral hole-burning. For semiconductor lasers, however, the hole width ($\sim 10^{13}$ s^{-1}) is nearly comparable to the spectral width of the gain profile because of an extremely short intraband relaxation time $\tau_{in} \sim 0.1$ ps (homogeneous linewidth $\sim \tau_{in}^{-1}$). As a result, the semiconductor gain medium saturates almost homogeneously. Indeed, the approximate result (19) shows that the frequency dependence of g_{NL} is identical to that of g_L, i.e., the gain is reduced homogeneously by the same amount over the whole spectral profile. The intensity dependence of g_{NL} is not, however, identical to that of a homogeneously broadened two-level system as assumed in some previous work.[5,11] In fact, the intensity dependence has a functional form identical to that of an inhomogeneously broadened two-level system since

$$g = g_L + g_{NL} = g_L/\sqrt{1+I}, \qquad (21)$$

as can be readily verified using Eq. (19). This is not surprising if we note that the range of possible transition frequencies is extremely wide for semiconductor gain media. For optical intensities much below the saturation level, $I \ll 1$, and the saturated gain can be approximated by

$$g \simeq g_L(1 - I/2). \qquad (22)$$

This is in agreement with the results of third-order perturbation theory[1–4] and justifies the form of nonlinear gain that is often used in the rate-equation analysis.[9–13]

For the purpose of comparison with experiments, it is important to estimate the power levels for which Eq. (22) is a reasonable approximation for the saturated gain. An order of magnitude estimate of the saturation intensity I_s can be obtained from Eq. (6) if we assume for simplicity that $\langle |U(r)|^2 \rangle \simeq 1$. Using estimated parameter values $\mu \simeq 7 \times 10^{-29}$ C m, $\tau_{in} = 0.1$ ps, $\tau_c = 0.3$ ps, and $\tau_v = 0.07$ ps, thought to be appropriate for InGaAsP lasers,[4] we obtain $I_s \sim 10$ MW/cm^2. Since the mode cross section is typically ~ 1 μm^2 for index-guided semiconductor lasers, the saturation power is ~ 100 mW. Thus, Eq. (22) is a good approximation for operating powers up to about 10 mW. At higher power levels Eq. (19) or (21) should be used in the single-mode rate equations to accurately model the dynamic and modulation properties of semiconductor lasers.

It is important to note that the linear gain g_L in Eq. (21) is not the small-signal gain but includes the saturation effects resulting from interband transitions (due to clamping of the carrier density at threshold). As is well known,[19,20] the linear gain saturates homogeneously according to $g_L = g_0/(1 + I/I_0)$, where the interband saturation intensity $I_0 \sim 1$ MW/cm^2 is nearly two orders of magnitude lower than the intraband saturation intensity I_s. Thus, for semiconductor lasers or amplifiers operating at power levels $\sim$1–5 mW, the gain meduim saturates almost homogeneously, as also observed experimentally,[19] since the intraband saturation or spectral hole-burning is negligible. However, departures from this homogeneous-saturation behavior should be observable at power levels exceeding 10 mW or so.

To study the implications of the saturated gain of Eq. (21) on the device performance, we have solved the modified rate equations in the small-signal approximation using the standard method.[13] The modulation response (normalized to its dc value) is given by

$$\frac{H(\omega)}{H(0)} \simeq \frac{\Omega^2}{(\Omega^2 - \omega^2 + i\omega\Gamma)}, \qquad (23)$$

where Ω is the relaxation-oscillation frequency and the damping rate Γ is given by

$$\Gamma \simeq \frac{I/2}{(1+I)^{3/2}} \tau_p^{-1}. \qquad (24)$$

Here τ_p is the photon lifetime (typically 1–2 ps) and we have neglected a small contribution to Γ arising from spontaneous emission.[13] Equation (24) predicts a sublinear increase of Γ with the operating power. Such a behavior has been experimentally observed for InGaAsP lasers and was attributed to the onset of multilongitudinal modes at high powers.[21] Our analysis suggests that the gain saturation may have been responsible in part for the observed sublinear behavior. A clear-cut verification of Eq. (24) would require the development of a distributed feedback semiconductor laser that maintains a single longitudinal mode up to relatively high operating powers. In conclusion, we note that several other predictions (e.g., the nonlinear-gain-limited modulation bandwidth[9]) may need revision when the form (21) in place of (22) is used in the single-mode rate equations.

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Resist heating in excimer laser lithography

Takayuki Abe, Tsunetoshi Arikado, and Tadahiro Takigawa
VLSI Research Center, Toshiba Corporation, 1 Komukai, Toshiba-cho, Saiwai-ku, Kawasaki 210, Japan

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Intensity dependence of the photochemical reaction rate, and thus the exposure reciprocity, for excimer laser beam irradiation to resist has been investigated. The reciprocity law did not hold for KrCl excimer laser beam irradiation concerning the sensitivity of the poly(methyl methacrylate) (PMMA) resist. It is also shown that this failure of the reciprocity law originates from a resist sensitivity change caused by the resist temperature rise due to irradiation (resist heating). For PMMA, the laser beam with a pulse energy of 15 mJ/cm^2 and a wavelength of 222 nm raised the resist temperature by about 30 °C, resulting in a thermally assisted photochemical reaction.

The design rule for very large scale integrated circuits (VLSI) devices will decrease down to 0.5–0.25 μm in the near future. Conventional optical lithography has a resolution limit around 0.5 μm. Therefore, a next generation lithography is stongly desired. Excimer laser reduction technology is expected to break through such a resolution limit.[1–6]

However, many phenomena to be clarified exist in excimer laser lithography. One of the phenomena is the failure of the reciprocity law to hold. The reciprocity law comes from the fact that chemical reaction between photons and the sensitive material (resist) only depends on the exposure dose, that is, its functional is f [(intensity) × (exposure time)] and not f (light intensity). The law holds in conventional optical lithography. However, in excimer laser lithography, some experimental results have shown the failure of reciprocity. Kawamura, Toyoda, and Namba[2] reported that the development rate of the resist (i.e., chemical reaction rate) increases with excimer laser beam intensity.

Dill's model for the photochemical reaction of a resist satisfies the reciprocity law in conventional optical lithography.[7] However, the model does not explain the failure of the reciprocity law in excimer laser lithography. Albers and Novotny[8] introduced a two-step kinetic model and estimated the intensity dependence of the photochemical reaction rate. They showed that the chemical reaction rate decreases with the increase in light intensity, over 5 kW/cm^2. This dependence is contrary to that of the above experimental results.

In this communication it is demonstrated that the failure of the reciprocity law originates from a change in the chemical reaction rate caused by the resist temperature rise due to irradiation (resist heating). The resist heating effect is a serious problem in mask making by electron beam writing which deforms the resist pattern, resulting in degradation of pattern accuracy.[9–11]

Figure 1 shows the residual resist thickness for several exposure doses. A KrCl excimer laser (222 nm) was used as the light source. A 1-μm poly(methyl methacrylate) (PMMA) resist was coated on Si substrates and was exposed with several laser intensities. The exposed resist was developed with methyl isobutylketon (MIBK) in 6 min. The development rate increased with beam intensity for the same exposure dose. The reciprocity law did not hold.

The temperature rise of the resist was estimated as follows. In excimer laser lithography, the resist is exposed with a pulse energy of about 30 mJ/cm^2 for about 10 ns. The laser intensity reaches 3 MW/cm^2. The pulse exposure is repeated with an interval of 1–100 ms. The accumulation of heat between each pulse exposure was ignored as follows. The Si substrate acts as a heat sink because the thermal diffusion coefficient of Si is about 10^3 times as large as that of the PMMA resist. The temperature rise of the resist may be ignored after the heat, generated at the resist surface, diffuses and reaches the heat sink (Si). This diffusion time t_d is represented by

$$t_d = d^2/(4D_r), \qquad (1)$$

where d represents resist thickness and D_r the thermal diffusion coefficient. Substituting the values of $d = 1$ μm and $D_r = 10^{-7}$ m^2/s into Eq. (1), a diffusion time of 3 μs is ob-