Cancellation of *B*-Integral Accumulation for CPA Lasers

Self-phase modulation (SPM) plays an important role in determining the final shape of the compressed pulse in chirped pulse amplification (CPA) lasers, even at relatively low values of the cumulative *B*-integral, $B\sim1$ to $2^{.1,2}$ The SPM distorts the linear chirp and causes the compressed pulse duration to increase noticeably near $B\sim1$ and approximately double for $B\sim2^{.1,2}$ It is possible to reset the expansion or compression gratings to compensate for this effect on average, but the radial variation of the *B*-integral means that it cannot be compensated for exactly. Temporal structure (wings) will unavoidably appear in a recompressed pulse, even for relatively small values of *B*.

We report the cancellation of the *B*-integral accumulated by a chirped pulse in a regenerative amplifier by using a GaAs plate that has a negative nonlinear index of refraction at 1.053 μ m.³ The absolute value of the nonlinear index of refraction is about three orders of magnitude larger than that of Nd:glass or KDP, so a thickness of less than 1 mm is required. We show that the compressed pulse duration and wings measured as a function of accumulated *B*-integral increase without the compensator plate as *B* exceeds 1, while with the GaAs plate installed the pulse duration remains fixed and the wings are reduced to almost their unmodulated value. The cancellation of the nonlinear phase was demonstrated with a 4.6-m Xe gas cell for a 10-ps KrF laser operating at 248.4 nm, which is just above two-photon resonance with the Xe 6*p*[1/2]₀ state.⁴

1.053- μ m chirped pulses are generated in a Nd:YLF oscillator coupled into a 0.8-km, single-mode fiber. They are subsequently expanded in a conventional single-grating, lens and mirror stretcher up to a 0.45-ns duration. One pulse is seeded into a *Q*-switched regenerative amplifier. Due to strong gain narrowing, the stretched pulse becomes Gaussian in time with a 0.3-ns duration.⁵ As the pulse envelope builds up in the regenerative amplifier, the *B*-integral is accumulated on every pass. Near the peak of the *Q*-switched envelope the *B*-integral is approximately 0.18 per pass. By varying the switch-out time of the pulse relative to the peak of the pulse train, we vary the *B*-integral accumulated by the output pulse. The pulse passes through a pair of compression gratings, and the pulse duration is measured with an autocorrelator.

We modeled the regenerative amplifier using the Frantz-Novdik system of equations similar to Ref. 6 to estimate the cumulative *B*-integral. The round-trip loss factor is 50% due to the output coupler, and the 6.3% additional intracavity losses derived from equations are in good agreement with 55% measured with a cold cavity. The gain-narrowed pulse width is still slightly longer than the terminal-level lifetime of the active host. Although the final simulation values of output parameters are indistinguishable from the experimental ones, we still have 25% of systematic error and 10% random error in *B*-integral evaluation. The parameters of the regenerative amplifier are shown in Table 67.I.

A polished, 400- μ m-thick, GaAs plate was inserted before the compression gratings to cancel the accumulated *B*-integral. To have a negative n_2 from the direct-beam semiconductor without strong single-photon absorption, the gap energy should be³

$$\hbar\omega < E_g < 1.42 \ \hbar\omega, \tag{1}$$

where $\hbar\omega$ is the incident photon energy. The region of $x = \hbar\omega/E_g$: 0.5 < x < 0.7 can, in principle, be used for cancellation, but in that case the bound electronic contribution and the generated free-carrier contribution will have opposite signs, and the free-carrier contribution will be greatly reduced due to reduction of two-photon absorption coefficient that scales as⁸ $\beta_2 \propto x^{-6}(2x-1)^{3/2}$. The nonlinear index of refraction of the GaAs plate estimated for 1.053 μ m is -4.2×10^{-13} cm²/W using an experimental value of -3.3×10^{-10} esu obtained for 1.064 μ m⁹ and scaling laws for $n_2(\hbar\omega/E_g)$.³

The power incident on the GaAs wafer was adjusted to cancel the accumulated *B*-integral and remove the temporal distortion of the compressed pulse (see Fig. 67.30). The 1.65-ps pulse generated with low accumulated *B*-integral (~0.5) is shown on trace (a). The pulse with time-bandwidth

Active element	<i>Q</i> -98 Nd:phosphate glass, 6%; $L = 115$ -mm athermal rod, $\phi = 7$ mm; $n_2 = 3.5 \cdot 10^{-16}$ cm ² /W
Resonator	Stable $g \approx 1/2$, plano-concave, 100% end cavity dumped; 50% output flat; round-trip time 11.8 ns
Switch-in Q-switch Switch-out	external $\lambda/4$ and step $\lambda/4$ voltage to Pockels cell external
Saturation fluence ⁷	4.7 J/cm ²
Round-trip, net small-signal gain	1.23
Seed fluence (J_{in}/J_{sat})	2 • 10 ⁻⁹
Pump fluence (J_{sto}/J_{sat})	1.035
Peak fluence (without self-focusing)	0.146
Peak pass	86
Round-trip loss factor	0.437
Net gain Total gain	67 dB 363 dB
Waist on output coupler (w_0)	750 μ m at low intensities and 500±50 μ m at $B \sim \pi/2$

Table 67.I: Parameters of Regenerative Amplifier



Figure 67.30

Experimentally measured autocorrelation of pulses compressed with (a) $B \approx 0.5$; (b) $B \approx \pi/2$; and (c) $B \approx \pi/2$, accumulated in glass and canceled in the GaAs plate before compression.

product of 0.44±0.04 is generated by switching out the regenerative amplifier pulse 6 to 7 pulses before the peak of the Q-switched envelope. Pulses switched out at the peak of the Q-switched envelope have an accumulated B-integral of 1.6 (~ $\pi/2$) and a compressed pulse duration of 2.0±0.1 ps [as shown in trace (b)]. They show significant non-Gaussian structure as expected.^{1,2} Pulses compressed after passing through the GaAs wafer are shown in trace (c). In this case, the pulse duration reduced to the original 1.65 ± 0.1 ps with the structure significantly reduced. The wings at 5% of peak intensity level are due to higher-order phase aberrations in our CPA laser utilizing fiber¹ and not due to the regenerative amplifier.

Figure 67.31 shows the compressed pulse duration (FWHM) as a function of cumulative *B*-integral. The solid circles show that the pulse duration increases in the absence of cancellation, while the diamonds show that with cancellation, the pulse duration remains constant up to a *B*-integral of approximately $\pi/2$. The pulses in Fig. 67.31 have a slightly longer duration than those in Fig. 67.30 but are also transform limited. The graph from Fig. 67.31 with uncompensated pulses has a distinctive knee at *B* between 1 and 1.5 as expected.^{1,2} A *B*-integral of 1 corresponds to an output intensity of 190 MW/cm². Internal losses for $B \approx \pi/2$ were near 10%.

The *B*-integral accumulated in the semiconductor can be found by solving the coupled intensity and free-generated carrier-density equations:⁸

$$\frac{dI(r,t,z)}{dz} = -\alpha I - \beta_2 I^2 - \sigma_{\rm ex} NI, \qquad (2a)$$



Figure 67.31

Pulse duration of switch-out pulses versus accumulated *B*-integral in the regenerative amplifier without subsequent cancellation in the GaAs plate (circles) and with subsequent cancellation in the GaAs plate (diamonds). The *B*-integral (*x* axis) has 25% systematic and 10% random error.

$$\frac{dN(r,t,z)}{dt} = -\frac{N(r,t,z)}{\tau_{\rm rel}} + \frac{\beta_2 I^2(r,t,z)}{2\hbar\omega},$$
 (2b)

$$\frac{dB(r,t,z)}{dz} = \frac{\omega}{c} (n_2 I - \sigma_r N).$$
(2c)

Here $\alpha = 2.3 \text{ cm}^{-1}$ and $\beta_2 = 20 \text{ cm/GW}$ are the linear and twophoton absorption coefficients; σ_{ex} is the exited carrierabsorption cross section; τ_{rel} is the characteristic relaxation time of exited carriers; n_2 is the nonlinear index of refraction caused by bound electrons; and σ_r is the change in index of refraction per unit of photoexited charge-carrier density, N:⁹

$$\sigma_r = \frac{2\pi \hbar^2 e^2 E_p}{n_0 E_g^3 x^2 (1 - x^2)}.$$
(3)

The exited carrier absorption term $-\sigma_{ex}NI$ can be neglected up to intensities⁸ $I_{cr} \approx 2\hbar\omega/[\sigma_{ex}(1-R)\tau_{FWHM}]$, which is ~1 GW/cm² where R = 0.3 is the reflectivity of the uncoated crystal and $\sigma_{ex} = 5 \times 10^{-18}$ cm².

The *B*-integral at the exit plane of the wafer, z = L, can be written in two terms. The fast term is caused by bound electrons and is

$$B_{\text{bnd}}(r,t) = \frac{2\pi n_2}{\lambda_{\text{vac}}\beta_2} \bullet \ln\left[1 + \beta_2 I_0(r,t)(1-R)L_{\text{eff}}\right], \quad (4)$$

where $L_{\text{eff}} = (1 - e^{-\alpha L})/\alpha$. The thickness of the crystal *L* was assumed to be much smaller than the confocal beam parameter. The dependence of $B_{\text{bnd}}(r,t)$ on $I_0(r,t)$ allows effective cancellation of the *B*-integral, when $\beta_2 I_0(1-R)L_{\text{eff}} \ll 1$ (0.15 for our case).

Free carriers lead to the slow term,

$$B_{\rm ex}(r,t) = -\frac{2\pi}{\lambda_{\rm vac}} \frac{\sigma_r \beta_2}{2\hbar\omega}$$
$$\int_0^L dz e^{-t/\tau_{\rm rel}} \left[\int_{-\infty}^t e^{-t'/\tau_{\rm rel}} I^2(r,z,t') dt' \right].$$
(5)

The relaxation time τ_{rel} is estimated to be ~100 ps — about one third of the laser-pulse duration.¹⁰ The free-carrier nonlinearity $\sigma_r N$ is proportional to a temporal integral of I^2 and cannot compensate for the accumulated *B*-integral in glass for long relaxation times $\tau_{rel} \ge \tau_p$. Moreover, for intensities higher than ~20 MW/cm² this term becomes comparable to B_{bnd} and exceeds B_{bnd} by more than an order of magnitude for $I_{inc} \approx 1 \text{ GW/cm}^2$ with laser-pulse duration $\tau_{FWHM} \approx \tau_{rel}$. However, for a relaxation time of one third of the pulse width, the contribution from the two terms is comparable at sub GW/cm² intensities. For short relaxation times ($\tau_{rel} \ll \tau_p$), B_{ex} follows the intensity dependence as well.

The *B*-integral of the pulses incident onto the GaAs crystal was taken as $B_{\text{glass}}(r,t) = B_{\text{glass}} \cdot I_0(r,t)/I_0(0,0)$, where B_{glass} is the peak *B*-integral accumulated in the regenerative amplifier. The total accumulated integral $B_{\text{tot}}(r,t)$ after the GaAs crystal,

$$B_{\rm tot}(r,t) = B_{\rm glass}(r,t) + B_{\rm bnd}(r,t) + B_{\rm ex}(r,t),$$
 (6)

can be reduced to values allowing wingless compression of the pulse for appropriate beam and crystal parameters. Figure 67.32 shows a calculation of $B_{tot}(0,t)$ and $B_{tot}(r,0)$ for a laser pulse that is Gaussian in time and space,

$$I_0(r,t) = I_0 \exp\left[-2(r/w_0)^2 - 4\ln 2(t/\tau_{\rm FWHM})^2\right]$$

with $I_0 = 285$ MW/cm² and $\sigma_r = 5.4 \times 10^{-21}$ cm³ (calculated

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Theoretically estimated residual phase after accumulation of the $B \approx \pi/2$ in absorptionless material (dashed line) (pure positive n_2) and cancellation in GaAs (solid line) with $\alpha = 2.3 \text{ cm}^{-1}$, $\beta_2 = 20 \text{ cm/GW}$, $n_2 = -4.2 \times 10^{-13} \text{ cm}^2/\text{W}$, $\sigma_r = 5.4 \times 10^{-21} \text{ cm}^3$, $L = 400 \ \mu\text{m}$, $I_{\text{inc},0} = 285 \text{ MW/cm}^2$, R = 0.3 (n = 3.47), with duration of stretched pulse $\tau_{\text{FWHM}} = 300 \text{ ps}$ and $\tau_{\text{rel}} = \tau_{\text{FWHM}}/3$: (a) radial profile at the temporal peak of the pulse, (b) temporal profile though the axis of the beam.

for $E_g = 1.4 \text{ eV}$ and $\hbar \omega = 1.177 \text{ eV}$).⁹ The intensity was chosen so that $-B_{\text{bnd}}(0,0) - B_{\text{ex}}(0,0) = B_{\text{glass}}(0,0) = \pi/2$. The cancellation reduced the *B*-integral to a temporal maximum of 0.5 and 0.3 radially at the temporal peak of the pulse. The computation shows that for a slightly longer crystal and smaller intensities the *B*-integral can be reduced by another factor of 2. The optimal choice of low-temperature (LT)-grown semiconductors with special dopants and/or longer pulses can reduce the $\tau_{\text{rel}}/\tau_{\text{laser}}$ ratio and allow reduction of the *B*-integral to negligible values. The estimated dispersion (wavelength dependence) of α , β_2 , or n_2 is negligible for a 10-Å spectrum.

AlGaAs with $E_g = 1.57$ eV has the same properties for $\lambda \approx 850 \text{ nm} - \alpha = 1 \text{ cm}^{-1}$, $n_2 \approx -5 \times 10^{-13} \text{ cm}^2/\text{W}$ (Ref. 11), $\sigma_r \approx 5 \times 10^{-21} \text{ cm}^3$, and $\beta_2 = 10 \text{ cm/GW}$ — as GaAs at 1.053 μ m. However, for $x \approx 0.92$ one should take into account the wavelength dependence (i.e., dispersion) of these parameters for efficient compensation of the *B*-integral.

The spatial *B*-integral accumulated in high-power lasers can be precompensated by placing a semiconductor wafer somewhere upstream in the laser path. Aside from coupling (fresnel) losses, which can be removed by an appropriate AR



coating, linear and TPA losses should be small enough for sufficient *B*-integral accumulation to justify placing the semiconductor for cancellation. The conditions $\alpha L \ll 1$, $\beta_2 IL_{eff}$ < 1, and $B = (2\pi/\lambda)\Delta nL_{eff}$ are the same as those used in all optical-switching inequalities.¹² These inequalities $(n_2/\beta_2\lambda > B/2\pi, \Delta n/\alpha\lambda > B/2\pi)$ are satisfied for *B* as large as π due to fairly large nonlinearities of GaAs for a fixed wavelength. For our case the bound electronic contribution is $n_2/\beta_2\lambda = 0.2$, and free-carrier index change is comparable to that caused by bound electrons.

Thus, the same semiconductor technique can be used to cancel the radially dependent *B*-integral acquired due to a nonuniform intensity profile in a long-pulse, high-power laser system. The wafer can be used as a pre- or post-compensator, depending on experimental conditions. Because the accumulated positive *B*-integral in the amplifier and the negative *B*-integral in the wafer are both proportional to the intensity, input intensity fluctuations do not affect the quality of cancellation.

The simulation in Fig. 67.33 shows cancellation of spatial and temporal *B*-integral with a GaAs wafer. A supergaussian in space (n = 8) and Gaussian in time ($\tau_{FWHM} = 1$ ns) pulse

Figure 67.33

Theoretically estimated residual phase after the propagation of a 1-ns pulse with an incident peak *B*-integral of unity before (dashed line) and after (solid line) GaAs plate with $\alpha = 1 \text{ cm}^{-1}$, L = 2 mm, $I_{\text{inc},0} = 90 \text{ MW/cm}^2$, $\tau_{\text{rel}} = \tau_{\text{FWHM}}/10$: (a) radial profile at the temporal peak of the pulse; (b) temporal profile though the axis of the beam. with peak intensity incident onto GaAs wafer I_0 (0,0) = 90 MW/cm² has a pre-accumulated *B*-integral of 1. This pulse is close to ~3 times the expanded LARA output beam.¹³ All parameters of the GaAs wafer were the same as above except the thickness, which was 2 mm, and the linear absorption coefficient $\alpha = 1 \text{ cm}^{-1}$. The phase accumulated after the cancellation is more than five times smaller than the initial phase and represents reduction of the phase error from $\lambda/6$ to $\lambda/30$. A 10% incident-energy fluctuation results in variation of residual phase by $|\delta B_{\text{max}}| \approx 0.08$.

In conclusion we have demonstrated for the first time that the nonlinear phase can be canceled in CPA lasers by using a semiconductor with negative nonlinear index of refraction.

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