Nonlinear dynamics inside femtosecond enhancement cavities

K.D. Moll, R. Jason Jones, and Jun Ye

JILA, National Institute of Standards and Technology and University of Colorado, Boulder, CO 80309-0440

mollk@jilau1.colorado.edu

http://jilawww.colorado.edu/YeLabs/

Abstract: We have investigated the effect of intracavity nonlinear dynamics arising from enhanced peak powers of femtosecond pulses inside broad-bandwidth, dispersion-controlled, high-finesse optical cavities. We find that for $\chi^{(3)}$ nonlinearities, when a train of femtosecond pulses are maximally coupled into a cavity by active stabilization of its frequency comb to the corresponding linear resonances of a cavity, enhancement ceases when the peak nonlinear phase shift is sufficient to shift the cavity resonance frequencies by more than a cavity linewidth. In addition, we study and account for the complex spectral dynamics that result from chirping the input pulse and show excellent qualitative agreement with experimental results.

OCIS codes: (190.7110) Ultrafast nonlinear optics, (320.7110) Ultrafast nonlinear optics, (230.5750) Resonators

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1. Introduction

Investigations into many nonlinear optical interactions require the use of amplified femtosecond laser systems. In addition to a potential increase in amplitude noise and pulse timing jitter, a serious limitation of conventional amplifier systems is the drastic reduction in the pulse repetition rate. This not only reduces the average power of the system and prolongs the data acquisition time for many experiments, but also limits the ability to actively stabilize the laser parameters against fluctuations (i.e. noise processes, such as amplitude fluctuations, occurring faster than half the repetition time become totally stochastic). The higher powers inside a mode-locked laser cavity can be utilized, such as for second harmonic generation [1], but this has limited use due to the dynamics of the particular mode-locking process and the need to keep the intracavity intensity in a range required for stable mode-locking.

The use of passive optical cavities external to a laser has been an effective approach to enhance laser power for nonlinear optical interactions. Although commonly used with CW lasers for efficient optical frequency conversion [2–6], pulse generation [7], or increasing the interaction length for precision spectroscopy [8–10], enhancement cavities have found limited use with sub-100 fs pulses due to many technical difficulties. For instance, coupling the entire spectrum of a mode-locked pulse train to an external high-finesse cavity requires high reflectivity mirrors with sufficient dispersion control across a broad bandwidth defined by the short laser pulse [11,12]. Recently, however, with improving mirror-coating technology, femtosecond laser stabilization techniques [13, 14], and a more complete understanding of the underlying "femtosecond comb" associated with a modelocked laser [15], sub-50 fs pulses can be efficiently coupled and "stacked" inside high-finesse (> 1000) cavities, leading to significant enhancement of the stored femtosecond pulse energy. Unlike traditional active amplifiers, the laser's original high repetition rate is maintained inside the cavity, while the stable cavity additionally provides temporal and spatial-mode filtering. This provides an environment suitable for precise measurements of nonlinear light-matter interactions at laser intensities not previously achievable without active amplification [16]. For example, in a recent experiment [17], significant spectral distortion was observed when the cavity contained a fused-silica acousto-optic modulator (AOM) at an intracavity focus that was used to dump the pulse from the cavity. The strong nonlinear response of the fused silica also limited the ultimate peak pulse intensities that were achieved in the AOM. An earlier theoretical study of nonlinear Fabry-Perot cavities predicted that significant pulse shaping can occur with ultrashort pulses [18]. Considering the potential use of the femtosecond enhancement cavity for future extreme nonlinear optics studies, it is essential that we develop a complete understanding of the intracavity nonlinear dynamics and identify the key mechanisms responsible for the experimental observations. In this article, we

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present a detailed investigation of the effect of intracavity nonlinearities on femtosecond enhancement cavities.

2. Theoretical model

As a model, we have numerically investigated a cavity similar to that studied experimentally [17], as depicted in Fig. 1. To simulate the physical system, we have assumed that the cavity has a net zero group-delay dispersion (GDD) around 790 nm and a residual third-order dispersion of ~ 250 fs^3. In addition, we have included in our model a nonlinear medium equivalent to a 3.8-mm fused-silica AOM ($n_2 = 3.8 \times 10^{-16}$cm^2/W) as well as a small loss term (0.5%) which accounts for the experimentally present absorption and scattering losses from the mirrors and small residual reflections from the nonlinear medium. The cavity length is such that the round-trip group delay corresponds to a 100-MHz repetition rate.

In the time domain, we model a pulse train incident on the input coupler with a periodic repetition frequency of f_{rep}, and the relative phase between pulses is fixed. These two variables control the two degrees of freedom of the femtosecond comb in the frequency domain (f_{rep} controls the comb mode spacing, and the phase between pulses determines the carrier-envelope offset frequency, f_o). These two degrees of freedom of the comb allow us to adjust the coupling efficiency of the pulse train to the cavity by aligning the femtosecond comb modes to the cavity resonances. For ultrashort pulses, though, net cavity GDD plays the dominant role in limiting the coupling efficiency. For studying cavity effects, a convenient way to represent the net cavity GDD is to compute the free spectral range (FSR) of neighboring cavity resonances as a function of wavelength. For the cavity in Fig. 1(a), the corresponding FSR is shown in Fig. 1(b), where the zero GDD of the cavity occurs at the maximum of the FSR. Because the FSR is not uniform across the relevant pulse spectrum, it can be seen that for high finesse cavities with narrow-

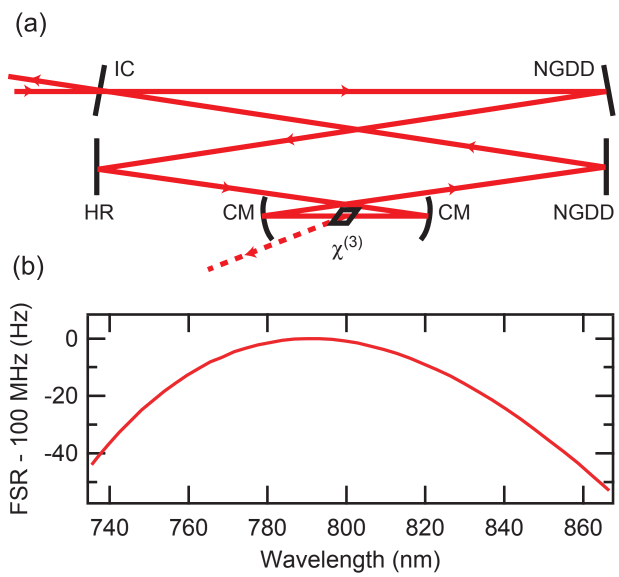

Fig. 1. (a) Schematic of the enhancement cavity being modeled. The cavity is composed of a 0.9% input coupler (IC), two negative dispersion mirrors (NGDD), a high reflector (HR), and two curved mirrors (CM) to focus the beam inside the nonlinear medium $\left(\chi^{(3)}\right)$. (b) Numerically calculated free spectral range of the cavity versus wavelength.

linewidth resonances, a femtosecond comb (which has a constant separation, f_{rep}, between comb components) will be spectrally filtered due to misalignment between the comb modes and the cavity resonances. This aspect of dispersive cavities when probed by femtosecond combs can be utilized to provide precise characterization of the cavity dispersion [19].

Since the cavity in Ref. 17 was typically under vacuum, we have assumed linear propagation except for the nonlinearities resulting from the $\chi^{(3)}$ response of the nonlinear material, specifically self-phase modulation (SPM) and self-steepening [20]. We numerically modeled the intracavity pulse by using a split-step method in which the nonlinear interaction (equivalent to propagating through a 3.8-mm fused silica AOM) is calculated in the time domain while the linear-dispersion effects are computed in the frequency domain where we model a subset of the frequencies that compose the underlying femtosecond comb of the pulse train. The spatial dynamics of the simulation were neglected; in order to model the nonlinear effects, the intensity is calculated using the beam radius at the focus inside the AOM.

3. Results

The first property that we investigated was the intensity dependence of the pulse-energy enhancement. In a cavity with a completely linear response, maximum enhancement is achieved when the pulse spectrum is centered around the zero GDD of the cavity, the laser repetition frequency equals the local FSR of the cavity, and f_o is adjusted such that the average comb frequency matches that of the corresponding cavity modes such that successive pulses constructively interfere at the input coupler. In order to isolate the effect of SPM on cavity enhancement, we simulated a pulse train composed of 45-fs pulses which have been chirped to 1 ps for which self-steepening is negligible. A series of simulations were conducted in which the input-pulse energy was increased, and the steady-state energy enhancement that was achieved was recorded. The results are presented in Fig. 2 (squares). A linear analysis of the cavity shows

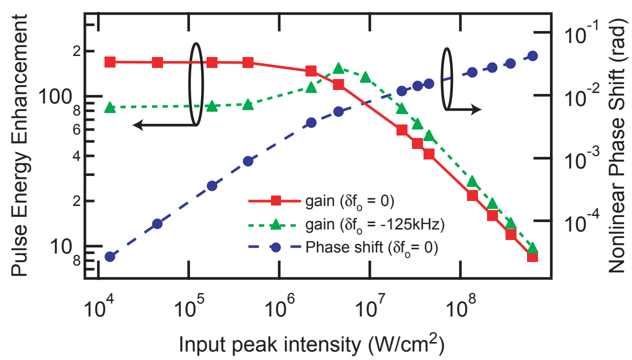

Fig. 2. Cavity pulse-energy enhancement as a function of input intensity and the associated nonlinear phase shift. Increased amplification can occur at higher input peak intensities if the comb is detuned from the linear cavity resonances.

that an enhancement of ~ 180 is expected for a CW laser locked to a particular cavity resonance mode; however, we only see an enhancement of ~ 170 when low-intensity femtosecond pulses are coupled to the cavity. This reduced enhancement results from the spectral filtering due to the nonuniform spacing between the cavity resonances arising from the residual third-order dispersion. As the intensity increases, we observe that the enhancement factor starts to rapidly decrease from the linear-cavity response. If we calculate the nonlinear-phase shift experienced by the pulse after propagating through the AOM, we observe that when the energy enhancement has dropped by a factor of 2, the single-pass nonlinear phase-shift is approximately 8 mrad. From the linear analysis of the cavity, we determine that the cavity resonance has a half-width half-maximum of ~ 112 kHz. Comparing this to the FSR of 100 MHz, this means that a phase-shift of $2\pi(112\text{kHz}/100\text{MHz}) \sim 7$ mrad in the cavity will shift the resonance by half its linewidth. Hence, we find that the degradation of the cavity enhancement is due to the dynamic change of the effective cavity resonance positions as the intra-cavity pulse builds up. It is interesting to note, however, that even though the pulse-amplification factor is decreasing as the input pulse-energy increases, the total intracavity energy (as can be inferred from the nonlinear phase shift) continues to increase, at least over the range of input-pulse energies that we studied.

In order to obtain higher energy enhancement at high intensities, we next considered the effect of coupling a pulse train with its comb structure slightly detuned from the linear cavity resonances. Since the nonlinearities effectively shift the resonance frequencies to lower values, we down shifted f_o of the pulse train by approximately one cavity resonance half-width (-125 kHz). The results are shown as triangles in Fig. 2. As expected, we find that it is possible to obtain increased amplification at higher intensities if the pulse train is slightly detuned from the linear cavity resonances. However, there is a delicate balance in order to see an improved enhancement; if too large of a detuning is used, the intracavity pulse never reaches a high enough intensity to add a nonlinear shift to the resonance. In addition, the enhancement never achieves the value predicted from a linear analysis of the cavity because the nonlinear frequency chirp imparted on the pulse always introduces some degree of destructive interference at the input coupler which cannot be compensated.

We also simulated the situation in which the underlying frequency comb of the input pulse train shifts. This occurs, for example, in experiments where the laser is scanned through the cavity resonance, when feedback is applied to the laser to keep the laser and enhancement cavity locked, or when random noise perturbs the laser. As an example, we modeled the limiting case in which a single shift in the frequency comb occurs, for example from a random perturbation to the laser cavity. An intracavity pulse associated with a specific f_o is built-up to steady state. We then simulate the perturbation by turning off the original driving pulse train, and a new pulse train with a shifted f_o becomes incident on the cavity. Inside the cavity, we follow the dynamics of two pulses: the original pulse which decays within the cavity lifetime and the second pulse which grows as it is driven by the shifted pulse train. Just as an intense intracavity pulse can shift the cavity resonances it experiences, it can also shift the cavity resonance that a second weaker pulse experiences through cross-phase modulation (XPM). Hence, because of XPM, a highly detuned pulse train can efficiently couple to the cavity. As a specific example, we considered a chirped 1-ps input pulse with a peak intensity of $4.5 \times 10^{7}\text{W/cm}^2$ coupled to the cavity. If a pulse train with a detuning $\delta f_o = -550$ kHz relative to the cavity resonance is directly coupled to the cavity, an enhancement of ~ 9 is observed (dotted line in Fig. 3). However, if first a pulse train of $\delta f_o = -300$ kHz is coupled to the cavity and then, after reaching steady state, a second pulse train of $\delta f_o = -550$ kHz is coupled to the cavity, the highly detuned pulse reaches a maximum enhancement of ~ 75. On a time scale much longer than the cavity lifetime, though, the pulse gradually decays to the value achieved by directly coupling the pulse train. This effect of the dynamic “memory” of the cavity resonance is analogous to experiments with CW lasers in which a cavity can “self-lock” to the laser frequency due to thermal changes in the intracavity medium [21]. However, because the long-decay time in our model is due to an electronic nonlinearity, these transient-behavior features can occur on a much faster time-scale and can play an important role in the presence of noise mechanisms. Thus, we have identified three relevant time scales when performing experiments on nonlinear cavities. The shortest time scale is governed by the natural lifetime of the cavity and represents the time for an intracavity pulse to decay if it is no longer driven by an incident pulse train. The intermediate time scale is dominated by the transient behavior of the nonlinear cavity. As can be seen in Fig. 3, this can be

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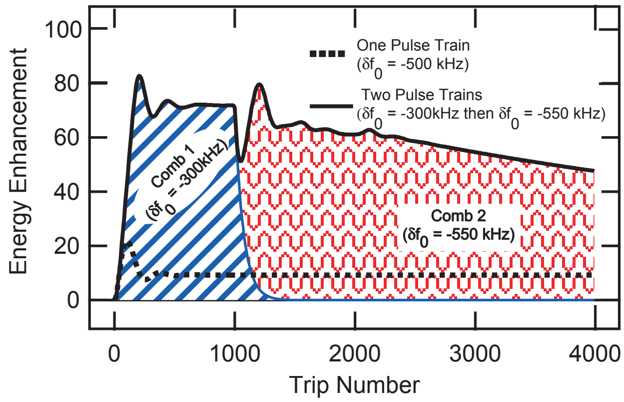

Fig. 3. Cavity buildup dynamics. A detuned comb (Comb 2) can temporarily grow to higher amplifications if another pulse (Comb 1) is already present in the cavity when compared to the absence of the first pulse (dotted line).

considerably longer than the lifetime of the cavity. For a linearly-responding cavity, a change of the frequency comb requires a time on the order of the cavity lifetime to buildup to steady state. In a nonlinear cavity, though, the long transient behavior severely limits how quickly the steady-state behavior (e.g., the cavity linewidth) can be measured. The third relevant time scale corresponds to the time in which the feedback system can make corrections to the laser cavity in order to keep the laser optimally coupled to the nonlinear cavity. Typically, piezo-actuated mirrors have a bandwidth of several tens of kilohertz. For a repetition frequency of 100 MHz, this implies that the feedback system cannot react to changes until $\sim 10^4$ pulses are incident on the cavity.

The results presented so far have been obtained from chirped input pulses such that a higher enhancement factor is achieved compared to their transform limited equivalents. However, the higher energies obtained by using chirped input pulses have come at the cost of increased spectral distortion at the output. Even though the center wavelength of the pulse is at the zero-GDD of the cavity, there is still a strong net third-order dispersion term. In combination with SPM, very complicated spectral features develop. In Fig. 4, we present numerically calculated spectra of different pulse profiles with the same initial spectral intensity distributions: a 45-fs transform limited pulse with a peak intensity of 10^9W/cm^2, the same pulse chirped to 1 ps by a normal dispersion material (+chirp), the same pulse chirped to 1 ps by an anomalous dispersion material (-chirp), and a 45-fs transform limited pulse with the same peak intensity as the 1-ps chirped pulses. Additionally, we present an experimental spectrum which resulted from input conditions similar to (c). As can be seen, the simulations reproduce the complicated fringe pattern observed in the experiment quite well.

As expected, when using pulses of equal input energy, the highest intracavity pulse energies are obtained with chirped input pulses. This indicates that if the enhancement cavity is used as an amplifier as done in Ref. 17, then it is advantageous to stretch the input pulse and compress it after it is dumped from the cavity. However, when intracavity peak intensities are important, using transform limited pulses leads to the highest intensities when compared to a chirped pulse of equal input energy. This is because the increased enhancement of the chirped pulse cannot compensate for its significantly reduced input peak intensity required to achieve the high amplification.

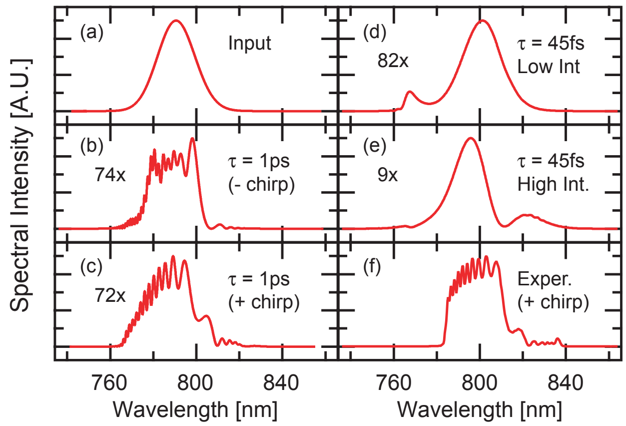

Fig. 4. Effect of chirp on output spectrum. All simulated pulses start with the same input spectrum (a). Two transform limited pulses are shown in which the peak intensity (d) and the pulse energy (e) are the same as the chirped pulses (b) and (c). The fringe pattern in the output spectrum from experimental observations (f) show excellent qualitative agreement with equivalent simulated conditions (c). The observed energy enhancement is also indicated in each panel.

4. Conclusions

In conclusion we have presented a detailed study of nonlinear dynamics inside a passive, femtosecond enhancement cavity where peak pulse energies can reach interestingly high levels. We have found that the pulse-energy enhancement is limited by a dynamic shifting of the cavity resonances brought about by the nonlinear phase shift imparted onto the intracavity pulse. We have identified that the transient behavior of the buildup dynamics can persist for times much longer than the natural cavity lifetime when nonlinear elements are introduced into the enhancement cavity and that the complicated spectral features that were experimentally observed can be explained by the interaction of SPM and GDD.

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