Q-Switched Operation with Carbon-Based Saturable Absorbers in a Nd:YLF Laser

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Abstract: We have numerically studied the influence of the absorption modulation depth of carbon-based saturable absorbers (graphene and carbon nanotubes (CNTs)) on the Q-switched regime of a diode-pumped Nd:YLF laser. A short-length cavity was used with an end mirror on which CNTs or mono- or bi-layer graphene were deposited, forming a saturable absorber mirror (SAM). Using a standard model, the generated energy per pulse was calculated, as well as the pulse duration and repetition rate. The results show that absorbers with higher modulation depths, i.e., graphene, deliver higher energy pulses at lower repetition rates. However, the pulse duration did not have a monotonic behavior and reaches a minimum for a given low value of the modulation depth typical of CNTs.

Keywords: diode-pumped lasers; Q-switched Nd:YLF laser; mode-locked Nd:YLF; graphene saturable absorber; carbon nanotube saturable absorber

1. Introduction

The passive Q-switched and mode locking operation of lasers using saturable absorbers to produce intense ultrashort pulses is a well-settled and understood technique [1], both from the theoretical and the experimental point of view. Among the systems that can act as saturable absorbers, we find dyes, SESAMs (semiconductor saturable absorber mirrors) [1] and, more recently, carbon nanotubes (CNTs)
and graphene. One advantage of the last two in relation to dyes and SESAMS is that they are not wavelength specific, so you can use the same absorber for a large variety of lasers. Carbon nanotubes have already been used to produce Q-switched [2] and mode-locked [3–6] laser pulses in bulk solid-state lasers. CNTs have been also used in fiber lasers using single-walled CNTs for the high-order harmonic mode locking [7] or single or double-walled CNTs for passive mode locking of erbium-doped fibers [8].

More recently, graphene has been employed for both temporal regimes, too, both in monolayer [9–11] and multilayer forms [12–14], in bulk solid-state lasers. While in fiber lasers, multilayer graphene [15] or graphene nanoparticles [16] have been used to produce mode locking in erbium-doped fiber lasers.

Both temporal regimes are not always easy to isolate experimentally from each other. Q-switch-modulated mode locking can appear or simply Q-switch can hinder mode locking. Hence, when looking for mode locking operation in a laser using a saturable absorber to produce ultrashort laser pulses, some attention should be paid to the possibility of Q-switched operation, which can make it more difficult to find pure mode locking. For example, the titanium:sapphire laser, which has optimal performance in mode locking, can be affected by the appearance of Q-switch in certain cavity designs [17–19]. In this paper, we want to evaluate the convenience of using carbon nanotubes or graphene in diode-pumped lasers, evaluating their performance as saturable absorbers for Q-switched operation. We will use as an example Nd:YLF laser. The choice of this type of laser is due, on one side, to the fact that it can be transversally pumped (side-pumped lasers have the advantage of a higher average output power) and, on the other side, because we have obtained good agreement between experimental and theoretical results in this type of laser in Q-switched operation using a monolayer graphene SAM [10].

2. The Model

In previous works, a model given by Haus and Spühler [20,21] has given good results when comparing experimental with simulated Q-switched operation using a monolayer G-SAM (graphene saturable absorber mirror) in a Nd:YLF laser [10] or predicting Q-switched operation in a short-cavity titanium:sapphire laser [19]. In this model, the intracavity power $P$, the gain $g$ and the density of population inversion $q$ can be calculated with the following set of equations [20,21]:

$$\frac{dP}{dt} = \frac{1}{t_r} \left( g - l - q \right) P \quad (1)$$

$$\frac{dg}{dt} = -\frac{1}{t_L} \left( (g - g_0) + gP/P_L \right) \quad (2)$$

$$\frac{dq}{dt} = -\frac{1}{t_A} \left( (q - q_0) + qP/P_A \right) \quad (3)$$

where $t_r$ is the cavity round-trip time; $t_L$, $t_A$ the relaxation times of the laser medium and absorber, respectively; $l$ the linear cavity losses; and $P_L$, $P_A$ the saturation power of the laser medium and absorber. The meaning of $g_0$ and $q_0$ is explained below.

To compare the results obtained with graphene or carbon nanotubes as saturable absorbers deposited on a high-reflecting mirror (G-SAM or CNT-SAM), we have chosen a simple linear cavity (Figure 1), which employs of a Nd:YLF laser rod. Nd:YLF is a birefringent crystal, which has a thermal lens effect and can emit laser radiation at 1047 nm and 1053 nm with crossed polarizations, 1047 nm being the strongest line [22,23]. In our calculations, we used the $\lambda = 1047$ nm emission line, and the dimensions of the rod had typical values of $2a = 5$ mm diameter, $L_r = 5$ cm length, refractive index 1.45.
The rod was placed in a plane-mirror resonator (SAM and output mirror (OM)) with a short physical length of 100 mm, corresponding to an optical length of \( L = 102.25 \) mm. Short-length cavities give shorter pulse durations, higher output powers and higher repetition rates in the Q-Switched regime [10,19] than longer ones.

![Figure 1. Cavity design for a transversally-pumped Nd:YLF laser. L: cavity length, \( L_r \): laser rod length, L: focusing lens, OM: output mirror, SAM: saturable absorber mirror.](image)

The SAM was considered to have a reflectivity \( R_{\text{SAM}} = 0.998 \) without the saturable absorber on it and the output mirror \( R_{\text{OM}} = 0.98 \). A lens L (f = 10 mm) was used to focus the laser beam onto the SAM to saturate the graphene or the CNTs. With this configuration, a stability analysis can be done where the thermal effects in the laser rod were introduced with a thermal lens with a focal length f = 200 mm. This analysis reveals that the TEM\(_{00}\) is practically collimated in the zone rod-OM and is strongly focused in the SAM. However, the geometry of the cavity has a high Fresnel number (i.e., \( N_F = a^2/(L\lambda) \sim 58 \), for \( a = 2.5 \) mm, \( L = 102.25 \) mm, \( \lambda = 1047 \) nm), and we can assume transversal pumping, so it is more realistic to think of a multimode operation and estimate larger beam sizes at the rod and at the SAM than the values given at the stability analysis, which is done for the TEM\(_{00}\). Hence, we have done the calculations estimating \( w_r \sim 800 \) \( \mu \)m at the rod and \( w_{\text{SAM}} \sim 300 \) \( \mu \)m at the SAM. This estimation is based on the fact that these orders of magnitude gave good results when comparing experimental and theoretical results in a transversally-pumped Nd:YLF [10].

Internal cavity losses were taken as \( \alpha_i L_r = 0.1 \), which is a value within the typical order of magnitude for solid-state lasers, where \( \alpha_i \) is the internal absorbance and \( L_r \) is the length of the laser rod. With these values, the linear cavity losses can be calculated as \( l = 2(\alpha_i L_r - \ln(R)) \), being the average reflectivities given by \( R = \sqrt{R_{\text{SAM}}R_{\text{OM}}} \). The quantity \( q_0 \) is the reflectivity change in the end mirror from absorbing to saturated absorber; this is the modulation depth. Carbon nanotubes have lower \( q_0 \) values than graphene. Modulation depths are typically around 1% and below [4–6], which correspond to \( q_0 = 0.01 \) and below. A single graphene layer absorbs \( \sim 2.3\% \); the absorbance of \( n \) graphene layers is \( q_0 = -2n \ln(1-0.023) \). Hence, with carbon nanotubes and mono- and multi-layer graphene, we can cover a large range in the parameter \( q_0 \). All graphene or nanotube layers were taken as uniform within the spot size of the beam at the SAM.

Other parameters, like relaxation times \( t_A \) for the absorber, saturation powers \( P_A \) and saturation fluences \( F_A \), are more similar between graphene and carbon nanotubes. \( t_A = 1.45 \) ps for graphene [9], while \( t_A \) ranges from 0.75 ps to 1 ps for carbon nanotubes [4,6]. For graphene, \( F_{\text{sat}} \) is 14.5 \( \mu \)J/cm\(^2\) [9], while \( F_{\text{sat}} \) is 10 \( \mu \)J/cm\(^2\) for carbon nanotubes [4,6]. Therefore, it seems reasonable to choose for all parameters their values in graphene and to study just the influence of \( q_0 \).
Saturation powers for the absorber or the laser medium $P_{sat} = P_A, P_L$ were calculated from the saturation fluence $F_{sat}$. We used $P_{sat} = F_{sat} \pi w^2 / \tau$, with $\tau$ the relaxation time of the laser medium ($t_L = 480 \mu s$ [22]) and of the absorber ($t_A = 1.45 \text{ ps}$ [9]), $w$ the beam waists at the laser rod ($w_r$) or at the SAM ($w_{SAM}$), which, as mentioned, were estimated considering multimode operation. The saturation fluence for Nd:YLF is given by $F_{sat} = h \nu / (2 \sigma)$, with $h$ the Planck’s constant, $\nu$ the laser emission frequency and $\sigma$ the emission cross-section ($\sigma = 1.8 \times 10^{-19} \text{ cm}^2$ for the 1047-nm line of Nd:YLF [22]). $g_0$ is the unsaturated round-trip gain, and this parameter grows with increasing pumping intensity. We gave $g_0$ values ranging from 2- to 10-times the linear losses $l$, in steps of 1.

By solving Equations (1) to (3) using a 4th order Runge–Kutta method, we have studied the Q-switched regime (pulse duration, pulse energy and repetition frequency of the Q-switch train) as a function of $g_0/l$, for different values of $g_0$. The values chosen were $g_0 = 0.046$ (two layers of graphene), 0.023 (one-layer of graphene) and 0.015, 0.01, 0.005 (representative values for CNTs) The integration time was always set long enough so that the stable regime (constant peak power) was reached. Tables 1 and 2 summarize the value of the parameters used in the calculations.

### Table 1. Cavity parameters (see the text for details).

<table>
<thead>
<tr>
<th>Laser Rod Diameter</th>
<th>Laser Rod Length</th>
<th>Refractive Index</th>
<th>Cavity Length</th>
<th>Output Mirror Reflectivity</th>
<th>SAM Reflectivity</th>
<th>Beam Waist at Laser Rod</th>
<th>Beam Waist at SAM</th>
</tr>
</thead>
<tbody>
<tr>
<td>2$a = 5$ mm</td>
<td>$L_r = 5$ cm</td>
<td>1.45</td>
<td>L = 100 mm</td>
<td>$R_{OM} = 0.98$</td>
<td>$R_{SAM} = 0.998$</td>
<td>$\omega_r = 800 \mu m$</td>
<td>$\omega_{SAM} = 300 \mu m$</td>
</tr>
</tbody>
</table>

### Table 2. Laser and saturable absorber parameters (see the text for details).

<table>
<thead>
<tr>
<th>Relaxation Times</th>
<th>Internal Cavity Losses</th>
<th>Saturation Fluences</th>
<th>Cavity Losses</th>
<th>Modulation Depth</th>
<th>Unsaturated Round-Trip Gain</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t_L = 480 \mu s$</td>
<td>$\alpha_i L_r = 0.1$</td>
<td>$F_{sat} = h \nu / (2 \sigma)$</td>
<td>$l = 2(\alpha_i L_r - \ln (R))$</td>
<td>$q_0 = 0.046, 0.023, 0.015, 0.010, 0.005$</td>
<td>$g_0 = 2l$ to 10$l$</td>
</tr>
<tr>
<td>$t_A = 1.45 \text{ ps}$</td>
<td>(\sigma = 1.8 \times 10^{-19} \text{ cm}^2)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 2 shows the pulses obtained by integration of Equations (1) to (3) in the case $q_0 = 0.015$, $g_0/l = 4$, where it can be seen how the regime is stationary only after half a millisecond approximately.

Figure 2. Q-switched regime obtained for $q_0 = 0.015$, $g_0/l = 4$.

The lower the value of $q_0$, the longer the integration time needed to reach the stationary Q-switched operation for low $q_0/l$ values (for example, 5 ms, for $q_0 = 0.005$ and $g_0/l = 2$). Figure 3 is a temporal zoom on the last pulse of this train.
Figure 3. Temporal zoom in a Q-switched pulse in the train of Figure 2a obtained for \( q_0 = 0.015, g_0/l = 4 \).

In other cases, the temporal shape does not show a sharp peak, and the absorber is bleached for a longer time, giving rise to a flat-top-like temporal pulse, like in Figure 4, obtained for \( q_0 = 0.046 \) and \( g_0/l = 4 \).

Figure 4. Temporal zoom in a Q-switched pulsed obtained for \( q_0 = 0.046, g_0/l = 4 \).

From this output, the temporal widths, the energy per pulse and the repetition rate can be deduced. Figure 5a shows the pulse durations obtained for \( q_0 \) ranging from 0.046 to 0.005. Very short pulse durations can be achieved in all cases, with very similar values with respect to \( g_0/l \). The behavior is not monotonic and changes from \( q_0 = 0.01 \) to \( q_0 = 0.005 \). This behavior in the pulse duration is studied later in closer detail. Figure 5b shows the pulse energy obtained for the same range of \( q_0 \) values. Very high energies are obtained, and the highest values are obtained for the highest \( q_0 \). Like the pulse duration, the pulse energy does not depend significantly on the pump value \( g_0/l \) and remains practically constant for increasing pumping values from \( g_0 = 2l \) to \( g_0 = 10l \), being the unsaturated round-trip gain \( g_0 \) higher for higher pumping power. Figure 5c shows the repetition frequency of the pulse train and is the magnitude, which changes the most, increasing with \( g_0/l \), and it is apparent that, unlike in the mode locking regime, the repetition rate in Q-switched operation mode is not fixed by the cavity round trip time. The lowest frequencies are obtained for the highest values of \( q_0 \). In Figure 5, the case \( q_0 = 0.046 \) for bilayer graphene and \( g_0/l = 2 \) did not reach Q-switch operation and extinguished after a couple of spikes.
Figure 5. Pulse duration, pulse energy and repetition frequency for different absorptions $g_0$ as a function of the pump parameter $g_0/l$.

The behavior of the pulse duration and pulse energy has been studied in closer detail for $g_0/l = 8$ and an absorption range for the CNTs from $g_0 = 0.005$ to $g_0 = 0.01$ in steps of 0.001, as well as for monolayer and bilayer graphene. Figure 6a shows how the pulse duration reaches a minimum for $g_0 = 0.01$, while the pulse energy grows monotonically with $q_0$ in the whole range. According to the behavior of these two magnitudes in Figure 5a,b, which have a low dependence on $g_0/l$, the same behavior is expected for other values of $g_0/l$.

Figure 6. Pulse duration and pulse energy for $g_0/l = 8$ in an absorption range $g_0 = 0.005$ to $g_0 = 0.01$ in steps of 0.001 for CNTs, $g_0 = 0.023$ for monolayer graphene and $g_0 = 0.046$ for bilayer graphene.
Although the Q-switched regime is present in the range studied, when $q_0$ is very low (for example $q_0 = 0.001$), then only relaxation oscillations appear, tending to the cw state for longer times (Figure 7).

![Figure 7. Relaxation oscillations obtained for $q_0 = 0.001$, $g_0/l = 2$.](image)

3. Conclusions

Using a standard model, we have calculated the pulse duration, pulse energy and repetition rate of the Q-switched pulse train generated in a transversally-diode-pumped short-length cavity Nd:YLF laser using a graphene or CNT saturable absorber mirror. Higher energy pulses can be obtained for high values of the modulation depth $q_0$; this is for graphene. However, pulse duration reaches a minimum for a certain low $q_0$ value; this is for CNTs. Hence, when designing the laser, the choice of the proper $q_0$ must be done with a compromise between the desired energy and duration for the pulse. We have also observed that these two magnitudes do not depend significantly on the pump $g_0/l$ for the geometry studied. The repetition rate of the train Q-switched pulses is higher for lower $q_0$ values and increases with the pump value $g_0/l$. The results also show that Q-switch can be hindered using CNTs with very low modulation depth ($\sim 0.001$), the relaxation oscillations being the temporal regime in these cases. The results have certain universality and can be expected for other types of transversally-pumped short-length cavity lasers with an active medium with a lifetime in the order of hundreds of microseconds.

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Author Contributions

José Manuel Guerra Pérez adapted and applied the model. Rosa Weigand programmed and solved the equations for the different cases and wrote the article. Margarita Sánchez Balmaseda processed the data.

Conflicts of Interest

The authors declare no conflict of interest.
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