Stark induced contribution to mode locking in cw solid-state lasers with semiconductor saturable absorber

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ABSTRACT

It is shown, that the fast saturable absorber effect due to the Stark shift of the excitonic resonance in the quantum-confined semiconductor can contribute to ultra-short pulse formation and stabilize the pulses with extremely short duration at the below band-gap excitation, i. e. in the presence of the Stokes mismatch between gain band center and excitonic line. This mechanism is strong enough to provide self-starting over the full region of the cavity stability.

Keywords: solid-state laser, mode locking, optical Stark effect, semiconductor, saturable absorber

2. INTRODUCTION

Recently, a considerable progress has been made in self-starting femtosecond lasers using semiconductor structures. This allows the pulses as short as 6.5 fs to be generated directly from the resonator. The laser systems with semiconductor absorbers combine the advantages of the Kerr-lens mode locked system, the self-starting ability and the cavity alignment insensitivity. The most striking feature of semiconductor absorbers used in the experiments is a long recovery time T (hundreds of fs) as compare with pulse duration. To explain an extremely-short pulse generation a soliton mode-locking mechanism was proposed. This mechanism involves the stabilization of the Schrödinger soliton against laser continuum (noise) due to noise decaying within the positive net-gain window, which is much longer than the pulse duration. This decaying is the result of the dispersion spreading of the noise and the difference between the noise and pulse group velocities. However, as well as semiconductor loss saturation and slow change of the refractivity, other nonlinear effects, in particular, ac Stark-effect can contribute to mode-locking and produce a strong self-amplitude modulation (see, for example). It was shown, that the intracavity intensities in the real solid-state lasers are high enough to produce an ultrafast nonlinear response due to ac Stark effect at below resonance excitation and thus has to be taken into account.

Here we present a simple model for mode-locking mechanism in the presence of the quadratic blue Stark shift of the excitonic resonance at below resonance excitation. One should note the difference with, where an external signal driven antiresonant Fabry-Perot modulator based on quantum-confined Stark-effect in multi-quantum well semiconductor structure was used to actively mode-lock a diode-pumped Nd: YLF laser. In our case the mode-locking mechanism is purely due to the pulse self-action which is a power dependent and can self-start an ultrashort pulse operation.

3. MASTER EQUATIONS

The ac Stark shift $\Delta \omega$ which is due to the influence of nonresonant transitions on excitonic resonance is proportional to the polarizibility difference between ground and excited states $\Delta \alpha$: $\Delta \omega = \Delta \alpha |E|^2 / h$, where $E$ is the field strength. The typical values of $|\Delta \alpha|$ for semiconductors are of order $10^{-19} \div 10^{-21}$ cm$^3$ (see ref.), that corresponds to the Stark shift coefficient $\zeta = 8\pi |\Delta \alpha| / (n \chi n) = 1.26 \times 10^{2} \pm 10^{3} n$ cm$^{-2}$ J$^{-1}$, where $n$ is the index of refractivity.

The evolution of the complex field envelope $a(t)$ in the laser system containing the gain medium, saturable absorber, frequency filter and dispersive element obeys nonlinear operator's equation: $a^{k+1}(t) = AGDa^k(t)$, where $k$ is the...
round trip number, $t$ is the local time. The Lorenzian gain band is described as $G = \exp\left( -\frac{g L_g}{1 + L_g t} \right)$, where

$L_g = \frac{1}{1 + i(\omega - \omega_g) t}$, $\omega$ is the pulse carrier frequency, $\omega_g$ is the gain band center, $t$ is the inverse gain bandwidth (we assumed $t = 20$ fs for the Cr: forsterite laser with modulator based on PbS quantum-dot doped glass), $g$ is the saturated gain. $D = \exp(id \frac{\phi^2}{\omega^2})$ is the second-order group velocity dispersion operator, where $d$ is the dispersion coefficient.

Neglecting the population of higher energy levels of the semiconductor and for weak exciton-exciton, exciton-phonon bound approximation, one can describe the interaction between laser field and quantum-confined absorber by the generalized two-level model. The evolution of the off-diagonal element of the density matrix $\pi$ and the population difference between ground and excited states $\eta$ obey the differential equation set:

$$\frac{d\pi}{dt} + \left( \frac{t_a^{-1} - i(\omega - \omega_a - \Delta\omega)}{\hbar} \right)\pi = i \hbar \omega \eta,$$

$$\frac{d\eta}{dt} + \frac{\eta - \eta_0}{T} = -\frac{4}{\hbar} \text{Im}(\pi^{\alpha^*}),$$

where $t_a$ is the inverse bandwidth of the absorption line, $\omega_a$ is the resonance frequency, $\pi$ is the matrix element of the interaction, $\eta_0$ is the equilibrium population difference. The Stark shift which is possible only in generalized two-level model, in quasi-monochromatic approximation, i.e., when one neglects the cross-modulation between different pulse spectral components is proportional to $\gamma |\alpha(t)|^2$. In the incoherent approximation, the combined effects of saturable absorption and the Stark-effect contribution are contained in the operator

$$A = \exp\left\{ \frac{\text{Re}(L_a)}{1 + L_a t} \exp\left[ -\frac{\text{Re}(L_a)}{U_a} \int_0^t \frac{|\alpha(t')|^2 e^{-\frac{t-t'}{T}}}{U_a} dt' \right] \right\},$$

where $L_a = \frac{1}{1 + i(\omega_1 - (\omega_2 + |\alpha(t)|^2)^2/\gamma}$, $U_a$ is the saturation energy, $\gamma$ is the saturated loss at the time $t_0$ corresponding to the pulse peak, $r$ is the unsaturable loss. Integral in $A$ accounts for the ordinary slow saturable absorption with recovery time $T$. We have assumed a Lorenzian profile for the excitonic resonance and neglected the two-photon absorption (TPA) (justified later) as well as other high-order intensity dependent effects. We normalized all times on $t\_g$, frequencies on $t\_g^{-1}$, intensity on $U_a/t\_g$.

Using the expression for $A$ one may obtain the instant intensity transmission:
where \( t \) is the local time, \( \tau = \frac{\omega a}{\gamma} \), \( \Gamma \) is the nonsaturated loss, \( \omega = \omega_t - \omega_a \), \( \chi = \frac{\gamma}{\omega_0 \Omega} \). \( \chi = 13 \) used in our calculations corresponds to absorber saturation fluency \( U_a = 390 \mu \text{J cm}^{-2} \). It is seen, that for the below resonance excitation with \( \omega < 0 \) (i.e. for the red shift of the pulse carrier frequency from the excitonic resonance) the expression of Eq. (1) describes the power-dependent loss saturation. The fast saturable absorber action results from "pushing out" of the excitonic resonance from the red-shifted pulse spectrum due to intensity-dependent blue Stark shift. The corresponding parameter of self-amplitude modulation \( \rho = \frac{\mu}{M} \left. \frac{\partial M}{\partial |a|^2} \right|_{|a|^2=0} \) is plotted in Fig. 1 (laser mode cross-section in absorber \( \mu = 30 \mu \text{m} \)) depending on the mismatch between pulse carrier frequency and excitonic resonance. One can see, that \( \rho \) is of order of \( 10^{-7} \) W\(^{-1}\) that is close to corresponding parameter typical for Kerr-lens mode locked systems\(^{10}\), but in contrast to the latter is cavity alignment insensitive.

An expansion of the laser operator equation in series in \( t \), local field energy and intensity, provided that the pulse duration is much shorter than \( T \), gives the following laser dynamical equation similar to the generalized Landau-Ginzburg equation (a similar equation for the other physical situation was solved in ref.\(^{11}\)):

\[
\frac{\partial a(k,t)}{\partial k} = \left[ c_1 + ic_2 \frac{\partial}{\partial t} + (c_3 + ic_4) \frac{\partial^2}{\partial t^2} + (c_5 + ic_6) a(k,t)^2 + \right.
\]
\[
\left. (c_7 + ic_8) + (c_9 + ic_{10}) \frac{\varepsilon^2}{2} + (c_{11} + ic_{12}) \frac{\partial}{\partial \phi} a(k,t), \right]
\]

where \( \varepsilon = \int [a(k,t')]^2 \, dt' \), \( c_1 = gJ_g^2 - gJ_a - r \), \( c_2 = 2g\Omega J_g^2 - 2\gamma \omega \rho J_a^2 \), \( c_3 = (1 - 3\Omega^2)J_g^3 - \gamma (1 - 3\omega^2 \tau^2) J_a^3 \), \( c_4 = g(\Omega^3 - 3\Omega)J_g^3 - \gamma (\omega^3 \tau^3 - 3\omega \Omega) J_a^3 \), \( c_5 = -2\gamma \rho J_g^2 \), \( c_6 = -\gamma \chi J_a^2 \), \( c_7 = \rho J_g^2 \), \( c_8 = -\rho \omega J_a^2 \), \( c_9 = -\rho J_g^3 \), \( c_{10} = \gamma \rho J_a J_g \), \( c_{11} = -\gamma (1 - \tau^2 \omega^2) J_g^3 \), \( c_{12} = 2\omega \rho \tau J_a^3 \), \( J_a = \frac{1}{1 + \omega^2 \tau^2} \), \( J_g = \frac{1}{1 + \Omega^2} \), \( \Omega = \omega_1 - \omega_g \). Here, the term \(-2\chi J_g^2 \omega \rho |a(k,t)|^2 a(k,t)\) is responsible for the fast saturable absorption.

### 4. LASER QUASI-SOLITON

The Eq. (2) has quasi-soliton solution \( a(k,t) = a_0 \text{sech}^{1/\psi} \left[ (t - k \delta) / t_p \right] e^{i \omega t} \), where \( t_p \) is the pulse duration, \( a_0 \) is the amplitude, \( \psi \) is the chirp, \( \delta \) and \( \phi \) are the time and phase delay after the full round-trip, respectively. Pulse duration and frequency shift for chirp-free solution and group velocity dispersion predicted by such a solution are presented in Fig. 2. It is seen, that the minimal pulse durations (close to the limit defined by \( t_g \)) are provided by normalized absorber-gain lines mismatch \( \left( \omega_a - \omega_g \right)/t_g \approx 0.3 \div 0.7 \). The increase of nonsaturated loss \( \Gamma \) shortens the pulse (curve 3 in compare with 1 in Fig. 2, a), that corresponds to growth of \( \rho \) (curve 2 in Fig. 1). The minimum of the pulse duration on mismatch \( \omega_a - \omega_g \) corresponds approximately to the maximum of \( \rho \) in Fig. 1. Using the absorber with smaller \( \omega_a \) increases \( t_p \) (curve 2) because the absorber operates in the regime of strong saturation. Strong saturation introduces an additional blue shift from the gain band center (curve 2 in Fig. 2, c, for the mechanism of this shift see ref.\(^{12}\)). Thus, the stronger saturation does not favor the Stark-induced mode locking. Strong self-phase modulation due to ac Stark effect, which is described by coefficient \( c_6 \) requires the negative dispersion for chirp compensation (Fig. 2, b). When the absorber band width is narrower than the band width (curve 4), the region of \( \omega_a - \omega_g \), where chirp-free pulses exist, reduces, which is explained by bad overlapping of the pulse spectrum and excitonic line. However, for \( t_p > t_g \) the generation of chirped pulses with
duration close to \( t_e \) is possible (Fig. 3). The large \( \tau \) requires positive dispersion \( d \) for pulse existence (curve 2 in compare to 1), which may be explained by change of the sign of self-phase modulation coefficient in \( \sigma_C \). However, for large \( \tau \), i.e. for the absorption line much narrower than the gain line, it would be necessary to account for the coherent nature of pulse-semiconductor interaction, which may cause new effects, for example self-induced transparency\(^\text{13}\) and may transform mode-locking dynamics essentially.

The described dependence of the pulse parameters on the frequency shift from excitonic resonance is confirmed by experimental results presented in ref.\(^5\). It was shown, that the resonance interaction with quantum-confined semiconductor structure does not favor an ultra-short pulse formation (fig. 2, a). Relatively small Stokes shift of the laser frequency from the excitonic resonance causes fs-pulse operation, while the increase of the Stokes shift brings the pulse duration into picosecond region.

### 5. ULTRA-SHORT PULSE STABILITY

In order to perform the stability analysis we calculated the net-gain \( \Sigma \) behind the pulse tale. \( \Sigma \) is the sum of saturated gain in active medium, loss in absorber and unsaturable loss. Evidently, the pulse would be unstable, if \( \Sigma > 0 \) for some frequency \( \omega_p \) within the noise spectrum. Fig. 4 shows \( \Sigma \) calculated for five selected values of \( \omega_p \) (curves 1 - 5 in Fig. 4). It is seen that the decrease of \( \Gamma \) (Fig. 4, a in compare with c), or, equally, decrease of \( U_0 \) (Fig. 4, a, \( U_0 = 390 \mu J \) cm\(^{-2} \), in compare with \( U_0 = 240 \mu J \) cm\(^{-2} \) in Fig. 4, b) or growth of \( \tau \) (Fig. 4, d) destabilizes the chirp-free pulse, but the suitable detuning between absorber and amplifier lines \( \omega_{a} - \omega_{g} \) and suitable absorbers parameters \( \tau \) and \( U_0 \) allow for the generation of stable pulses with short duration, which is much shorter than the absorption recovery time.

To investigate the self-starting ability of the laser analyzed the stability of cw-regime. As the self-starting criterion the condition of modulational destabilization of cw operation and setting up of pulsations with the repetition rate correlated with the laser repetition rate\(^\text{14}\) was chosen. A cw-solution of Eq. (2) is \( N = N_0 \exp(ik\xi) \), where real amplitude \( N_0 \) and phase \( \xi \) satisfy the equations:

\[
\frac{\alpha_{\max} J_g}{1 + \sigma V_0 J_g} - \frac{F_{J_a}}{1 + N_0^2 J_a} = \frac{2G\chi}{\omega_0 N_0^2}, \\
\xi = \phi + (1 - \omega^2 \tau^2) \frac{2G\chi}{\omega_0 N_0^2}.
\]  

(3)
Here, all parameters have the same meaning as in Eq. (2), \( \sigma \) is the ratio of the loss saturation energy to the gain saturation energy \((\sigma=10^3)\). The cw power is normalized to the loss saturation power, \( \alpha_{\text{max}}=1 \). We investigated the stability of cw solution against complex perturbation \( \psi = \psi_0 e^{i\theta} \). The self-starting occurs when \( \text{Re}(\lambda)>0 \) and \( \text{Im}(\lambda)=\pm 1 \), \( l \) is the natural number (nondecaying oscillations set up with repetition rate which is multiply of inverse cavity round-trip). The linearization of eq. (2) and summing up with complex conjugated gives the equation for eigen values of perturbation:

\[
\lambda = gJ_g - pJ_a + \left[ e_a N_0^2 J_a - \frac{\epsilon_g \sigma N_0^2 J_g}{1 - i\omega T_c^g} \right] \left[ 1 + \Re(\lambda) - \omega c_2 - \omega^2 c_3 + \omega^4 c_4 + (c_5 - i\epsilon_c) N_0^2 \right] + \left[ e_a N_0^2 J_a - \frac{\epsilon_g \sigma N_0^2 J_g}{1 - i\omega T_c^g} \right] \left[ 1 - \Re(\lambda) + \omega c_2 + \omega^2 c_3 - \omega^4 c_4 - 2(c_5 - i\epsilon_c) N_0^2 \right],
\]

where

\[
\Re(\lambda) = \frac{\frac{\epsilon_a N_0^2 J_a}{1 - i\omega T_c^a} - \frac{\epsilon_g \sigma N_0^2 J_g}{1 - \omega T_c^g} + (c_5 - i\epsilon_c) N_0^2}{\frac{\epsilon_a N_0^2 J_a}{1 - i\omega T_c^a} - \frac{\epsilon_g \sigma N_0^2 J_g}{1 - \omega T_c^g} + (c_5 - i\epsilon_c) N_0^2},
\]

\[
T_c^a = \frac{T}{1 + N_0^2 J_a},
\]

\[
T_c^g = \frac{T}{1 + \sigma N_0^2 J_g},
\]

\( \epsilon_g = \frac{\alpha_{\text{max}}}{1 + \sigma N_0^2 J_g} \), \( \epsilon_a = \frac{\Gamma}{(1 + N_0^2 J_a)^2} \).

The dependence of \( \text{Im}(\lambda) \) versus \( \text{Re}(\lambda) \) is shown in Fig. 5 for three different mismatches between gain and loss lines. One can see, that the cw regime is unstable for all three cases, but self-mode locking is possible only for the case 1, where the condition \( \text{Im}(\lambda)=1 \) is satisfied and self-amplitude modulation coefficient \( \rho \) (see Fig. 1) is close to its maximum.

Now we will check the validity of approximation that neglected the contribution of TPA in compare to one-photon absorption made in the beginning of the article. Comparing curves 3 and 2 in Fig. 1, plotted for the situation with and without TPA, one may conclude, that in our case the TPA contribution is small and we may neglect it.

In conclusion, we have analyzed the mode-locking mechanism in cw solid-state laser with semiconductor saturable absorber in the presence of the Stark shift of the excitonic resonance. Calculations showed that it is strong enough to self-start and maintain an ultra-short pulse generation.

6. REFERENCES


Fig. 3. Chirp \( \psi \) (a) and duration of the pulse \( t_p \) (b) versus normalized group velocity dispersion. \( \tau = 1 \) (1), 30 (2), \((\omega_c - \omega_g) t_g = 0.5 \) (1), 0.2 (2); \( \chi = 13 \), \( \Gamma = 0.1 \), g - r = 0.001.
Fig. 4. Net-gain $\Sigma$ behind the pulse tail for five selected noise frequencies $(\omega_n-\omega_0)t_g = 1$ (curve 1), 0.5 (2), 0 (3), -0.5 (4), -1 (5). $\chi = 13$ (a, c, d), 8 (b), $\Gamma = 0.05$ (a, b), 0.1 (c, d), $\tau = 1$ (a, b, c), 3 (d).

Fig. 5. Complex increment $\lambda$ of cw perturbation to cw-solution. $\chi = 13$, $\Gamma = 0.1$, $\tau = 1$, $g_r = 0.01$, $\omega_{r}$ $t_r = 0.7$ (1), 0.5 (2), 1 (3).