Theory of passive mode-locking of semiconductor disk lasers in the blue spectral range by metal nanocomposites

Kwang-Hyon Kim,[1,2] Uwe Griebner,[1] and Joachim Herrmann[1,*]

[1]*Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born-Str. 2a, Berlin, D-12489, Germany*
[2]*Institute of Lasers, State Academy of Sciences, Unjong District, Pyongyang, DPR Korea*
**jherrman@mbi-berlin.de*

Abstract: We theoretically study femtosecond pulse generation by passive mode-locking of semiconductor disk lasers operating in the blue spectral range using metal nanocomposites as slow saturable absorbers. By using the relation for the nonlinear dielectric response of a layer of silica glass doped with spherical silver nanoparticles and the master equation for mode-locking, we investigate the dynamics of pulse formation and the achievable pulse parameters and predict the generation of pulses as short as 50 fs at 420 nm in such lasers.

OCIS codes: (320.7090) Ultrafast lasers; (250.5403) Plasmonics.

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1. Introduction

Metal nanocomposites exhibit saturable absorption most prominently in the visible spectral range (see e.g. [1,2]). The recovery time of such materials is in the range of a few picoseconds [3,4]. A prospective application of this type of absorbers is passive mode-locking. In comparison with other well-established mode-locking elements such as semiconductor saturable absorber mirrors (SESAMs) [5], quantum dots [6] and carbon nanotubes [7,8], its operation range can be extended to much shorter wavelengths, down to the blue spectral range and exhibit several additional advantages. In the early works of mode-locking, saturable absorbers in the visible spectral range consisted of dyes dissolved in different solvents [9]. The disadvantages of such saturable dyes, such as maintaining a constant stream in the dye jet, temperature sensitivity and long-term instability restricted their versatility.

Semiconductor disk lasers (SDLs) are compact and low cost devices covering a wide spectral range extending from the blue to the mid-IR [10]). In combination with a fast saturable absorber, mode-locked pulses can be achieved, typically at high pulse repetition rates larger than 1 GHz [11], which are attractive, e.g., for frequency comb generation [12]. Mode-locking of SDLs was demonstrated at different wavelengths in the infrared spectral range using SESAMs [11,13]. The shortest pulses to date were generated at around 1 μm with 107 fs and 198 fs in the fundamentally and harmonically mode-locked regime, respectively [13]. Theoretical studies of SDLs mode-locked by SESAMs have been published in [14,15]. Recently, several continuous-wave SDLs emitting in the blue spectral range were realized [16,17]. Special applications of femtosecond optical pulses in the blue spectral range are in bio-imaging or in the next generation of high-density optical storage [18], but the present laser sources are rather complex and expensive. Therefore, blue femtosecond mode-locked SDLs would be an attractive alternative source. In order to attain passive mode-locking of SDLs, composite materials doped with metal nanoparticles (NPs) are a promising candidate because of their distinguished saturable absorption properties in the visible range. In particular, compared to the wide-spread semiconductor-based saturable absorbers, NP-based saturable absorbers can be fabricated much simpler and more cost-effectively. In [19], the authors have theoretically studied passive mode-locking of dielectric solid-state lasers emitting in the green spectral range by using metal nanocomposites as slow saturable absorbers and predicted pulse generation with a duration as short as 100 fs in a Ho:YLF laser at 545 nm. Unlike dielectric solid-state gain media, semiconductor gain materials dynamically respond with a much shorter response time, which is in the order of a few nanoseconds, and the mode-locked dynamics differ by this reason.

In this paper, we analyze passive mode-locking of SDLs in the blue spectral range with metal nanocomposites as slow saturable absorbers from a theoretical point of view. To utilize the strong saturable absorption of NPs one has to ensure that the surface plasmon resonance (SPR) is located at the central lasing wavelength of the gain medium. In general, this is accomplished by tailoring the size and shape of metal NPs and choosing an appropriated

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embedding medium. Here we study passive mode-locking of a GaN-based SDL operating at a central wavelength of 420 nm. As will be shown, such laser can be mode-locked by a thin layer of silica glass doped with spherical silver NPs. The plasmon resonance of the latter is located at 414 nm.

2. Theoretical fundamentals

In metal NPs, the absorption near the plasmon resonance becomes saturated with increasing intensity [1,2] corresponding to a saturation intensity in the range of 10 MW/cm^2. The physical reason for this effect is related to the intensity-dependent dielectric function of metals leading to a nonlinear shift of the plasmon resonance [2]. The transient nonlinear response of the NPs is determined by the electron thermalization and the cooling of the hot electrons through the thermal exchange with the lattices in the metal. These processes can be described by the semiclassical two-temperature model for the electron temperature T_e and the lattice temperature T_L.

For a moderate pump fluence the change of the dielectric function of the metal is proportional to the change of the temperature of the electrons in the metal NPs. Based on these facts, an equation for the transient dielectric function of the metal NPs can be derived, which is given by [19]

$$\frac{\partial \varepsilon_m}{\partial t} = -\frac{\varepsilon_m - \varepsilon_m^{(0)}}{\tau_{ep}} + \frac{\chi_m^{(3)}}{\tau_{ee}\tau_{ep}} \int_{-\infty}^{t} \left| x(t') E(t') \right|^2 \exp\left(-\frac{t-t'}{\tau_{ee}} \right) dt', \tag{1}$$

where $\varepsilon_m^{(0)}$ is the linear and $\varepsilon_m(t)$ the nonlinear dielectric function, $\chi_m^{(3)}$ the degenerate third-order susceptibility of the metal at the pump wavelength, τ_{ee} and τ_{ep} are the electron-electron scattering time and the electron-phonon coupling time, respectively, $x(t) = 3\varepsilon_h / \left[\varepsilon_m(t) + 2\varepsilon_h \right]$ is the time-dependent field enhancement factor, ε_h is the permittivity of host medium, and $E(t)$ is the electric field of the incident light.

The effective dielectric function of metal nanocomposites can be directly calculated by using the Maxwell-Garnett model for nanospheres smaller than 10 nm, given by

$$\varepsilon_{eff} = \varepsilon_h \frac{1 + 2f(1-x)}{1 - f(1-x)}, \tag{2}$$

where f is the volume filling factor of the metal NPs. For larger or non-spherical NPs, the effective medium approximation in combination with the discrete dipole approximation can be used (for details, see [19]).

To study the lasing dynamics of SDLs containing a metal nanocomposite as a slow saturable absorber in the laser cavity, we apply the following standard master equation

$$T_R \frac{\partial A(T,t)}{\partial T} = -iD\frac{\partial^2 A}{\partial t^2} + \left[(1 - i\alpha) g - l + D_{g,f} \frac{\partial^2 A}{\partial t^2} - q(T,t) \right] A(T,t), \tag{3}$$

where T_R is the round trip time, A is the envelope of the intracavity field, α is the linewidth enhancement factor, D is the group delay dispersion (GDD), $D_{g,f} = g/\Omega_g^2 + 1/\Omega_f^2$ is the gain and intracavity filter dispersion, Ω_g and Ω_f are the gain and filter bandwidth (HWHM), $g(t)$ and l are the gain and the loss of passive cavity, and $q(t)$ describes the saturable absorber loss and the nonlinear refractive index of the NP-nanocomposite,

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respectively (see e. g [14,15,17].). In the above equation, the gain coefficient $g(t)$ is given by

$$\frac{\partial g}{\partial t} = -\frac{g - g_0}{\tau_g} - g\frac{|A(t)|^2}{E_g}, \tag{4}$$

where τ_g is the gain recovery time, E_g is the gain saturation energy dependent on the saturation fluence and the beam diameter on the gain medium. The absorber loss is given by

$$q(T,t) = -i\frac{4\pi}{\lambda_L}\sqrt{\varepsilon_{eff}}\,d, \tag{5}$$

where λ_L is the central wavelength of the laser, ε_{eff} is the effective dielectric function of the composite, and d is its thickness. Since the effective dielectric function ε_{eff} is complex, $q(T,t)$ becomes also a complex quantity leading to the modulation of both the loss and the refractive index during one round trip.

3. Numerical results

Figure 1(a) shows the transient transmittance of a 1-µm thin silica glass layer doped with Ag nanospheres smaller than 10 nm for different pump pulse fluences at 430 nm. The pump pulse duration is 50 fs and τ_{ee} and τ_{ep} were chosen to be 100 fs and 1 ps, respectively. The dielectric functions of silver and silica have been taken from [20] and $\chi_m^{(3)}$ from [21]. The calculated recovery time of the NP-nanocomposite yields 3 ps and compiles with the needed requirements of an absorber recovery time of few ps for femtosecond SDLs [13].

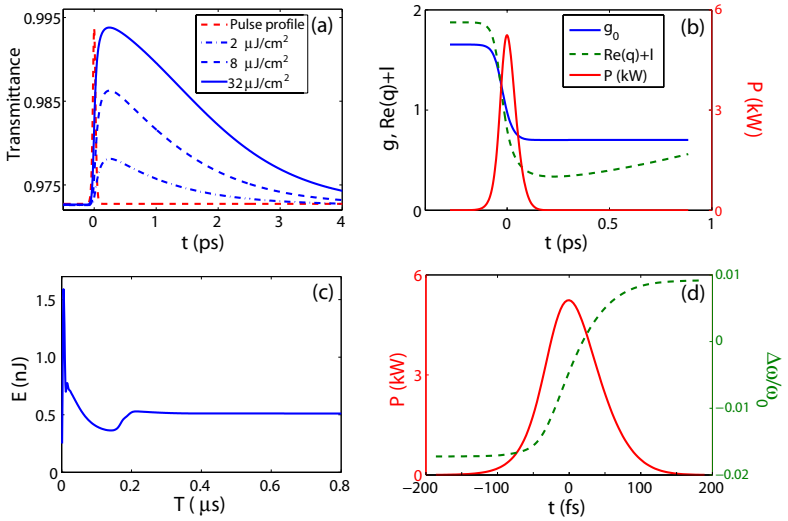

Fig. 1. (a) Transient transmittance of silica glass doped with spherical Ag NPs smaller than 10 nm at 430 nm for different pump pulse fluences; Ag NP filling factor: 10^{-4}, thickness of composite layer: 1 μ m, pump pulse duration: 50 fs. (b)-(d) Pulse evolution in the GaN-based semiconductor disk laser (operating at 420 nm) passively mode-locked by silica glass doped with Ag nanospheres for GDD parameter $D = 100$ fs^2 and filling factor $f = 3.5\times10^{-3}$. (b) Behavior of gain and loss during the pulse formation, (c) evolution of pulse energy on a µs-time scale, (d) pulse intensity and frequency shift during the pulse.

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Here we study a mode-locked semiconductor disk laser containing GaN as gain material as an example [16,17]. The laser is operating at a central wavelength of 420 nm and the length of the resonator is set to 20 cm. The small signal gain is chosen to be $g_0 = 2$, the gain linewidth $\Omega_g = 2\pi \times 20.39$ THz, and the linewidth enhancement factor $\alpha = 2.8$ [16,17]. The gain recovery time is approximately 1 ns [22,23] and the saturation energy of the gain medium 0.6 nJ. As a saturable absorber we have chosen a 1-µm thin silica glass film doped with Ag nanospheres smaller than 10 nm. In this case, the SPR is located at 414 nm and the composite exhibits strong saturated absorption at the lasing wavelength of 420 nm. The main contribution to nonsaturable loss of NP-SAs is related to scattering. For the estimation of the latter we refer to the precise calculations for gold NPs of [24]. For NP diameters smaller than 20 nm scattering loss is negligible in the visible spectral range [24]. Therefore, applying metal nanoparticles smaller than 10 nm, as in our case, we can approximately neglect nonsaturable loss caused by scattering. Additionally, loss related to NP size distributions are not relevant for such small NPs. The substrate itself is also silica glass and only the 1-µm thick surface layer is doped with Ag NPs. The substrate thickness can be chosen to adjust the positive dispersion in the cavity when required.

In Figs. 1(b)-1(d), we present calculated results and properties of the passively mode-locked laser operation of this laser with a group delay dispersion (GDD) of $D = 100$ fs^2 and a beam area on the metal nanocomposite of 0.002 mm^2. Figure 1(b) demonstrates the mechanism of mode-locking. Pulse shortening and stability is explained by the combined action of the dynamics of saturable gain and saturable loss. This can be seen in Fig. 1(b) because the net gain is negative both at the leading and the trailing fronts of the generated pulse. The leading edge of the pulse is suppressed by the action of the absorber, until the pulse energy has reached a value at which the absorption is greatly diminished due to saturation. On the other hand, the trailing edge is suppressed as well because above a certain pulse energy the amplification is decreased due to the depletion of the population inversion in the gain medium. This mechanism is similar as in passively mode-locked dye lasers. In Fig. 1(c) the evolution of the pulse energy is presented. The formation of a pulse containing the highest pulse energy occurs at the very beginning of the process (~20 ns) while the pulse stabilization with the formation of a cw regime takes place on a much longer timescale (200 ns) because of the energy-depending dynamic of the pulse build-up with increasing round-trip numbers. After reaching the cw-regime, the resultant pulse duration is 83 fs. Figure 1(d) shows the intensity profile and the chirp of the pulse. The pulse is positively chirped which can be explained by the nonlinear index of gain and absorber.

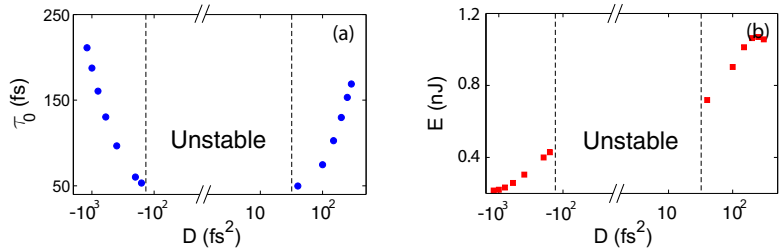

Fig. 2. Pulse duration (a) and pulse energy (b) as a function of the group delay dispersion D for the GaN-based semiconductor disk laser at 420 nm passively mode-locked by Ag nanospheres doped silica glass: filling factor is $f = 3.5 \times 10^{-3}$.

Figure 2 shows the dependencies of pulse duration and energy on the GDD. The other parameters are the same as in Fig. 1. Pulse shaping is unstable only for a small dispersion range between −150 fs^2 and 40 fs^2. This is attributed to the imbalance between the dispersion-induced pulse broadening and compression by the dynamic gain and loss due to the slow response of the gain and the metal NP-nanocomposite. The pulse duration in the positive and negative GDD ranges attain similar values. However, for the same pulse duration the absolute

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value of negative GDD is larger than those for positive GDD value which can be interpreted by the fact that the pulse broadening effect is stronger for positive GDD because of the positive chirp of the pulses. As Fig. 2(a) indicates the shortest pulse duration of about 54 fs is achieved for a negative dispersion parameter of $D = -160$ fs^2, while the shortest duration in the positive GDD range is very similar, about 55 fs for D = 50 fs^2. In Fig. 2(b), the dependence of the pulse energy on the GDD parameter is presented. A remarkable point is the larger pulse energy in the positive GDD range than in the negative GDD range, which can be explained by the advancement of the pulse due to the negative GDD and the positive chirp leading to strong suppression in the leading part of the pulse by the absorber loss. Since the shortest pulse durations are very similar, the positive GDD range is more favorable due to the higher pulse energy.

Passive mode-locking by NP-composites is possible only in a small interval of filling factors. For filling factors smaller than $f = 3\times10^{-3}$ the mode-locked operation becomes unstable due to the excessive dynamic range of saturable loss. For filling factors larger than $f = 4\times10^{-3}$, lasing itself becomes impossible due to the negative small signal net gain. For $f = 3\times10^{-3}$ and $f = 3.5\times10^{-3}$, the resultant pulse durations and pulse energies are $\tau_0 = 87$ and $\tau_0 = 83$ fs, and $E = 1.75$ and $E = 0.55$ nJ, respectively. These values are calculated for $D = -300$ fs^2. Other parameters were taken to be the same as in Fig. 2.

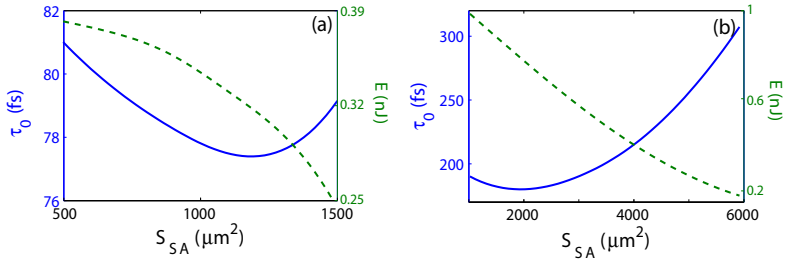

Fig. 3. Dependencies of pulse duration and energy on the beam area on the saturable absorber for $D = -300$ fs^2 (a) and $D = 300$ fs^2 (b).

In Fig. 3 we show the dependencies of pulse duration and energy on the beam area on the metal nanocomposite absorbers for the same absolute values of the pump power for GDD parameters $D = -300$ fs^2 and $D = 300$ fs^2. The other parameters are the same as in Fig. 1. The figure shows that the pulse duration has a shortest value for an optimum beam area (or correspondingly for an optimum pump fluence) but is not altered significantly when changing the beam area. This is in contrast to the case of dielectric solid-state lasers mode-locked by metal NP-nanocomposites, where the pulse duration depends much stronger on the beam area [18]. With the above given parameters, the fluence on the silver NP saturable absorber is in the range from ~15 to ~45 μJ/cm^2.

4. Conclusion

To conclude, we have theoretically studied passive mode-locking of semiconductor disk lasers with metal nanocomposites as saturable absorbers in blue spectral range. For a GaN-based semiconductor disk laser operating at 420 nm and 1μm thick layer of silica glass doped with silver NPs, we studied the dependence of pulse parameters on the absorber and laser parameters and predicted a shortest pulse duration of about 50 fs. Compared with other saturable absorbers, the application of composites containing metal NPs offers several advantages. It allows the development of very compact and cheap mode-locking devices with tunable operation regions, extending from IR down to the blue spectral range.

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