Ultrafast laser heating of non-plasmonic nanocylinders

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Abstract. We develop a model describing non-equilibrium processes under the excitation of resonant semiconductor nanostructures with ultrashort laser pulses with a duration of about 100 fs. We focus on the heating effects related to pulsed excitation with account on free carriers generation, thermalization, and relaxation. The heat exchange between the electron and phonon system is treated within the two-temperature model. We applied the developed model to describing pulsed heating of silicon nanocylinder on top of a dielectric substrate. We come up with estimations of the thermal damage threshold of the considered structures which provides the limits for the experimental conditions and ensures thermal stability of the samples.

1. Introduction
The recent years have been highlighted with the rapid and extensive development of nonlinear all-dielectric photonics [1]. Compared to plasmonics, all-dielectric nanostructures possess high mode volume and large nonlinear coefficients resulting in extremely high efficiency of second [2, 3] and third [4] order nonlinear optical effects as well as time varying optical response [5, 6]. At the same time, the residual free-carrier absorption along with single and multiphoton interband absorption may result in heating of dielectric and semiconductor nanostructures which form the main platform of all-dielectric photonics [7]. The efficient optical heating has been recently observed in continuous wave regime [8] opening a way for strong thermo-optical nonlinearity [9, 10, 11].

The rigorous approach of pulsed excitation heating modelling has been developed in plasmonics [12]. It allows to consider the resonant properties of metal nanoparticles for exact temperature rise estimation. However, this model is inapplicable to the semiconductor structures because of the free carriers generation during photoexcitation which changes significantly the optical properties of the investigated material [13]. The numerical simulation of ultrafast optical heating of semiconductor microscale waveguide structures has been performed with analyzing exact material properties [14]. At the same time, the thermooptical effects in the regime of pulsed excitation of resonant subwavelength structures have not been studied so far, despite of extensive studies of nonlinear optical effects under femtosecond pulse illumination. In this work we develop a semi-analytical model describing the ultrafast heating of semiconductor nanoparticles under femtosecond laser pulse excitation. We combine the exact numerical simulation with extracted key model parameters such as the rate of heat flow into the substrate and absorption cross section. Basing on our model, one can estimate the critical repetition rate of the laser pulses for different intensities which provides the limits of thermooptical damage threshold. The obtained results are highly important for describing the optical response of dielectric and semiconductor nanostructures under high-intensity laser illumination in nonlinear optical experimental setups.

2. The model
We consider cylindrical silicon nanoparticles placed on top of a glass substrate (see the inset Fig. 1(a)) and subjected to linearly polarized femto-second laser illumination. We utilize two-temperature electron-phonon model taking into account three correlated processes: i) absorption of incident femtosecond laser radiation accompanied by electron-hole plasma generation, ii) thermalization process...
between the electron and lattice subsystems associated with electron-phonon interaction, and iii) heat leakage into the substrate.

We simulate the process of nanocylinder heating with the following system of differential self-consistent time-dependent equations for the free carrier concentration $n$ which corresponds to both electron and hole concentration since we have undoped semiconductor; the electron and phonon (lattice) temperatures $T_e$ and $T_{ph}$:

$$\begin{align*}
\frac{\partial n}{\partial t} &= \frac{\alpha I(t)}{\hbar \omega} + \frac{\beta I(t)^2}{2\hbar^2 \omega} - \gamma n^3, \\
C_{e-h}(n) \frac{\partial T_e}{\partial t} &= g(T_{ph} - T_e) + Q(t), \\
C_{ph} \frac{\partial T_{ph}}{\partial t} &= g(T_e - T_{ph}) - \kappa (T_{ph} - T_0).
\end{align*}$$

(1)

Here $I$ is the incident field intensity; $\alpha = 1.0086 \cdot 10^4$ cm$^{-1}$ and $\beta = 2$ cm/GW are the coefficients of one- and two-photon absorption respectively; $\gamma = 3.8 \cdot 10^{-31}$ cm/s is the Auger recombination coefficient; $\hbar \omega$ is the energy of incident photons; $C_{e-h}(n)$ and $C_{ph}$ are the electron-hole plasma and phonon specific heat capacities; $Q(t)$ is the volume density of the absorbed electromagnetic power; $g$ and $\kappa$ are the electron-phonon coupling and heat outflow coefficients respectively.

The first equation of the system corresponds to the free carriers concentration change in the system. It increases due to the processes of one- or two-photon absorption proportional to the first and second power of the incident field intensity $I$ respectively. The free carrier concentration decreases because of Auger recombination which is the dominant mechanism of electron relaxation for the high density plasma.

The last two equations of system (1) describe the evolution of electron-hole plasma and phonon systems temperature. The femtosecond laser pulse excitation with the volume power density $Q(t)$ generates hot electron gas which subsequently interacts with lattice phonons. Electron-phonon coupling underlies the transfer of energy from hot electrons to lattice vibrations defined. The coupling constant between the subsystems $g = C_{e-h}/\tau_{e-ph}$ is governed by electron-phonon relaxation time $\tau_{e-ph} = 240$ fs.

The nanocylinder phonons are coupled to the substrate thermal reservoir, which we describe with the constant $\kappa$ which depends on the material properties and on the particular configuration of the thermal contact between the nanoresonator and the substrate. We extracted the $\kappa$ coefficient from exact numerical modelling of the the cylinder on the glass substrate with help of commercially available COMSOL Multiphysics software. We modeled transient cooling of nanocylinder initially heated to homogeneous temperature $T_0$ in the absence of optical excitation and nonequilibrium carriers. Assuming exponential dependence of phonon temperature $T_{ph} \sim \exp(-\kappa t/C_{ph})$, we obtained the relaxation constant $\kappa$ from the temporal dependence of the cylinder temperature.

The absorbed density of electromagnetic power could be estimated from formula (2):

$$Q_{abs} = \frac{\omega}{2} \text{Im} \varepsilon \int |E|^2 dV \equiv \sigma_{abs} \cdot I,$$

(2)

where $\varepsilon$ is the dielectric permittivity of the nanoparticle, $\omega$ is the incident light frequency, $|E|$ is the electric field amplitude inside the particle and the integral is taken over the nanocylinder volume, $\sigma_{abs} = Q_{abs}/I$ is the absorption cross section.

The energy absorbed during a single optical pulse can not be considered as constant since the free carriers concentration significantly changes during the excitation, which modulates the imaginary part of the dielectric constant according to the Drude model:

$$\Delta \text{Im} \varepsilon(n) = \frac{\omega_p^2(n) \tau_{e-e}}{\omega(1 + \omega^2 \tau_{e-e}^2)}.$$  

(3)

Here $\tau_{e-e}$ is the characteristic electron-electron scattering time which can be as small as $\tau_{e-e} = 1$ fs for high density electron-hole plasma [15] and $\omega_p(n)$ is the plasma frequency defined as the function of generated electron-hole plasma concentration

$$\omega_p(n) = \sqrt{\frac{n e^2}{\varepsilon_0 m_{eff}}}.$$  

(4)
where \( m_{\text{eff}} = 0.18m_e \) is the effective mass of holes and electrons in crystalline silicon (c-Si), \( \varepsilon_0 \) is the permittivity of vacuum, and \( e \) is the elementary charge.

Here, for the simplicity of the model, we suppose that the real part of the refractive index and consequently the spectral position of the resonances does not change significantly during the excitation, which is a valid assumption for not very high quality resonances. Then the absorbed power as the function of the free carrier concentration can be estimated as

\[
Q_{\text{abs}}(n) = \sigma_{\text{abs}}(n_0) \left( 1 + \frac{\Delta \text{Im}(\varepsilon(n))}{\text{Im}(\varepsilon(n_0))} \right) I.
\]

The absorption cross-section \( \sigma_{\text{abs}}(n_0) \) was modelled using the COMSOL Multiphysics package. We reduced fully three-dimensional electromagnetic problem to the two-dimensional one using the axial symmetry of the structure under consideration as it was done in our previous works, for example, in [17].

3. Results

Taking into account all the equations presented above, one can solve the system of differential equations with the help of the key parameters \( \sigma_{\text{abs}}(n) \) and \( \kappa \) obtained from full wave numerical simulation. For a quantitative consideration of the model, we simulate numerically the heating of a nanocylinder with radius \( r = 200 \) nm and height \( h = 500 \) nm under normal plane wave incidence as shown in Fig. 1(a) inset. System (1) is solved numerically using MATLAB software for the experimentally measured physical constants for silicon [14].

The results of the modelling are shown in Fig. 1(a). At the initial stage of heating, we observe the sharp rise in the electron temperature under laser pulse excitation with duration of \( \tau_p = 100 \) fs. After the pulse is over, the electron temperature goes down and there appears slow and gradual increase in the phonon temperature. Since the characteristic exchange time between electron and phonon systems is \( \tau_{e-ph} = 240 \) fs, the thermal equilibrium between them is reached at the scale of \( \sim 2 \) ps.

![Figure 1](image_url)

**Figure 1.** a) Time dependence of phonon \( T_{\text{ph}} \) and electron temperature \( T_{\text{el}} \) for a single laser pulse excitation with an intensity \( I = 100 \) GW/cm\(^2\) and pulse duration \( \tau_p = 100 \) fs. The inset shows a schematic view of nanocylinder excitation under a series of pulses with repetition rate \( \nu \). b) Dependence of the maximal temperature on the pulse repetition rate \( \nu \) for various incident intensities. The melting point of c-Si is marked with a horizontal dashed line to estimate the critical repetition rate leading to the structural damage for a given intensity.

After modelling one pulse, we proceed to study the behavior of the phonon temperature during several consequent laser pulses (Fig. 2). Simulation of a finite number of pulses was conducted by applying the results of computation for a single pulse excitation as the initial conditions for the next pulse.
Figure 2. Dependence of the phonon temperature upon heating by successive pulses of femtosecond laser with intensity $I = 100 \text{ GW/cm}^2$ for different pulse repetition rates. The maximum temperature is reached during the first pulse for cases a) and b) corresponding to the repetition rates $\nu = 1 \text{ MHz}$ and $\nu = 10 \text{ MHz}$, respectively. For c) and d) with $\nu = 50 \text{ MHz}$ and $\nu = 100 \text{ MHz}$, the temperature is established over a long period of time and the maximum is reached only after a series of pulses.

The modelling results lead to the existence of two different cases for temperature evolution. In the first case, at relatively low frequencies (Fig. 2(a, b)), the phonon temperature reaches its maximum during the first pulse. At the scale of a single cycle in case of a low repetition rate, the temperature drops to initial values due to the drain of heat into the substrate. An interesting fact is that the concentration of free carriers decreases much slower than the temperature of the cylinder. Since that the heat capacitance of free carriers stays relatively high $C_{\text{e-h}} = 3nk_B$ and significantly exceeds the initial value before the first pulse came. Thus, the first and subsequent pulses are significantly different in the temperature increase as it is seen in Fig. 2(a). In the case of higher repetition rate (see Fig. 2(c)), the next pulse comes before the temperature drops to initial and the averaged temperature increases with subsequent pulses coming. Therefore, with each pulse, the temperature rises until it stabilizes (Fig. 2(c, d)). Under stable temperature we understand the situation in which the maximum temperature of the structure stays the same for all further pulses. It occurs when the initial conditions for all the parameters of our problem coincide for the subsequent pulses.

So, we estimate the maximum temperature during the multi-pulse heating process as a function of the pulse repetition rate for different intensities (Fig. 2(b)). By comparing the obtained results with the melting point of silicon, we can predict the maximum allowable pulse repetition rate before the structure is damaged. It is remarkable that for typical power of 150 GW/cm$^2$ the thermal damage is achieved at 80 MHz repetition rate which corresponds to real experimental conditions.
4. Conclusion
In conclusion, we studied a self-consistent two-temperature model of semiconductor nanocylinders heating via pulsed laser excitation. We took into account both the heat diffusion into the substrate and the change in the concentration of free carriers due to the optical excitation. On the basis of this model, the temperature evolution of nanodisks over a series of pulses has been observed. We estimated critical repetition rate of the laser pulses for different incident intensities before the structure under consideration is damaged. We assume further development of this model in the case of other nanoparticles shape or some resonant features for application to specific problems, and we are convinced that described calculations will be useful for analytical evaluation of heating when observing various pulse-excited effects in nanophotonics.

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