Light induced temperature decrease of semiconductor nanoparticle

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Abstract. We demonstrate model of light induced refrigeration of single semiconductor resonant spherical nanoparticles. In this paper, we reveal that cooling efficiency can be increased by several times in comparison with bulk material due to excitation of optical Mie-resonances in the nanoparticles. We believe that such approach can be promising for the implementation of all-optical laser cooling devices.

1. Introduction
All-dielectric nanophotonics attracted a lot of attention in recent years among leading scientific groups due to the ability of a single nanoresonator to localize and significantly enhance electromagnetic field on a sub-wavelength scale [1]. Such field localization occurs owing to excitation of so-called Mie-resonances [2] of both nature - electric and magnetic ones in the visible range [3, 4]. Resonant high-Q optical Mie-modes lead to the amplification of the nanoparticles’ absorption, thus making them promising for heat generation at the nanoscale applications. Indeed, it was proven both theoretically and experimentally that resonant crystalline silicon nanosphere can be more efficient in terms of optical heating and nanothermometry [5, 6] in comparison with conventional plasmonic structures [7]. Moreover, all-dielectric laser heating approach demonstrated great performance in thermo-optical applications, including local crystallization of amorphous nanostructures [8], metasurface reshaping via precise melting [9], real-time tracing of protein unfolding processes [10] and others. Therefore, resonant semiconductor materials and nanostructures were studied thoroughly for thermal applications, but only as a light-to-heat conversion tool. However, we believe that semiconductor resonant nanostructures are highly efficient all-optical cooling platform that has not been studied yet.

In this regard, an optical refrigeration of solid structures and objects is found to be attractive mechanism of temperature decrease. Such approach one can find optimistic for the possible fabrication of compact cooling devices. Indeed, first experimental demonstration of this effect was revealed for Yb³⁺-doped fluoride glass in 1995 [11]. Subsequent research improved this material and recent studies demonstrated the concept of a solid-state all-optical cryocooler [12].

On the other hand, laser cooling of semiconductors one can found more attractive because as expected temperatures in such systems can be significantly lower than the RE-doped glass structures achieve. Moreover, additional advantage of the semiconductor cooling systems is the possibility for simplified integration with the devices.
Optical cooling of semiconductors was recently demonstrated for CdS nanobelts systems [13]. The system was cooled down by more than 40K. The ability of the nanostructures to support optical cooling is maintained by the cooling ratio, which was introduced by [14] as following expression:

\[ \eta_c = \eta_{ext} \frac{\omega_{PL}}{\omega} - 1, \]  

where \( \eta_{ext} \) is an external radiative quantum efficiency, \( \omega_{PL} \) is a PL frequency and \( \omega \) is a frequency of absorbed light. Firstly, resonant Mie-nanoparticle allows to decrease incident light frequency maintaining possibility for optical cooling due to enhanced absorption in the bang gap region. On the other hand, radiating dipole located in spherical resonant nanoparticle can achieve an increase in emission rate [15, 16], thus making possible to increase the quantum efficiency \( \eta_{ext} \). Balance of these two effects determines cooling efficiency.

In this work, we theoretically study an optical cooling of single resonant spherical nanoparticle of semiconductor materials with strong exitonic contribution. We reveal, that such nanoparticles with pronounced magnetic dipole and quadrupole optical modes can support the highest efficiency in optical cooling process.

2. Results

According to the classic work on the optical cooling we will solve the kinetic equation for the light-generated electron-hole density [14], but also taking into account radiative recombination term and absorption for nanoparticle:

\[ \frac{dN}{dt} = AN + BF_p N^2 + CN^3 - \frac{I \sigma_{abs}}{\bar{V} \bar{\hbar} \omega} = 0, \]  

where \( I \) is the laser intensity, \( F_p \) is the emission rate, \( \sigma_{abs} \) is the absorption cross section and \( \bar{V} \) is the volume of the nanoparticle. The recombination processes are nonradiative - \( AN \), radiative - \( BF_p N^2 \), and Auger - \( CN^3 \). Typical values of \( A, B, C \) for semiconductors with strong exitonic contribution were taken elsewhere [17].

For such materials the dependence of the dielectric permittivity \( \varepsilon \) is given by the relation [18]:

\[ \varepsilon(\omega) = \varepsilon_0 + \frac{\omega_p^2}{\omega_{exc}^2 - \omega^2 - i\gamma \omega}, \]  

where \( \omega_{exc} \) is the frequency of excitonic transition, \( \omega_p \) is the strength of a dipole oscillator, \( \gamma \) is the damping factor, \( \varepsilon_0 \) is the background dielectric constant. In this work we use following values: \( \varepsilon_0 = 5.2, \omega_p = 0.387 \text{ eV}, \gamma = 0.09 \text{ eV}, \omega_{exc} = 2.55 \text{ eV} \).

Absorption cross section \( \sigma_{abs} \) for a spherical nanoparticle with the permittivity from Equation 3 was calculated according to Mie-theory [2]. Subsequently, normalized average emission rate for the nanosphere was calculated using model described elsewhere [19]. The results of are shown in the left part of the Figure 1. Different peaks correspond to the different modes: magnetic dipole (MD), magnetic quadruple (MQ) and magnetic octopole (MO), respectively. Solving Equation 2 for different nanosphere radius and wavelength of absorbed light gives the value of light induced electron-hole carrier density \( N \).

The cooling efficiency \( \eta_c \) is given by the ratio of the cooling power \( (P_{lum} - P_{abs}) \) and absorbed power \( (P_{abs}) \) [20]:

\[ \eta_c = \frac{P_{lum} - P_{abs}}{P_{abs}} = \frac{BF_p N^2}{AN + BF_p N^2 + CN^3} \frac{\hbar \omega_{PL}}{\bar{\hbar} \omega} - 1, \]
Figure 1. Left side: emission rate of a spherical nanoparticle with ε calculated from Equation 3 for different nanoparticle radius (R). Right side: cooling efficiency of a spherical nanoparticle calculated from Equation 4. Black dashed lines correspond to the optical modes excited in the nanosphere on the emission rate wavelengths. Green dashed lines correspond to the optical modes excited in the nanosphere on the incident light wavelengths.

where \( \omega \) is the frequency of absorbed light and \( \omega_p \) is the photoluminescence frequency. The results of the calculated \( \eta_c \) are presented in right part of the Figure 1. Black dashed lines are denoted to the optical modes supported by the sphere at the emission wavelength. Whereas green dashed lines correspond to the optical modes excited in the absorption cross sections.

The optical cooling takes place when \( \eta_c \) is positive. The positive cooling efficiency is observed in the region of intersection of the ‘absorption’ and the ‘emission’ modes which are shown in the Figure 1. Thus, we reveal that the highest cooling efficiency is achieved when the nanosphere supports one optical mode at the absorption (incident light) wavelength and the other optical mode at the emission (photoluminescence) wavelength.

3. Conclusions
In summary, we have demonstrated the model of light induced refrigeration of a single semiconductor resonant spherical nanoparticle. The cooling efficiency enhancement by several times in comparison with bulk material due to the excitation of optical Mie-resonances inside the nanoparticle is presented. We believe that such concept will pave the way for the nanoscale all-optical cooling devices.

Acknowledgments
This work was supported by Russian Science Foundation (project 18-79-00338) and Ministry of Science and Higher Education of the Russian Federation (Project 16.8939.2017/8.9).
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