Can Laser Light Cool Semiconductors?

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(Received 14 August 2003; published 18 June 2004)

Laser cooling in semiconductors is theoretically investigated including arbitrary external efficiency and photon recycling. Experimental conditions needed to attain net cooling in GaAs are derived.

DO: 10.1103/PhysRevLett.92.247403 PACS numbers: 78.55.Cr, 32.80.Pj, 78.20.–e, 78.66.Fd

The concept of laser cooling (optical refrigeration) by luminescence up-conversion in solids dates back to 1929 [1]. Pringsheim recognized that thermal vibrational energy can be removed by anti-Stokes fluorescence if a material is excited with photons having energy below the mean fluorescence energy (see Fig. 1). Material purity problems prevented observation of this type of laser cooling until 1995, when it was first demonstrated in ytterbium-doped glass [2]. This was followed soon after by reports of cooling in dye solutions [3] and thulium-doped glass [4]. Attaining net cooling in semiconductors, however, has remained elusive. A key problem has been the inability of luminescence to efficiently escape from a semiconductor due to total internal reflection [5,6]. Although the theory of semiconductor cooling has been tackled previously [7,8], the critical issues of luminescence trapping and redshifting have not been taken into account. These processes have the potential to frustrate attempts to achieve semiconductor net cooling. Here, we resolve this problem and show that laser cooling of semiconductors is feasible.

A primary advantage of semiconductors compared to rare-earth doped solids is their potential for achieving temperatures \(\sim 10\) K and below. This is due to the difference of the ground state populations in the two systems. As the temperature drops below 100 K in a rare-earth doped system, the population at the top of the ground state manifold dramatically decreases, rendering the cooling process highly inefficient. This is a consequence of Boltzmann statistics. Semiconductors, on the other hand, obey Fermi-Dirac statistics, which keeps the lower energy valence band populated even at absolute zero.

In this Letter, we describe laser cooling in semiconductor structures allowing for arbitrary external efficiency. Our analysis, for the first time, accounts for the luminescence redshift due to reabsorption. We combine this theory with the established plasma theory of semiconductor optical absorption including many-body Coulomb effects and band filling. The necessary experimental conditions for observing laser cooling in bulk semiconductor heterostructures are obtained.

We investigate an intrinsic (i.e., undoped) semiconductor system uniformly irradiated with a laser light at photon energy \(h\nu\). We make the realistic assumption that only a fraction (\(\eta_v\)) of the luminescence can escape the material; the remaining fraction \((1 - \eta_v)\) is trapped and reabsorbed, which causes the reexcitation of electron-hole pairs and subsequent reemission. This is known as photon recycling. For steady-state conditions at a given temperature, the electron-hole \((e-h)\) carrier density \((N)\) is obtained from the following equation:

\[
0 = \frac{\alpha(\nu, N)}{h\nu} I - AN - BN^2 - CN^3 + (1 - \eta_v)BN^2, \tag{1}
\]

where \(I\) is the laser irradiance and \(\alpha(\nu, N)\) is the interband absorption coefficient. Density dependent absorption results from (i) Coulomb screening and (ii) band filling. The latter effect is saturation due to the Pauli exclusion principle (i.e., Pauli band blocking). It is accounted for by taking \(\alpha(N, h\nu) = \alpha_0(N, h\nu)(f_v - f_c)\), where \(\alpha_0\) is the unsaturated absorption coefficient [9]. The bracketed term is the band-filling factor defined by the Fermi distribution functions \(f_v\) and \(f_c\) for the valence and conduction bands, respectively. The recombination processes are nonradiative \((AN)\), radiative \((BN^2)\), and Auger \((CN^3)\). All the above coefficients are temperature dependent. Rearranging the terms in Eq. (1) yields a radiative recombination rate that scales as \(\eta_v B\). A similar result has

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{Cooling cycle in laser refrigeration of a semiconductor in which a laser photon with energy \(h\nu\) is absorbed followed by emission of an up-converted luminescence photon at \(h\nu_f\).}
\end{figure}
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been derived by Asbeck [10]. It is important to emphasize that \( \eta_e \) is averaged over the entire luminescence spectrum, i.e., \( \eta_e = \int S(\nu)R(\nu)\,d\nu / \int R(\nu)\,d\nu \). Here \( S(\nu) \) is the geometry-dependent escape probability and \( R(\nu) \) is the luminescence spectral density, which is related to the absorption coefficient through the van Roosbroek-Shockley relation [11]. The radiative recombination is obtained from \( BN^2 = \int R(\nu)\,d\nu \) [9].

The net optical power deposited in the semiconductor is the difference between the absorbed power and escaped luminescence:

\[
P_{\text{net}} = \eta_i BN^2 (h\nu - h\nu_i) + ANh\nu + CN^3 h\nu + \Delta P, \tag{2}
\]

where we define an “escaped” mean luminescence photon energy \( h\nu_i = \int S(\nu)R(\nu)h\nu\,d\nu / \int S(\nu)R(\nu)\,d\nu \). A residual heating term \( \Delta P \) accounts for free-carrier absorption and other parasitic absorptive processes. Equation (2) completely describes laser cooling of a semiconductor. It incorporates luminescence trapping via inhibited radiative recombination (\( \eta_i B \)) and includes redshifting of the escaped luminescence \( h\nu_i \).

In the limit of high external efficiency where \( S(\nu) = 1 \), Eq. (2) approaches previous results that assume \( \eta_i = 1 \) and \( \nu_i = \nu_f [5,7,8] \). Equation (2) indicates laser cooling occurs when \( P_{\text{net}} < 0 \), which requires a dominant contribution from radiative recombination. Cooling efficiency \( \eta_e \) is defined as the ratio of cooling power density \( P_c = -P_{\text{net}} \) to the absorbed laser power density \( P_{\text{abs}} = \alpha I + \Delta P \). Ignoring the \( \Delta P \) contributions for the moment, \( \eta_e \) is

\[
\eta_e = \eta_i (\nu_f - \nu_i), \tag{3}
\]

where \( \eta_i = \eta_i BN^2 / (AN + \eta_i BN^2 + CN^3) \) describes the “inhibited” or external radiative quantum efficiency. Assuming the radiative term dominates recombination (i.e., \( \eta_e B > N > A/\eta_e B \)) and ignoring band filling, we can analytically solve for optimum carrier density and incident laser irradiance. We account for parasitic absorption by taking \( \Delta P = \alpha_b I + \sigma_{fca} NI \), where \( \alpha_b \) represents background parasitic absorption from defects and impurities and \( \sigma_{fca} \) is the free-carrier absorption cross section. Net cooling \( (P_{\text{net}} < 0) \) occurs if \( A < A_0 \), where \( A_0 \) defines a “break-even” nonradiative recombination rate:

\[
A_0 = \left( \frac{\eta_i}{\alpha_b} - \frac{\sigma_{fca}}{\alpha(\nu)} \right)^2 \frac{(\eta_e B)^2}{4C^3}. \tag{4}
\]

where we have defined a quantum cooling efficiency \( \eta_q = (h\nu_f - h\nu_i)/h\nu_i \) and \( C = C^3 + \sigma_{fca} \eta_i B/\alpha(\nu) \). Cooling takes place for a laser irradiance in the range \( I_1 < I < I_2 \), where \( I_{1,2} = \{h\nu\eta_i B/\alpha(\nu)\}N_{1,2} \), and the \( n-H \) densities are

\[
N_{1,2} = \frac{A_0}{C^3} \left( 1 \mp \sqrt{1 - \frac{A}{A_0}} \right). \tag{5}
\]

Inclusion of band filling, however, will limit the maximum carrier density attainable with optical pumping \( (N_{\text{max}}) \) that occurs when \( f_c = f_n \). As we discuss below, cooling may be inhibited at low temperature, where \( N_1 \) can approach and even exceed \( N_{\text{max}} \). The parameters \( B \) and \( C \) are fundamental properties of a semiconductor; these coefficients can be calculated and measured for bulk or quantum-confined structures. Reported values, however, vary considerably. In bulk GaAs, for example, the numbers range from \( 2 \times 10^{-16} < B < 7 \times 10^{-16} \text{ m}^3/\text{s} \) and \( 1 \times 10^{-42} < C < 7 \times 10^{-42} \text{ m}^6/\text{s} \). To assess the feasibility of laser cooling, we use average values in our calculations: \( B = 4 \times 10^{-16} \text{ m}^3/\text{s} \) and \( C = 4 \times 10^{-42} \text{ m}^6/\text{s} \) and initially ignore background and free-carrier absorption. The break-even nonradiative lifetime at room temperature \( \tau_{\text{nr}} = 1/A_0 \) is plotted in Fig. 2 as a function of \( \eta_e \). We set \( h\nu_f - h\nu_i = kT \) and use the mean luminescence wavelength of \( \lambda_f \sim 860 \text{ nm} \). The gray area under the curve designates the undesired heating region. The horizontal line corresponds to \( \tau_{\text{nr}} = 40 \mu\text{s} \), which is the longest reported nonradiative lifetime in a GaAs/InGaP double heterostructure, although 5–10 \mu\text{s} is typical [12]. These lifetimes set a lower limit on extraction efficiency \( \eta_e = 15\% \) for the best material; in practice 20%–30% may be required.

Equation (4) suggests increasing the quantum efficiency \( \eta_q \) by decreasing the incident photon energy reduces the break-even lifetime. At \( h\nu_f - h\nu_i = 2kT \), for example, the required lifetime decreases by a factor of 4. As the photon energy moves into the Urbach tail, however, the interband absorption drops rapidly and therefore background and free-carrier absorption are no longer negligible. Recently, it was found that \( \sigma_{fca} \) at band-edge wavelengths is much smaller than previously expected [13]. For GaAs, \( \sigma_{fca} = 10^{-22} \text{ m}^2/\text{Hz} \), which requires \( \alpha(\nu) \gtrsim 10^3 \text{ m}^{-1} \) to ensure that free-carrier losses are

![FIG. 2. The break-even nonradiative lifetime as a function of the luminescence extraction efficiency in bulk GaAs at 300 K. This calculated lifetime delineates the cooling zone and heating zone (shaded area). The horizontal dashed line corresponds to \( \tau_{\text{nr}} = 40 \mu\text{s} \), the longest lifetime reported in a GaAs/InGaP double heterostructure.](image-url)
negligible compared to Auger recombination (i.e., \( C' = C \)). This condition is satisfied even at \( \lambda = 890 \text{ nm} \) (corresponding to \( h\nu_f - h\nu = 2 \text{ K} \)), where \( \alpha(\nu) = 10^4 \text{ m}^{-1} \). Therefore, free-carrier absorption does not present a major limitation to laser cooling. Possible sources for \( \alpha_b \) are (i) impurity absorption in active and/or cladding layers and (ii) substrate absorption. It is also important that \( \alpha_b \) in Eq. (4) be scaled to the device geometry: \( \alpha'_b = \alpha_b \times (d/L) \), where \( d \) and \( L \) are the thicknesses of the lossy and active media, respectively. For an active medium \( L = 1 \mu\text{m} \) and substrate thickness of 1 cm, this requires \( \alpha(\nu) > 10^5 \alpha'_b \). This, in turn, demands \( \alpha'_b < 10 \text{ m}^{-1} \) at \( \lambda = 870 \text{ nm} \), and \( \alpha'_b < 0.1 \text{ m}^{-1} \) at \( \lambda = 890 \text{ nm} \). A ZnS substrate with \( \alpha'_b \leq 10^{-2} \text{ m}^{-1} \) meets such a requirement. A ZnSe substrate having \( \alpha'_b \leq 0.3 \text{ m}^{-1} \) can be effective only at \( \lambda = 870 \text{ nm} \) [14]. We shall later discuss using such substrates to make index-matching dome lenses to enhance \( \eta_c \).

Our analysis to this point has been concerned only with net cooling from room temperature. Of key importance is determining a break-even lifetime for cryogenic conditions. Using the expected scaling \( C(T) \propto \exp(1 - 300/T) \) [15], \( B \propto T^{-3/2} \) [9,16], keeping \( \eta_b = kT/E_g \), and ignoring parasitic losses and the small temperature dependence of the band-gap energy, we obtain

\[
\frac{A_0(T)}{A_0(300)} \approx \frac{300}{T} \exp\left(\frac{300 - T}{T}\right).
\]

This indicates that at \( T = 100 \text{ K} \), for example, the break-even nonradiative lifetime is lowered by \( \sim 20 \) times compared to room temperature. Furthermore, nonradiative recombination is primarily surface recombination, which decreases exponentially with temperature [16]. This makes observation of laser cooling even more favorable at a lower starting temperature, despite decreased efficiency \((= kT/E_g)\). Reduced cooling efficiency at a lower temperature is primarily due to the smaller probability for phonon absorption by electrons and holes. This is manifest in a temperature-dependent exciton linewidth [9]:

\[
\Gamma(T) = \Gamma_0 + \sigma T + \gamma N_{LO}(T),
\]

where \( \Gamma_0 \) is due to an inhomogeneous background distribution of impurities, \( \sigma \) is the contribution from acoustic phonon scattering, \( \gamma \) is the coefficient of LO-phonon scattering, and \( N_{LO}(T) \) is the Bose-Einstein phonon distribution. Exciton-exciton scattering is negligible at the densities encountered here [17]. At lattice temperatures approaching 10 K, the acoustic phonon component dominates and the scattering rate becomes comparable to the radiative recombination rate \((BN^2)\). This means that cold exciton recombination occurs before complete thermalization with the lattice. Hot exciton recombination is a similar process that has hindered observation of Bose-Einstein condensation in semiconductors. This problem has been significantly alleviated by employing quantum-confined systems where relaxation of wave-vector con-

![Image](https://via.placeholder.com/150)

**FIG. 3.** (a) A high index \( n_d \) dome is bonded to a semiconductor heterostructure of index \( n_s \) consisting of a GaAs active layer sandwiched between two thin layers of AlGaAs or InGaP for surface passivation. (b) Heterostructure designs with polished planar surfaces (I) and a textured bottom surface (II).
We evaluate this quantity (depicted in Fig. 3. A density of for Structure I (solid lines) and Structure II (dashed lines) cooling power (bottom) as a function of the GaAs thickness escaped mean luminescence photon energy (middle), and total escape. A polished (planar) surface exhibits a relatively energy photons suffer less absorption and can eventually enhanced efficiency. Photon recycling causes luminescence reabsorption and red-shift, which may adversely affect cooling in textured-surface structures. Highest cooling power is achieved with polished planar structures and index-matching encapsulation.

In summary, we have developed a model to treat laser cooling in semiconductors for arbitrary external efficiency. The break-even nonradiative lifetime and luminescence extraction efficiency are obtained. Photon recycling causes luminescence reabsorption and red-shift, which may adversely affect cooling in textured-surface structures. Highest cooling power is achieved with polished planar structures and index-matching encapsulation.

The authors gratefully acknowledge support from the Air Force Office of Scientific Research (Grants No. F49620-0201-0059 and No. F49620-02-1-0057) and the National Aeronautics and Space Administration (Grant No. NAG5-10373). This work was carried out in part under the auspices of the U.S. Department of Energy. We thank M. P. Hasselbeck for proofreading the manuscript.