# Theoretical study of laser cooling of a semiconductor

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A nonlocal energy-balance equation is derived for the optical absorption, photoluminescence and inelastic electron-phonon scattering, which determines the electron and hole temperatures for any given lattice temperature. The evolution of the lattice temperature is found to be determined by the difference between the power-loss density due to photoluminescence and the power-gain density due to optical absorption, as well as by the initial lattice temperature. We find that in addition to the expected decrease in the lattice temperature, the electron temperature also decreases with time. A laser-cooling power as high as 380 eV/s is predicted for the wide bandgap semiconductor AlN initially at room temperature when the pump-laser field is only 10 V/cm. Laser cooling is found to be more efficient for a large bandgap material, a weaker laser field, and a high initial lattice temperature. The laser-cooling rate then decreases as the lattice cools. The theory presented here provides quantitative predictions that can guide future experiments.

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The cooling of a solid via light-induced fluorescence has been of interest for a very long time.[1–5] This interesting phenomenon involves the excitation of an electron from the valence band edge to the conduction band edge by absorbing a pump photon. This cool electron quickly becomes hot by gaining thermal energy through ultrafast electron-phonon scattering. After a radiative lifetime, recombination of the hot electron will produce a spontaneous photon with energy higher than that of the pump photon. As a result, the lattice will be cooled due to the loss of thermal energy to the electron. It is only recently that this phenomenon has been observed experimentally. Laser-induced fluorescent cooling of heavy-metal-fluoride glass doped with trivalent ytterbium ions was the first realization of this concept.[6] Soon to follow were demonstrations of cooling in dye solutions[7] and thulium-doped glass.[8]

Laser cooling of a semiconductor, however, remains an elusive goal, although it has been pursued for many years.[9,10] Indeed, now more than ever the field of laser cooling is a topic of intense on-going theoretical and experimental investigations.[11] The only theories attempting to model the laser cooling phenomenon are local simulation theories that include rate equations for determining the steady-state carrier density and the loss of lattice energy with several kinetic coefficients. The problem with these theories is that they neglect important dynamical effects such as the change of the carrier distribution when the temperature is lowered. Therefore, they only apply to situations with little change of temperature. The main feature of the rate equation approach is its simplicity, but it is unable to elucidate the essential physics behind the laser cooling phenomena. The key question that still remains open is as follows: what are the best semiconductor materials and conditions for achieving the greatest laser cooling effect? This requires an accurate *nonlocal* theory on a microscopic level, which directly provides an evolution equation for the lattice temperature by including the dynamical effects. This theory should establish a criterion for the occurrence of laser cooling. The theory should also establish a criterion for the efficiency of the laser cooling if it does exist. In this paper, we will present such a *nonlocal* theory for the laser cooling of semiconductors. By including the effect of the carrier distribution, we will be able to uncover the essential physics underlying the phenomenon and we will be able to provide important quantitative predictions that can guide experimentalists toward achieving maximum efficiency of laser cooling in the future.

The conservation of total energy of carriers, phonons and spontaneous photons is assumed due to ultrafast carrier scattering with phonons. It forms the basis of our application of the energy-balance equation to electrons and holes in this paper. However, the energy of phonon system interacting with carriers due to vibration of thermally-isolated lattice can not be balanced by the thermal radiation from the surrounding. As a result, the lattice temperature drops with time due to transferring net energy to electrons and holes. At the same time, the energy-balance equation at a reduced lattice temperature quickly drives the carrier temperature to a new lower quasi-equilibrium value.

Let us first recall the transport force-balance theory for hot electron transport.[12] When a *dc* electric field is applied to a doped semiconductor, there is a drift of electrons as a result of the center-of-mass motion. This is described by a balancing of forces between the frictional forces due to scattering processes and the electrostatic force. At the same time, electrons form a quasi-equilibrium Fermi-Dirac distribution. The electron temperature of this distribution can be determined by the energy balance between the power-gain density due to Joule heating and the power-loss density due to electron-phonon scattering. As a result, the electron temperature becomes higher than the lattice temperature if the lattice is in thermal equilibrium with a heat bath and the drift velocity is large.

For the situation under consideration in this paper, a weak pump laser first excites electrons from the valence bandedge to the conduction bandedge. The excited carriers instantaneously form a nonequilibrium distribution.[13] It is well known that the quantum kinetics of the scattering of electrons with phonons or other carriers under a weak pump field can only be seen within the time scale of several hundred femtoseconds.[14] Subsequently, ultrafast carrier-phonon

and carrier-carrier scattering quickly adjusts the kinetic energies of these excited carriers by taking energy from the lattice.[15] As a result, a quasi-equilibrium Fermi-Dirac distribution of carriers is formed in about 0.1 ps,[13] with an electron temperature determined by the pump-field intensity, pump-photon energy, and lattice temperature. After a few tens of nanoseconds, radiative decay of the excited carriers will begin to affect the electron distribution. The electron temperature will be adiabatically readjusted according to an energy balance between the power-gain density due to optical absorption, the power-loss density due to photoluminescence, and the power-exchange density due to scattering with phonons. At the same time, the lattice temperature will evolve because of an imbalance between the power loss due to transferring phonon energy to carriers and the slight power gain from the external thermal radiation. Just before the radiative decay occurs, the lattice and the electrons are in thermal equilibrium with an initial temperature which can be determined by solving a semiconductor Bloch equation[15]

Using the nonlocal theory described below, we find that the laser-cooling rate is largest for a large bandgap material, a weaker pump-laser field, and a high initial lattice temperature. We also find that the laser-cooling power decreases as the lattice cools down.

In general, the power gain by electrons from the absorption of the pump laser cannot be balanced by the power loss due to spontaneous photon emission alone. As a result, electrons either take energy from or give energy to phonons through inelastic scattering, which depends on the sign of the difference between the electron and lattice temperatures. The electron temperature can be determined by an energy-balance equation for any given lattice temperature. The pair scattering between electrons due to the Coulomb interaction conserves the total energy and does not contribute to the energy-balance equation. On the other hand, the single-particle electron-phonon scattering greatly contributes to the energy-balance equation.

The power-density loss due to spontaneous photon emission is calculated to be[16,17]

$$\begin{aligned}\mathcal{W}_{\mathrm{sp}} &= \frac{\sqrt{\epsilon_{\mathrm{r}}}e^2}{\pi\hbar^2 m_0\epsilon_0 c^3 V}\left(\frac{m_0}{m_{\mathrm{e}}^*}-1\right)\frac{E_{\mathrm{G}}(E_{\mathrm{G}}+\Delta_0)}{E_{\mathrm{G}}+2\Delta_0/3}\\ &\quad\times\sum_{\vec{k}}\left[E'_{\mathrm{G}}(k)+\frac{\hbar^2k^2}{2\mu^*}\right]^2 f_k^{\mathrm{e}}f_k^{\mathrm{h}}\\ &= \frac{\sqrt{\epsilon_{\mathrm{r}}}e^2}{2\pi^3\hbar^2 m_0\epsilon_0 c^3}\left(\frac{m_0}{m_{\mathrm{e}}^*}-1\right)\frac{E_{\mathrm{G}}(E_{\mathrm{G}}+\Delta_0)}{E_{\mathrm{G}}+2\Delta_0/3}\\ &\quad\times\int_0^\infty dkk^2\left[E'_{\mathrm{G}}(k)+\frac{\hbar^2k^2}{2\mu^*}\right]^2 f_k^{\mathrm{e}}\,f_k^{\mathrm{h}},\end{aligned}\tag{1}$$

where $f_k^{\mathrm{e}}$ ($f_k^{\mathrm{h}}$) is the quasi-equilibrium electron (hole) distribution at electron (hole) temperature $T_{\mathrm{e}}$ ($T_{\mathrm{h}}$), $E_{\mathrm{G}}$ is the bandgap, $\Delta_0$ is the spin-orbit splitting, $m_{\mathrm{e}}^*$ is the electron effective mass, $\mu^*$ is the reduced mass of electrons and holes, $\epsilon_{\mathrm{r}}$ is the average relative dielectric constant, $\mathcal{V}$ is the volume of the sample, and $E'_{\mathrm{G}}(k)$ is the renormalized bandgap. It is clear from Eq. (1) that the larger the bandgap or the higher the carrier temperature, the stronger the power-loss density will be.

The power-density gain due to pumping by a spatially-uniform laser for $\hbar\Omega_{\mathrm{p}}\geqslant E_{\mathrm{G}}$ is found to be[18]

$$\begin{aligned}\mathcal{W}_{\mathrm{p}} &= \frac{4}{\hbar\mathcal{V}}\sum_{\vec{k}}\Delta_k^2\left[E'_{\mathrm{G}}(k)+\frac{\hbar^2k^2}{2\mu^*}\right](1-f_k^{\mathrm{e}}-f_k^{\mathrm{h}})\\ &\quad\times\frac{\gamma_0}{\gamma_0^2+[E'_{\mathrm{G}}(k)+\hbar^2k^2/2\mu^*-\hbar\Omega_{\mathrm{p}}]^2}\\ &= \frac{2}{\pi^2\hbar}\int_0^\infty dkk^2\Delta_k^2\left[E'_{\mathrm{G}}(k)+\frac{\hbar^2k^2}{2\mu^*}\right](1-f_k^{\mathrm{e}}-f_k^{\mathrm{h}})\\ &\quad\times\frac{\gamma_0}{\gamma_0^2+[E'_{\mathrm{G}}(k)+\hbar^2k^2/2\mu^*-\hbar\Omega_{\mathrm{p}}]^2},\end{aligned}\tag{2}$$

where $\gamma_0$ is the homogeneous level broadening, $\hbar\Omega_p$ is the pump-photon energy, and $2\Delta_k$ is the renormalized Rabi splitting which is proportional to the square root of the pump-laser intensity. It is seen from Eq. (2) that the greater the pump-laser field, the higher the power-gain density will be. By comparing Eqs. (1) and (2), we find that the ratio of the power-loss density to the power-gain density scales as $E_{\mathrm{G}}^3/\mathcal{E}_{\mathrm{p}}^2$. For simplicity, we do not include recapture of photoluminescence photons here, which can be equivalently included as an adjustment of the pump-laser intensity if the carrier temperature is much smaller than $E_{\mathrm{G}}/k_{\mathrm{B}}$.

By keeping only the leading order interaction between electrons and phonons or impurities in the Heisenberg equation, we get the following power-exchange density $\mathcal{W}_{\mathrm{s}}^{\mathrm{e}}$ from impurities, phonons and scattering-assisted photons to electrons,[19,20]

$$\begin{aligned}\mathcal{W}_{\mathrm{s}}^{\mathrm{e}} &= 2\pi n_{\mathrm{i}}\sum_{\vec{q}}|U_{\mathrm{imp}}^{\mathrm{e}}(q)|^2\sum_{n=-\infty}^{\infty}\left[J_n\left(\frac{eq_{\parallel}\mathcal{E}_{\mathrm{p}}}{m_{\mathrm{e}}^*\Omega_{\mathrm{p}}^2}\right)\right]^2 n\Omega_{\mathrm{p}}\\ &\quad\times\sum_{\vec{k}}(f_k^{\mathrm{e}}-f_{|\vec{k}+\vec{q}|}^{\mathrm{e}})\delta(E_{|\vec{k}+\vec{q}|}^{\mathrm{e}}-E_k^{\mathrm{e}}-n\hbar\Omega_{\mathrm{p}})\\ &\quad-\frac{4\pi}{V}\sum_{\vec{q},\lambda}|C_{q\lambda}^{\mathrm{e}}|^2\sum_{n=-\infty}^{\infty}\left[J_n\left(\frac{eq_{\parallel}\mathcal{E}_{\mathrm{p}}}{m_{\mathrm{e}}^*\Omega_{\mathrm{p}}^2}\right)\right]^2(\omega_{q\lambda}-n\Omega_{\mathrm{p}})\\ &\quad\times\left[N_0^{\mathrm{ph}}\left(\frac{\hbar\omega_{q\lambda}}{k_{\mathrm{B}}T_{\mathrm{L}}}\right)-N_0^{\mathrm{ph}}\left(\frac{\hbar\omega_{q\lambda}-n\hbar\Omega_{\mathrm{p}}}{k_{\mathrm{B}}T_{\mathrm{e}}}\right)\right]\\ &\quad\times\sum_{\vec{k}}(f_k^{\mathrm{e}}-f_{|\vec{k}+\vec{q}|}^{\mathrm{e}})\delta(E_{|\vec{k}+\vec{q}|}^{\mathrm{e}}-E_k^{\mathrm{e}}+\hbar\omega_{q\lambda}-n\hbar\Omega_{\mathrm{p}}),\end{aligned}\tag{3}$$

which includes the phonon- or impurity-assisted photon absorption for $n\neq 0$. Here, $\mathcal{E}_p$ is the pump-laser strength, $E_k^{\mathrm{e}}$ is the renormalized electron kinetic energy, and $T_{\mathrm{e}}$ and $T_{\mathrm{L}}$ are the electron and lattice temperatures, $q_{\parallel}$ lies in the polarization direction of the pump-laser field, $N_0^{\mathrm{ph}}(x)=[\exp(x)-1]^{-1}$ is the Bose-Einstein function, $n$ is an integer, $J_n(x)$ is the *nth* order Bessel function, $\hbar\omega_{q\lambda}$ is the phonon energy for phonon wave number $q$ and mode $\lambda$, $n_{\mathrm{i}}$ is the impurity concentration, $|U_{\mathrm{imp}}^{\mathrm{e}}(q)|^2=|e^2/[\epsilon_0\epsilon_{\mathrm{r}}(q^2+\Lambda_{\mathrm{e}}^2)\mathcal{V}]|^2$ is the electron-impurity

coupling strength, $1/\Lambda_e$ is the static screening length, and $|C^e_{q\lambda}|^2$ is the electron-phonon coupling strength. For polar semiconductors, such as $Al_xGa_{1-x}N$, there exist both acoustic and optical phonon modes. For optical phonon modes, only the longitudinal-optical phonon mode can strongly couple to electrons. For acoustic phonon scattering, on the other hand, we use the deformation-potential approximation [21] with parameters given in the text. The detailed form of electron-phonon coupling strength $|C^e_{q\lambda}|^2$ can been found from publications.[21,22] Applying the Debye model to low-energy acoustic phonons, we get $\omega_{q\lambda}=c_\lambda q$ with $\lambda=\ell,t$. For holes we get the similar power-exchange density $\mathcal{W}^h_s$ with $T_h$. It is clear from Eq. (3) that the electron energy loss or gain from phonons under a weak pump field depends on whether the electron temperature is higher or lower than the lattice temperature, respectively. The same argument applies to holes. The detailed balance between interacting electrons and holes for any given lattice temperature gives rise to a uniform carrier temperature.

In order to cool the lattice, the power gain of the electrons due to laser pumping must be smaller than the power loss due to spontaneous photon emission. This requires a very weak laser field and a large bandgap. The energy conservation in steady state requires

$$\mathcal{W}_{ab}-\mathcal{W}_{sp}+\mathcal{W}^e_s+\mathcal{W}^h_s=0. \tag{4}$$

The solution of this equation provides the carrier temperature for any given lattice temperature $T_L$ since $\mathcal{W}^e_s+\mathcal{W}^h_s$ explicitly depends on the lattice temperature $T_L$. The sign of $\mathcal{W}_{ab}-\mathcal{W}_{sp}$ determines the signs of $T_e-T_L$ and $T_h-T_L$. The larger the value of $|\mathcal{W}_{ab}-\mathcal{W}_{sp}|$, the larger will be the deviation of the carrier temperature from $T_L$.

Although the phonons also stay in a quasi-equilibrium state, the phonon temperature $T_L$ directly evolves with time due to an imbalance between the power loss to electrons and holes and the power gain from any external thermal source (such as the background thermal radiation). As a result, the average phonon energy varies with time. This gives rise to

$$\frac{\hbar^2}{8\pi^2 k_B T_L^2}\left(\frac{dT_L}{dt}\right)\sum_\lambda\int_0^{\pi/a_L} dq q^2\omega^2_{q,\lambda}\sinh^{-2}\left(\frac{\hbar\omega_{q\lambda}}{2k_BT_L}\right)$$
$$=\frac{\sigma\mathcal{A}_s}{\mathcal{V}}(T_B^4-T_L^4)-(\mathcal{W}_{sp}-\mathcal{W}_{ab}), \tag{5}$$

where $a_L$ is the lattice constant, $\sigma=\pi^2k_B^4/60\hbar^3c^2$ is the Stefan-Boltzmann constant, and $\mathcal{A}_s$ is the surface area of the sample. We assume $T_L=T_0$ at $t=0$, where $T_0$ is the initial temperature of equilibrium phonons, and $T_B$ is the environmental temperature which is close to $T_0$ for band edge pumping with very weak laser field. The first term in Eq.(5) is much smaller than the second term even when $T_L\neq T_B$. The rate of reduction of $T_L$ is determined by $\mathcal{W}^e_s+\mathcal{W}^h_s$ which decreases with decreasing $T_L$ and the temperature difference $T_L-T_e$. Moreover, we note that the bandgap of semiconductors in general depends on the lattice temperature, but can be neglected for wide bandgap semiconductors such as $Al_xCa_{1-x}N$. From Eq. (5) we know that the cooling of the lattice implies $\mathcal{W}^e_s+\mathcal{W}^h_s=\mathcal{W}_{sp}-\mathcal{W}_{ab}>0$. This requires $T_e<T_L$ ($T_h<T_L$ for hole) from Eq. (3) with a weak pump field and a large bandgap.

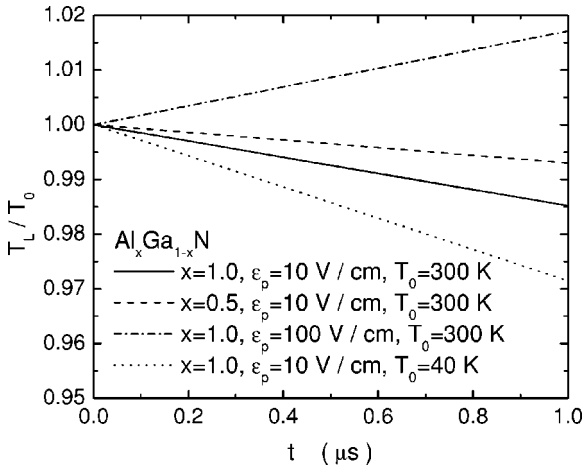


FIG. 1. Calculated scaled lattice temperature $T_L/T_0$ for $Al_xGa_{1-x}N$ as a function of time $t$ with $\hbar\Omega_p-E_G=10$ meV for four different cases. These cases include $x=1$ and $\mathcal{E}_p=10$ V/cm, $T_0=300$ K (solid curve); $x=0.5$ and $\mathcal{E}_p=10$ V/cm, $T_0=300$ K (dashed curve); $x=1$ and $\mathcal{E}_p=100$ V/cm, $T_0=300$ K (dash-dotted curve); and $x=1$, $\mathcal{E}_p=10$ V/cm, $T_0=40$ K (dotted curve). The other parameters are given in the text.

In this paper, we consider the semiconductor $Al_xGa_{1-x}N$ for our numerical calculations, where $x$ is the percentage of Al in the alloy. The bandgap increases with $x$.

For $Al_xGa_{1-x}N$, we choose the following parameters: $E_G=3.4+2.8x$ eV, $m^*_e/m_0=0.2+0.2x$, $m^*_h/m_0=1.4+2.13x$, $\Delta_0=0.02-0.001x$ eV, $\epsilon_s=8.9-0.4x$, $\epsilon_\infty=5.35-0.58x$, $\hbar\omega_{LO}=91.2+8.0x$ meV, $\rho=6.15-2.92x$ g/cm$^3$, $D=-(8.3+1.2x)$eV, $h_{14}=(2.81+4.09x)\times10^7$ V/cm, $c_t=(2.68+1.02x)\times10^5$ cm/sec, $c_\ell=(6.56+2.56x)\times10^5$ cm/sec, $a_L=5.12-0.14x$, $\gamma_0=\hbar/\tau$ with $\tau=0.1$ ps, $n_i=10^{10}$ cm$^{-3}$, $\epsilon_r=(\epsilon_s+\epsilon_\infty)/2$, $\hbar\Omega_p-E_G=10$ meV, and the sample is assumed to be cubic with an edge size of 1 cm.

Figure 1 displays our main results for the scaled lattice temperature $T_L/T_0$ as a function of time $t(0\leqslant t\leqslant 1\ \mu s)$ for $Al_xGa_{1-x}N$. From it we find that the laser cooling at $T_0=300$ K, $x=1.0$ and $\mathcal{E}_P=10$ V/cm (solid curve) is the largest compared to the other three cases, reaching as high as $k_B\Delta T_L/\Delta t=k_B(T_0-T_L)/\Delta t=380$ eV/s. The cooling effect becomes smaller when x is reduced to 0.5 (dashed curve) with a smaller bandgap. Moreover, the laser cooling changes into laser heating when $\mathcal{E}_P$ is increased to 100 V/cm (dash-dotted curve). Finally, the laser-cooling efficiency decreases to 100 eV/s when $T_0$ drops to 40 K (dotted curve). This indicates that the laser cooling of a lattice can be maximized for wide-bandgap semiconductors under the conditions of low pump-field strength and high initial lattice temperature.

In conclusion, by using the energy-balance equation for pump-laser induced conduction electrons and holes, we have demonstrated a laser-cooling power as high as 380 eV/s for the wide bandgap semiconductor AlN at room temperature when the pump-laser field is only 10 V/cm. The evolution of

the lattice temperature was found to be determined by the difference between the power-loss density due to photoluminescence and the power-gain density due to optical absorption, as well as the initial lattice temperature.

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