Large Excitonic Enhancement of Optical Refrigeration in Semiconductors

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We present a theoretical analysis for laser cooling of bulk GaAs based on a microscopic many-particle theory of absorption and luminescence of a partially ionized electron-hole plasma. Our cooling threshold analysis shows that, at low temperatures, the presence of the excitonic resonance in the luminescence is essential in competing against heating losses. The theory includes self-consistent energy renormalizations and line broadenings from both instantaneous mean-field and frequency-dependent carrier-carrier correlations, and it is applicable from the few-Kelvin regime to above room temperature.

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The conceptual foundation of optical refrigeration via luminescence up-conversion was laid in 1929 in Ref. [1]. The system to be cooled is pumped with a narrow-band light beam. In the subsequent thermalization, the absorbed light energy is redistributed among the system's excited states before the system deexcites by luminescing. If the pump (absorption) frequency can be chosen to be below the mean luminescence frequency, the system loses energy in each photon cycle and, thus, cools. Experiments using systems that have optical properties similar to those of molecules (namely, rare-earth doped glass and fluid solutions of laser dyes) have shown significant temperature changes (see, e.g., [2–6]).

It would be desirable to establish laser cooling of semiconductors in which excitonic resonances replace the atomic resonances in doped glass, but no net cooling of a semiconductor sample has been observed yet. An early experimental study suggested the possibility of local semiconductor cooling [7], and several theoretical studies have addressed various aspects of semiconductor cooling (see, e.g., [8–11]). Extending the foundations laid in Refs. [8,9,12], Sheik-Bahae and Epstein [10] developed a very general theoretical framework for analyzing the cooling characteristics of bulk GaAs (the SB-E model). It is based on rate equations for the electron-hole (e-h) density and the net optical power deposited in the semiconductor. General physical arguments and hypotheses rather than a microscopic theory were used in Ref. [10] to extract cooling criteria.

It is the purpose of this Letter to provide analysis and understanding of semiconductor cooling criteria based on a microscopic theory for the luminescence and absorption spectra. The microscopic processes underlying the cooling operation are influenced (i) by the appearance of excitonic resonances in the absorption and luminescence spectra, (ii) by many-particle effects such as excitonic correlations, Pauli blocking, and Coulomb screening, and (iii) possibly also by temperature-dependent (mostly phonon-induced) line broadening. In order to analyze the temperature dependence of the cooling criteria over a wide range of temperatures (from the few-Kelvin regime up to several hundred Kelvins), our theory needs to incorporate all of those effects. We note that, for bulk GaAs, there is a vast literature on plasma effects (see, for example, [13], and references therein), but very little is known about the role of excitonic populations [14–16]. Our theory combines both plasma and excitonic populations effects, allowing us to determine the effects of a partially ionized plasma on the cooling characteristics. In the following, we outline the central features of our theory and then focus on discussing the main results. Details of the theory will be presented elsewhere.

For stationary cooling conditions, we assume that the semiconductor can be modeled under the assumption of quasithermal equilibrium at temperature T and e-h pair density N [17]. We use a model with two parabolic bands. Our theory describes a mixed e-h plasma and exciton gas, similiar to Ref. [15]. We use a real time Green's function formalism with a dynamical T-matrix approximation for the two-particle (e-h, e-e, and h-h) Green's functions. For the single-particle self-energies, the theory contains ladder-type (T-matrix) as well as quasistatically screened Hartree-Fock (HF) and Coulomb-hole (CH) [19] contributions. We calculate self-consistently the optical absorption spectrum and the chemical potential $\mu = \mu_e + \mu_h$, where μ_e (μ_h) are the chemical potentials for the interacting electrons (holes). The e and h spectral functions entering the calculations of the chemical potentials contain features due to excitonic as well as plasma-induced renormalizations. The Coulomb potential entering all self-energies and T matrices is screened mainly by the unbound carriers (plasma). We specify below our estimate of the plasma fraction, which is used to determine the screening within the quasistatic plasmon-pole approximation [19].

We define an ionization degree as N_{free}/N, where N_{free} is approximated by the density of a hypothetical noninteracting plasma at the same chemical potential as that obtained for the full density N. The single-particle energies in the hypothetical plasma include only HF and CH shifts. In the low-density limit, our numerical results are in agreement

with the mass-action law given by the well-known Saha equation and its generalization, the Beth-Uhlenbeck formula [20] (see also [21–23]). In the high-density limit, our theory yields complete exciton ionization, which is often referred to as the Mott transition.

While it is appropriate to use the full T-matrix self-energies for the calculation of the ionization degree, there are indications that the corresponding (excitonic) vertex corrections may be important for the optical spectra [14]. However, such vertex corrections are not presently numerically feasible. Since it is reasonable to expect that contributions from excitonic resonances to the e-h T-matrix self-energy are partially cancelled by the vertex corrections, we compare the final results (see below, Fig. 3) using the full theory with those obtained when using only the second-order self-energy (which does not contain an excitonic resonance) for the calculation of the optical spectra. We believe the true result to be between these two sets of data.

We account for phonon-induced and other broadening effects (present even at zero e-h density) by using phenomenological line shapes consistent with experimental data [18]. We note that a Lorentzian exciton absorption line shape decays too slowly to yield a well-defined mean luminescence frequency. Our chosen line shape model has a more realistic exponential low-frequency tail. The temperature-dependent zero-density bandgap renormalization is also included using literature values [19].

Once the absorption spectrum [$\alpha(\omega)$] and chemical potential (μ) are known as functions of temperature and density, we obtain the luminescence spectral density using the Kubo-Martin-Schwinger (KMS) relation [14]

$$R(\omega) = \left(\frac{\omega n_b}{\pi c}\right)^2 \alpha(\omega) g(\omega), \tag{1}$$

where $g(\omega)$ is the Bose function $g(\omega) = (e^{(\hbar\omega-\mu)/k_BT} - 1)^{-1}$ and n_b is the refractive index. We stress that the chemical potential is strongly affected by interaction effects (both Coulomb interaction and related exciton effects as well as phonon-induced line broadening). The KMS relation implies that, for any nonzero e-h density, $R(\omega) \geq 0$ requires the absorption spectrum to cross over into gain for frequencies below μ, provided that there are optically active states at these frequencies. We note that the pump frequency (ω_a) must be in the absorption region ($\omega_a > \mu$), and the up-conversion $\Delta\omega = \omega_l - \omega_a$ (where ω_l is the mean luminescence frequency) must be positive for cooling to be possible. In other words, a necessary cooling condition is $\mu < \omega_a < \omega_l$.

Figure 1 shows, as an example, the absorption and corresponding luminescence spectra calculated for $T = 30$ K and a pair density of $N = 3 \times 10^{16}$ cm^{-3}. The arrows show the pump frequency optimized for cooling (discussed below) and the mean luminescence frequency. Clearly, in this case the above necessary condition of cooling is fulfilled. For a detailed analysis of sufficient conditions for cooling, we extend the approach of Ref. [10]. There, it was shown that cooling can occur only if the up-conversion dominates all other loss (heating) mechanisms, including nonradiative recombination, luminescence reabsorption, Auger recombination, free-carrier absorption, and other parasitic absorption processes.

The net power imparted to the system is given by

$$P_{\text{net}} = I[\alpha(\omega_a, N) + \alpha_b + \sigma_{\text{fca}} N] - \hbar\omega_l \eta_e B(N) N^2, \tag{2}$$

where, in the steady state,

$$\frac{dN}{dt} = \frac{\alpha(\omega_a, N)}{\hbar\omega_a} I - AN - \eta_e B(N) N^2 - CN^3 = 0. \tag{3}$$

Here I is the intensity of the pump laser, α_b is the (parasitic) background absorption, $\sigma_{\text{fca}} N$ is the free-carrier absorption, AN is the nonradiative recombination rate, η_e is the extraction efficiency (the probability that an emitted photon escapes from the semiconductor without being reabsorbed), $B(N)N^2$ is the radiative recombination rate, and CN^3 is the Auger recombination rate [10]. Cooling requires $P_{\text{net}} < 0$. In an experiment, T, A, C, η_e, α_b, and σ_{fca} are fixed parameters, while the intensity (and thereby N) and ω_a are variables that can easily be adjusted. A useful cooling criterion defined in Ref. [10] is the break-even A_b, which marks the boundary at which cooling becomes possible [$P_{\text{net}}(A_b) = 0$]. This boundary depends on all parameters and variables. In the following discussion, we fix all parameters ($\sigma_{\text{fca}} = 10^{-20}$ cm^2, $\alpha_b = 1$ cm^{-1} [24], $\eta_e = 0.25$, a temperature-dependent C taken from Ref. [10], and T values specified below). We then use a numerical maximization to find the optimal A_b over the space of all densities and absorption frequencies, $\bar{A}_b = \max A_b(N, \omega_a)$. We denote the density and absorption frequency necessary to achieve the maximum A_b (denoted by $\bar{A}_b$) by $\bar{N}$ and $\bar{\omega}_a$, respectively. Physically, $\bar{A}_b$ represents a condition on the sample quality that would be needed in an experiment given the fixed parameter values listed above.

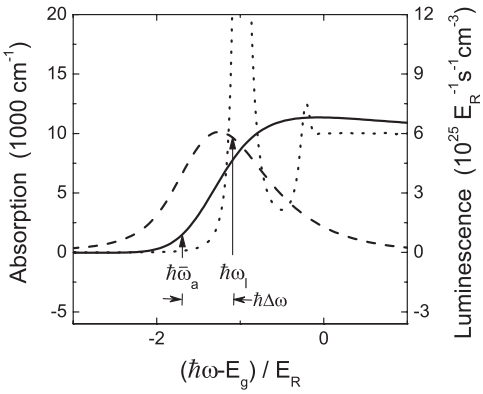

FIG. 1. The absorption (solid line, left scale) and luminescence (dashed line, right scale) calculated for $T = 30$ K at the optimal cooling density of 3×10^{16} cm^{-3} (E_g = band gap, E_R = 4.2 meV). The mean luminescence frequency ω_l and optimal pump frequency $\bar{\omega}_a$ are indicated. For reference, the low-density absorption is also shown (dotted line).

If $\Delta\omega$ and $B(N)$ are density-independent, then increasing the density allows the radiative recombination (cooling) to dominate over the nonradiative recombination (heating), but Auger recombination (heating) limits the maximum density [see Eq. (3)]. At high temperatures, the system operates in this Auger-limited cooling region. At low temperatures, Auger recombination becomes unimportant. However, that does not imply $\bar{N}$ would go to infinity, because, as shown below, realistic theories predict that band filling would limit $\bar{N}$. This is because, at high densities, strong band filling (and, thus, strong optical gain) causes the mean luminescence frequency ω_l to be in the gain region, thus violating the necessary cooling condition $\mu < \bar{\omega}_a < \omega_l$ [25].

We plot in Fig. 2 the radiative recombination coefficient B calculated with the present theory as functions of density for various temperatures ($\bar{N}$ marked by stars). Reference [10] used a high-temperature estimate for B obtained in a noninteracting, nondegenerate particle model: B is density-independent with a $T^{-3/2}$ temperature dependence. Our results in Fig. 2 show rather strong deviations from this behavior at low temperatures ($\lesssim$100 K) and/or high densities. According to the figure, B decreases as density grows because a finite exciton population fraction and Pauli blocking of single-particle orbitals both tend to drive the density dependence of the luminescence rate to $\sim N$ instead of $\sim N^2$. In the low-density limit, our calculated B does approach a constant [28]. We note that in our theory the temperature dependence of the low-density B values increases faster than $\sim T^{-3/2}$ as $T \downarrow 0$.

In Fig. 3, we show results for the optimized break-even nonradiative lifetime (defined as $\bar{\tau}_b = 1/\bar{A}_b$) as a function of temperature. The solid and dotted lines show the results of the full calculation and the second-order approximation defined above, respectively. The overall temperature dependence is similar for these two curves [29], suggesting that many-body correlation effects beyond the T-matrix approximation would probably not change the results significantly. To better understand our results, we also show $\bar{\tau}_b$ for several simplified models: (i) the SB-E theory (dashed line) but with slightly different parameter values from Ref. [10], $B(300\ \mathrm{K}) = 4.69 \times 10^{-10}\ \mathrm{s}^{-1}\,\mathrm{cm}^{-3}$, $\hbar\Delta\omega = 1.55 k_B T$, and neglecting background absorption [30], (ii) the noninteracting fermionic electron-hole model ("free e-h theory," dashed-dotted-dotted line), and (iii) same as the full interacting model, but without any T-matrix self-energies ("plasma theory," dashed-dotted line).

For each model, the predicted cooling region is above the corresponding $\bar{\tau}_b$ curve in Fig. 3. Above room temperature, all models show an increase of $\bar{\tau}_b$ (i.e., cooling becomes more difficult) with temperature. In this regime, Pauli blocking becomes negligible, and the free e-h (dashed-dotted-dotted line) and SB-E (dashed line) theories give converging results, both suffering from the $T^{-3/2}$ downward trend of the radiative recombination coefficient. Figure 3 shows that, with decreasing temperature, the SB-E theory predicts a vanishing $\bar{\tau}_b$. This is because the SB-E theory neglects band filling and background absorption. Without band filling, Auger recombination is the limiting process for $\bar{N}$. Since C decreases with temperature [10], $\bar{N}$ can become large and the radiative recombination dominates over nonradiative recombination. The free e-h theory adds band filling and background absorption effects to the SB-E theory. For $T \lesssim 200$ K, band filling limits $\bar{N}$. To maintain a positive up-conversion ($\mu < \bar{\omega}_a < \omega_l$), $\bar{N}$ decreases with decreasing T, entering the density regime where the nonradiative recombination dominates. Thus, $\bar{\tau}_b$ increases with decreasing T (dashed-dotted-dotted line). At even lower temperatures ($T \lesssim 30$ K), background absorption becomes important. Indeed, for $T \lesssim 15$ K, the free e-h theory predicts that the up-conversion becomes too small to overcome the background losses, making cooling impossible.

Finally, the three models including Coulomb effects yield $\bar{\tau}_b$'s significantly lower than the "free e-h" value, showing a dramatic improvement of the refrigeration criterion at low temperatures. In particular, cooling is still possible at very low temperatures, in sharp contrast to the free e-h model. This promising behavior is a consequence

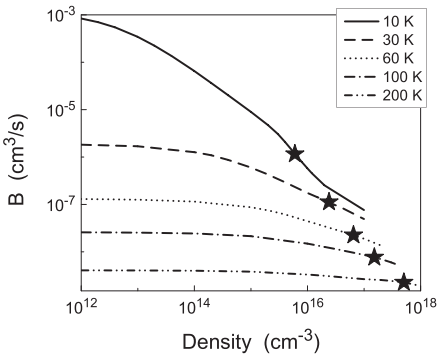

FIG. 2. The density-dependent radiative recombination coefficient for various temperatures. The optimal cooling density is marked with a star for each temperature.

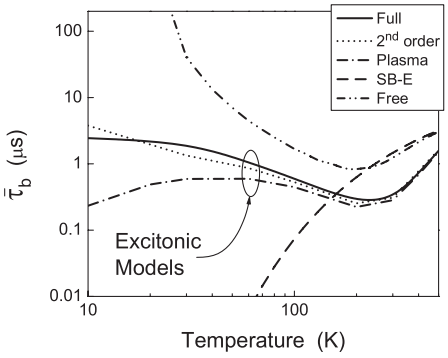

FIG. 3. Break-even nonradiative lifetime vs temperature for the full theory and various simplified models. A smaller $\bar{\tau}_b$ is beneficial to cooling (for each model, the predicted cooling region is above its curve).

of excitonic effects. At all temperatures, the excitonic enhancement of the low-frequency absorption raises B [cf. Eq. (1)] and, therefore, lowers $\bar{\tau}_b$. However, the dramatic improvement of $\bar{\tau}_b$ at low temperatures is due to the Coulomb-induced steepening of the low-energy tail of $\alpha(\omega)$. This can be shown to help overcome background absorption much more effectively than in the case without excitonic effects. We note that, in the free e-h theory, at low temperatures, $\bar{\tau}_b$ is increasingly sensitive to the level of α_b. For $\alpha_b = 0$, cooling becomes possible in the free e-h theory at temperatures even well below 20 K and $\bar{\tau}_b$ approaches roughly a T^{-1} behavior (see also [26]). In contrast, the prediction of the excitonic models are relatively insensitive to α_b. Among the excitonic models, the plasma theory modestly underestimates $\bar{\tau}_b$. This is due to the neglect of excitonic correlation contributions to the single-particle spectrum, which increases μ (but not enough to reduce the up-conversion) and, consequently, B.

We found that $\bar{N}$ is generally high enough so that neglecting phonon-induced broadening does not alter the results significantly. We finally note that our excitonic results shown in Fig. 3 support the conjecture in Ref. [10] that cooling should be easier to achieve at a reduced starting temperature, because the temperature dependence of typical nonradiative lifetimes $\tau = 1/A$ have been found to increase exponentially with decreasing temperature, whereas our calculation shows a subexponential increase of $\bar{\tau}_b = 1/\bar{A}_b$. If, therefore, at a given temperature a sample has a τ just above $\bar{\tau}_b$ (i.e., it barely admits cooling), reducing the temperature will increase its τ well above $\bar{\tau}_b$, which makes cooling easier.

In conclusion, we have developed a comprehensive theory for semiconductor laser cooling based on a microscopic theory for a partially ionized exciton gas in a bulk semiconductor. The theory is valid for a wide range of temperatures. We found that the cooling criterion expressed as a break-even condition for the nonradiative lifetime required for cooling ($\bar{\tau}_b$) is strongly affected by excitonic effects and that at low temperatures these effects lead to a dramatic reduction in $\bar{\tau}_b$. In the future, it would be desirable to study extensions of our theory, such as band structure effects beyond the parabolic two-band model, possible nonquasithermal equilibrium effects (e.g., kinetic holes in the center-of-mass distribution of excitons [31]) and polariton effects.

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