

# Radiation thermodynamics with applications to lasing and fluorescent cooling

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Laser cooling of bulk matter uses thermally assisted fluorescence to convert heat into light and can be interpreted as an optically pumped laser running in reverse. Optical pumping in such devices drives the level populations out of equilibrium. Nonthermal radiative energy transfers are thereby central to the operation of both lasers and luminescent coolers. A thermodynamic treatment of their limiting efficiencies requires a careful development of the entropy and effective temperatures of radiation, valid for the entire range of light from the blackbody to the ideal laser limiting cases. In particular, the distinct meaning and utility of the brightness and flux temperatures should be borne in mind. Numerical examples help illustrate these concepts at a level suitable for undergraduate physics majors. © 2005 American Association of Physics Teachers.

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## I. INTRODUCTION

Energy transfers between thermodynamic systems are often labeled as either work or heat.<sup>1</sup> However, it is generally recognized that the exchange of electromagnetic radiation cannot be neatly categorized as one or the other. Exceptions are blackbody radiation, which is one of the three traditional forms of heat transfer, and ideal laser radiation which can be characterized as work by the Carathéodory definition.<sup>2</sup> Some authors explicitly distinguish radiation as a third category of energy transfer.<sup>3</sup> However there is nothing special about radiation in this regard; any irreversible transfer of energy between two systems generally involves a mixture of heat and work. For example, when a block slides over a rough table, there is both mechanical work involved in the bending of asperities on the surfaces of the block and the table, and heat transfer between portions of the block and table at different temperatures.<sup>4</sup>

The primary reason for delineating heat from work is to introduce the second law of thermodynamics and arrive at the concept of entropy. The limiting efficiency of an engine or refrigerator depends on the entropies of the input and output energy fluxes. This paper explicitly treats photon engines and optical coolers, thereby requiring a robust understanding of the thermodynamic terms that apply to radiation.

## II. BASIC CONCEPTS OF LASER COOLING OF BULK MATTER

Before jumping into the abstractions of radiation entropy and temperature, consider the principles of operation of an optical engine or refrigerator. Figure 1 is a simple block diagram of a laser cooler, presented in the same style that physics textbooks typically use to introduce refrigerators. The left-hand arrow denotes an external source of energy  $E_{in}$ . In a conventional refrigerator this external source does electrical work, but in the present context it is a directed, coherent (and possibly polarized, particularly if the active material is axial) narrowband laser beam.

The refrigerator consists of an optically active medium that resonantly absorbs most, if not all, of the input laser radiation. (The absorption length may be increased either by shaping the material into the form of an optical fiber or by

placing it in an optical cavity tuned to the pump wavelength.) Subsequently the medium relaxes by spontaneous emission at a variety of different possible energy-level transitions, so that the output fluorescence (which carries away energy  $E_{out}$ ) is multidirectional (probably isotropic), incoherent, unpolarized (or at most, partially polarized), and broadened in bandwidth. (This fluorescence must be absorbed by some external heat sink which is thermally insulated from the refrigerator. Care must be taken to minimize reabsorption of the exiting fluorescence, for example by employing a fiber sample geometry that enables the fluorescence to readily escape out its sides.) In ordinary cases of laser absorption,  $E_{out}$  is less than  $E_{in}$ , and the difference appears as an increase in the internal energy of the material, thereby enabling lasers to cut, weld, or ablate substances. However, the crucial and surprising property of certain materials irradiated with lasers of just the right color is that they emit light of a higher frequency (and hence photon energy) than they absorb; this property is known as anti-Stokes or thermally assisted fluorescence. If anti-Stokes emission predominates over resonant and Stokes processes, then heat  $Q$  is withdrawn on average from the medium in each excitation–relaxation cycle.

The simplest possible level scheme of such an absorber is depicted in Fig. 2. The material has only three energy levels, where level 1 is the ground state. The pump laser frequency  $\nu_p$  is tuned to resonance with the 1–2 transition, so that  $E_{in} = h\nu_p$  (times the number of photons absorbed), where  $h$  is Planck's constant. Suppose that  $\nu_p$  is large enough that nonradiative relaxation from the excited states to the ground state is much less likely than radiative decay. In other words, the fluorescence quantum efficiency is essentially unity. According to the energy gap law,<sup>5</sup> this requirement is satisfied if  $\nu_p \gg \nu_{acceptor}$ , where  $\nu_{acceptor}$  is the frequency of any other modes of the substance to which the excited states can couple. For a solid material,  $h\nu_{acceptor} \cong kT$  (where  $k$  is Boltzmann's constant) could represent phonon energies, where  $kT$  is a typical thermal energy at the refrigerator's operating temperature  $T$ . Alternatively, the energy acceptors might be localized vibrational or rotational modes. If we assume that the radiative time constant for relaxation from the excited to the ground states is relatively long, as is typical of fluorescent materials, and that the spacing between levels 2 and 3 is

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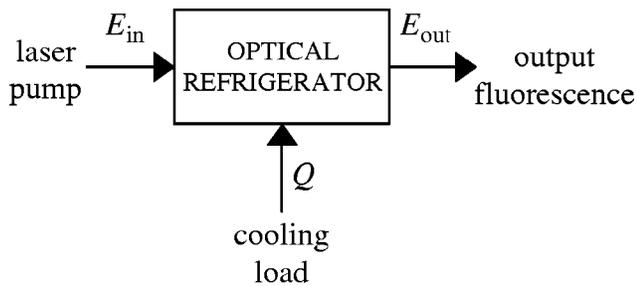


Fig. 1. Block diagram of the input and output energy flows for a laser cooler.

small (compared to  $kT$ ), so that rapid nonradiative exchanges occur between them, then a thermal (Boltzmann) distribution of population between these two excited levels will be established prior to relaxation to the ground state. In that case, some fraction of the active species (viz. ions, atoms, molecules, or electrons) will be thermally promoted into level 3. (The fraction depends on the relative degeneracies and energy spacing of levels 2 and 3.) Consequently, the average fluorescence photon energy  $h\bar{\nu}_f$  will be larger than the laser pump photon energy  $h\nu_p$ , with the difference representing thermal energy withdrawn from the system,  $Q = h(\bar{\nu}_f - \nu_p)$  per cycle.

This cooling energy  $Q$  may be used to chill an external load. Typical proposed applications<sup>6</sup> of optical refrigerators are to cool infrared detectors in satellites (because competing Stirling-cycle compressors introduce undesirable vibrations, and thermoelectric coolers are highly inefficient at the liquid-nitrogen temperatures of interest) and to cool small, specialized electronic or high-temperature-superconductor devices (which can justify the expense of designing an optical cooler). In the laboratory, the cooling load often is simply the absorbing sample itself. In that case, the cooling energy first reduces the temperature of the material (suspended in an optical-access vacuum chamber) and eventually balances the heat load on the sample from the surroundings. In a well-designed system, this load arises primarily from ambient thermal radiation, implying that all three inputs and outputs in Fig. 1 are radiative. Optical heat shields (sometimes called hot mirrors) can be used to further reduce this heat leak. The sides of the active sample are coated with a material that

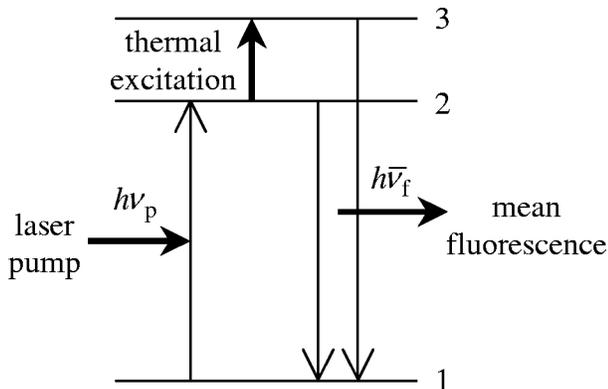


Fig. 2. A possible energy-level scheme of a cooling material. The three states could represent electronic levels of an atom, vibrational levels of a molecule, or levels within the valence and conduction bands of a semiconductor.

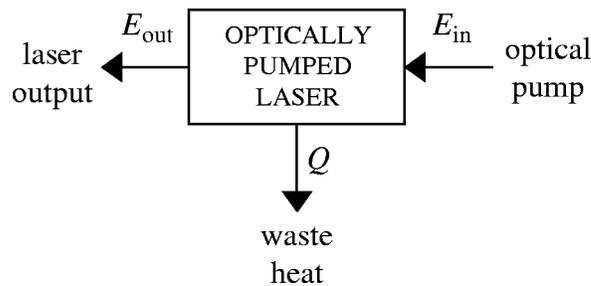


Fig. 3. Thermodynamic diagram analogous to Fig. 1 describing a laser pumped by an external light source.

transmits the fluorescence but reflects the ambient blackbody light (assuming that the fluorescence and the blackbody radiation, which peaks at  $10\ \mu\text{m}$  in the case of 300 K surroundings, are spectrally well separated from one another).

The current record temperature drop for an unshielded solid sample suspended in vacuum starting from room temperature<sup>7</sup> is an impressive  $65\ ^\circ\text{C}$ . This drop was achieved for optical pumping of the rare-earth ion  $\text{Yb}^{3+}$  doped into a heavy-metal fluoride glass (ZBLAN). The advantages of  $\text{Yb}^{3+}$  over other species are threefold: It consists of only two bands of energy levels, thus avoiding excited-state absorption of the pump or fluorescence radiation; the two bands are each split into a number of closely spaced levels, thus permitting efficient absorption of thermal energy from the host; and the energy gap between the two bands is large (corresponding to a near-infrared wavelength of  $1\ \mu\text{m}$ ), thereby minimizing nonradiative decay.<sup>8</sup> Although helpful, these three factors are not required for successful cooling of rare-earth ions, as evidenced by recent results<sup>9</sup> for  $\text{Tm}^{3+}$ . Laser cooling of gaseous carbon dioxide,<sup>10</sup> organic laser dyes in an alcohol solution,<sup>11,12</sup> and gallium arsenide heterostructures<sup>13,14</sup> also have been reported.

The alert reader may notice that these systems (rare-earth doped solids,  $\text{CO}_2$ , solvated organic dyes, and semiconductor heterostructures) also make efficient lasers. The properties required for a material to perform well as an optical cooler (efficient luminescence, large optical cross sections, minimal competing decay channels) also are properties that promote lasing. In fact, one need simply reverse every arrow in Fig. 1 to obtain the optically pumped laser sketched in Fig. 3. The optical pump might now be a flashlamp, which, like the fluorescence in Fig. 1, is broadband, incoherent, undirected, and unpolarized. Some fraction of this radiative energy is converted into laser light and the rest appears as waste heat to be removed say by circulating cooling water.

The realization that good lasers can also make good optical coolers suggests other possible cooling candidates. Both  $\text{Nd}^{3+}:\text{YAG}$  and ruby have been explored for this role,<sup>15,16</sup> although for technical reasons these particular materials have not lived up to their theoretical cooling potential.<sup>17</sup>

### III. REVIEW OF RADIATION THERMODYNAMICS

The rate at which entropy is carried by a steady, unpolarized beam of light (not necessarily collimated) in vacuum across a surface  $A$  is given by

$$\dot{S} = 2kc^{-2} \int_A \int_{\Omega} \int_{\Delta\nu} [(1+n)\ln(1+n) - n \ln n] \nu^2 d\nu \cos \theta d\Omega dA, \quad (1)$$

as discussed in the Appendix, where  $c$  is the speed of light, and  $\theta$  and  $\phi$  are the polar and azimuthal angles, respectively, at which a photon is traveling into element  $d\Omega = \sin \theta d\theta d\phi$  of a solid angle relative to the normal to an element  $dA$  of the surface. (Note that  $A$  could be part or all of an arbitrary cross section of the beam, or it could be a portion of the real surface of a material emitting or absorbing the light.) The radiation is distributed over a set of optical modes with mean occupation number  $n$  that depends on the photon frequency  $\nu$ , the direction of travel  $\theta$  and  $\phi$  of the photon (within a range of solid angle  $\Omega$ ), and two position coordinates of the photon on the surface  $A$  (affording a considerable opportunity for confusion if spherical coordinates are used to describe it). The frequency integration is over the relevant spectral bandwidth  $\Delta\nu$  of the light, where the factor of  $\nu^2$  comes from the density of states. Finally the term in square brackets in Eq. (1) arises from Bose–Einstein statistics. It is crucial to realize that Eq. (1) is valid whether or not the radiation is thermal, that is, even for a nonequilibrium photon distribution such as the fluorescence emission of Fig. 2.

Entropy leads to the concept of radiation temperature. Two definitions of this quantity have been proposed,<sup>18</sup> called the brightness and flux temperatures. Because the energy of a photon is  $h\nu$  and the average number of such photons in a particular beam mode is  $n$ , the replacement of  $k$  times the quantity in square brackets in Eq. (1) by  $nh\nu$  gives the rate at which energy is carried by the beam (that is, its power),

$$\dot{E} = 2hc^{-2} \int_A \int_{\Omega} \int_{\Delta\nu} n\nu^3 d\nu \cos \theta d\Omega dA. \quad (2)$$

From Eq. (2) the spectral radiance (often referred to as brightness,<sup>19</sup> although properly speaking that is a photometric not a radiometric term)  $L_\nu \equiv d\dot{E}/(dA_\perp d\nu d\Omega)$ , where  $dA_\perp \equiv \cos \theta dA$ , is seen to be

$$L_\nu = 2nh\nu^3/c^2. \quad (3)$$

Thus an experimental measurement of the spectral radiance (using an angle-resolved absolute-intensity spectrometer) directly gives the photon occupation numbers  $n$ .

The brightness temperature  $T_B$  of a beam of radiation is defined as the temperature of a blackbody whose spectral radiance integrated over the bandwidth of the radiation is equal to the (integrated) radiance of the beam,

$$\int_{\Delta\nu} n\nu^3 d\nu = \int_{\Delta\nu} \frac{\nu^3}{\exp(h\nu/kT_B) - 1} d\nu, \quad (4)$$

according to Planck's law. Further insight into the meaning of the brightness temperature can be obtained by examining two special cases of radiation. First consider a blackbody emitting isotropically into a hemisphere (via a small hole in a furnace for example). Its energy flux density (or irradiance)  $I_E \equiv d\dot{E}/dA$  is<sup>20</sup>

$$I_E^{\text{BB}} = \frac{2\pi h}{c^2} \int_0^\infty \frac{\nu^3}{\exp(h\nu/kT_B) - 1} d\nu = \sigma T_B^4. \quad (5)$$

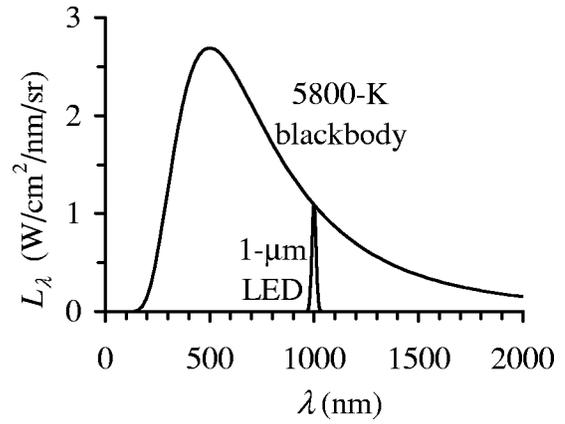


Fig. 4. The spectral radiance of a 5800 K (solar surface temperature) blackbody and a Gaussian source with a peak at  $\lambda_0 = 1 \mu\text{m}$  representing a LED with a hemispherical irradiance of  $1 \text{ W/mm}^2$  and a 25 nm bandwidth ( $w = 10 \text{ nm}$ ). Within this bandwidth, the area under the Planck curve is equal to the area under the Gaussian. We could therefore describe this LED as being as bright as the sun.

Here  $\sigma = 2\pi^5 k^4 / (15c^2 h^3) = 5.67 \times 10^{-8} \text{ W m}^{-2} \text{ K}^{-4}$  is the Stefan–Boltzmann constant.

Likewise its entropy flux density  $I_S \equiv d\dot{S}/dA$  is<sup>18</sup>

$$I_S^{\text{BB}} = \frac{2\pi k}{c^2} \int_0^\infty \left\{ \frac{h\nu/kT_B}{1 - \exp(-h\nu/kT_B)} - \ln[\exp(h\nu/kT_B) - 1] \right\} \times \nu^2 d\nu = \frac{4}{3} \sigma T_B^3. \quad (6)$$

The thermodynamic definition of temperature is

$$\frac{1}{T} \equiv \left. \frac{\partial S}{\partial E} \right|_V. \quad (7)$$

If we apply Eq. (7) to the cavity radiation escaping from the interior of a furnace of volume  $V$ , we find

$$T = \frac{dI_E^{\text{BB}}}{dI_S^{\text{BB}}} = \frac{4\sigma T_B^3 dT_B}{4\sigma T_B^2 dT_B} = T_B. \quad (8)$$

In agreement with Eq. (4), the brightness temperature of blackbody radiation is equal to the absolute temperature of the furnace emitting that radiation. Hence the irradiance of blackbody radiation is only a function of the temperature of its source, and it is in this sense that one describes blackbody radiation as “pure” heat or “low-grade” energy.

Consider next the case of narrowband radiation centered at peak frequency  $\nu_0$ , such as might be emitted by an LED or a real laser. It follows from Eq. (4) that its mean brightness temperature is

$$\bar{T}_B = \frac{h\nu_0}{k \ln(1 + 1/\bar{n})}, \quad (9)$$

where  $\bar{n} = c^2 \bar{L}_\nu / (2h\nu_0^3)$  according to Eq. (3). Here  $\bar{L}_\nu$  is the spectral radiance of the source averaged over its bandwidth  $\Delta\nu$ .

The intuitive meaning of Eq. (9) is easier to grasp from a graph such as Fig. 4. Suppose the source spectrum has a Gaussian distribution

$$L_\lambda = L_{\text{peak}} \exp\left[-\frac{1}{2}\left(\frac{\lambda - \lambda_0}{w}\right)^2\right], \quad (10)$$

where  $L_{\text{peak}}$  is the spectral radiance at the source's peak wavelength  $\lambda_0 = c/\nu_0$  and  $w$  is the spectral width corresponding to one standard deviation. If we define the source's bandwidth as

$$\Delta\lambda \equiv \int_0^\infty g(\lambda) d\lambda, \quad (11)$$

where  $g(\lambda) \equiv L_\lambda/L_{\text{peak}}$  is the normalized profile, then one can show that

$$\Delta\lambda = w\sqrt{2\pi} \approx 2w\sqrt{\ln 2}, \quad (12)$$

which is the full-width at half-maximum of the source's spectrum. It is left as an exercise to verify that

$$L_{\text{peak}} \approx L_\lambda^{\text{BB}}(\lambda_0) = \frac{2hc^2\lambda_0^{-5}}{\exp(hc/k\bar{T}_B\lambda_0) - 1}. \quad (13)$$

That is, to an excellent approximation one can determine the brightness temperature of a narrowband source by plotting its spectral radiance and then finding the blackbody curve which passes through its peak, as in Fig. 4.

For a very bright source, Eq. (9) becomes  $k\bar{T}_B \approx \bar{n}h\nu_0 = \lambda_0^2 \bar{L}_\nu/2$ . For example, an unpolarized 1 mW red helium–neon laser with a beam area of 1 mm<sup>2</sup>, a divergence (half-angle  $\delta$ ) of 0.5 mrad corresponding to a solid angle of approximately  $\pi\delta^2 = 0.8 \mu\text{sr}$ , and a bandwidth of 1 GHz has a mean brightness temperature on the order of  $2 \times 10^{10}$  K.<sup>19</sup> (The detailed variation in the brightness temperature with frequency and angle can easily be deduced for simple profiles such as Gaussians.<sup>21</sup>)

If the narrowband radiation is independent of the angular directions  $\theta$  and  $\phi$  over a circular cone of half-angle  $\delta$ , then its energy flux density is

$$I_E^{\text{NB}} = 2\pi hc^{-2} \bar{n} \nu_0^3 \Delta\nu \sin^2 \delta, \quad (14)$$

and its entropy flux density is

$$I_S^{\text{NB}} = 2\pi kc^{-2} [(1 + \bar{n}) \ln(1 + \bar{n}) - \bar{n} \ln \bar{n}] \nu_0^2 \Delta\nu \sin^2 \delta. \quad (15)$$

The analog of Eq. (8) becomes

$$T = \frac{dI_E^{\text{NB}}}{dI_S^{\text{NB}}} = \frac{h\nu_0 d\bar{n}}{k \ln(1 + 1/\bar{n}) d\bar{n}} = \bar{T}_B \quad (16)$$

using Eq. (9) in the last step. Therefore the brightness temperature remains an absolute thermodynamic temperature for nonequilibrium radiation.

It is worth noting what happens if either the bandwidth  $\Delta\nu$  or divergence  $\delta$  of the source collapses to zero. If its energy flux density is to remain finite, then Eq. (14) implies that  $\bar{n} \rightarrow \infty$ . In this limit, Eq. (15) becomes

$$\begin{aligned} I_S^{\text{NB}} &\approx 2\pi kc^{-2} [(1 + \bar{n}) \ln \bar{n} - \bar{n} \ln \bar{n}] \nu_0^2 \Delta\nu \sin^2 \delta \\ &= \frac{k}{h\nu_0} I_E^{\text{NB}} \frac{\ln \bar{n}}{\bar{n}} \rightarrow 0. \end{aligned} \quad (17)$$

That is, the entropy carried by monochromatic or unidirectional radiation is zero, so that one can characterize an ideal laser beam as “pure” work or “high-grade” energy. Note from Eq. (16), however, that  $\int T dI_S$  remains equal to the

irradiance (just as it did for blackbody radiation) and is not zero, as one might have expected if this integral defined a heat flux density. Entropy and heat are not directly related for nonequilibrium distributions.

To analyze the efficiency of optical devices, it is useful to introduce the flux temperature,

$$T_F \equiv \frac{I_E}{I_S}. \quad (18)$$

Equations (14) and (15) imply for a narrowband source that

$$T_F = \frac{h\nu_0}{k} \frac{\bar{n}}{(1 + \bar{n}) \ln(1 + \bar{n}) - \bar{n} \ln \bar{n}}. \quad (19)$$

If the source is no more than moderately bright, then  $\bar{n} \ll 1$ , and Eq. (19) becomes

$$T_F \approx \frac{h\nu_0}{k} \frac{\bar{n}}{(1)(\bar{n}) - \bar{n} \ln \bar{n}} = \frac{h\nu_0/k}{1 + \ln(1/\bar{n})}. \quad (20)$$

Equations (9) and (20) imply that  $\bar{T}_B/T_F \rightarrow 1$  as  $\bar{n} \rightarrow 0$ , so that the flux and brightness temperatures are equal for dim, narrowband radiation.

In contrast,  $T_F = 0.75T_B$  for blackbody radiation according to Eqs. (5) and (6). At first glance this result is surprising, because it implies that if a large body (of surface area  $A$ ) at temperature  $T$  radiates away a small amount of heat  $Q$  into free space, then the light carries away entropy  $S = 4Q/3T$  according to Eq. (18), rather than  $Q/T$ . This entropy transport appears to disagree with the first equality in Eq. (8), which can be rewritten in this context as  $\dot{S} = \int d\dot{Q}/T$ . But in fact Eq. (8) does not imply that  $S = \int dQ/T = Q/T$ , because  $\dot{Q}$  is only a function of temperature (specifically  $d\dot{Q} = 4\sigma AT^3 dT$ ), whereas  $Q$  is implicitly being interpreted as a function of time at the (nearly) constant temperature of the emitter. The lesson is to exercise caution when relating the fluxes to the total energy and entropy. The light carries away entropy  $4Q/3T$ , while the entropy of the body decreases by  $Q/T$ , and thus the total entropy change of the universe is positive in this irreversible emission process.

On the other hand, we expect entropy to balance if a hot body at temperature  $T + \Delta T$  is radiatively coupled to surroundings at temperature  $T$  that are only infinitesimally smaller in temperature by  $\Delta T$ . In this case, the net rate of radiative energy transport from the hot body to the surroundings is

$$\dot{Q} = \sigma A [(T + \Delta T)^4 - T^4] \approx 4\sigma AT^3 \Delta T, \quad (21)$$

while the net rate of entropy transport is

$$\dot{S} = \frac{4}{3} \sigma A [(T + \Delta T)^3 - T^3] \approx 4\sigma AT^2 \Delta T. \quad (22)$$

This time  $S = Q/T$ , as one would predict from the quasi-equilibrium of the bodies with the radiation.

Returning to our discussion of Eq. (19), for a very bright source ( $\bar{n} \gg 1$ ) we obtain

$$T_F \approx \frac{h\nu_0}{k} \frac{\bar{n}}{(1 + \bar{n}) \ln \bar{n} - \bar{n} \ln \bar{n}} = \frac{h\nu_0}{k} \frac{\bar{n}}{\ln \bar{n}}. \quad (23)$$

For ideal laser radiation,  $\bar{n} \rightarrow \infty$  and thus  $T_F$  becomes infinite, which is consistent with the fact that it carries zero entropy at a finite irradiance. One (but not the only) way to get zero

entropy transport is if the radiation occupies a single optical mode, corresponding to a monochromatic plane wave, so that the multiplicity of its macrostate is unity. Also note that Eqs. (9) and (23) imply that  $\bar{T}_B/T_F \cong \ln \bar{n}$  for a bright narrowband source. That is, the brightness temperature diverges even more rapidly than the flux temperature as  $\bar{n} \rightarrow \infty$ .

So far it has been implicitly assumed that the fluorescent emission in Fig. 1 is into free space. However if the cooling sample is bathed in thermal radiation from the surroundings at ambient temperature  $T_A$ , then one needs to account both for the occupation number  $n$  of fluorescence photons and the distribution  $n_A$  of ambient photons.<sup>22</sup> The net energy and entropy fluxes stem from the difference between the radiation leaving the body (fluorescence plus thermal radiation which is reflected or transmitted, assuming for simplicity that the sample has negligible thermal emissivity) and that incident on the body (thermal radiation).<sup>23</sup> Hence

$$\begin{aligned}\dot{E}_{\text{net}} &= 2hc^{-2} \int (n+n_A) v^3 dv \cos \theta d\Omega dA \\ &\quad - 2hc^{-2} \int n_A v^3 dv \cos \theta d\Omega dA \\ &= 2hc^{-2} \int n v^3 dv \cos \theta d\Omega dA,\end{aligned}\quad (24)$$

so that one recovers Eq. (2). In contrast

$$\begin{aligned}\dot{S}_{\text{net}} &= 2kc^{-2} \int [(1+n+n_A)\ln(1+n+n_A) \\ &\quad - (n+n_A)\ln(n+n_A) - (1+n_A)\ln(1+n_A) \\ &\quad + n_A \ln n_A] v^2 dv \cos \theta d\Omega dA,\end{aligned}\quad (25)$$

which does not simplify to Eq. (1). That is, the entropy carried by the net radiative emission from the refrigerator is not independent of the ambient temperature in general. However for practical optical refrigerators,  $n \gg n_A$ , and Eq. (1) is an excellent approximation to the net entropy flux. It is only in the limit of vanishing fluorescence radiance that the ambient radiation matters. Specifically Eq. (25) can then be approximated as

$$\begin{aligned}\dot{S}_{\text{net}} &\cong 2kc^{-2} \int [(1+n+n_A)\ln(1+n_A) - (n+n_A)\ln n_A \\ &\quad - (1+n_A)\ln(1+n_A) + n_A \ln n_A] v^2 dv \cos \theta d\Omega dA \\ &= 2kc^{-2} \int n \ln(1+1/n_A) v^2 dv \cos \theta d\Omega dA \\ &= \dot{E}_{\text{net}}/T_A\end{aligned}\quad (26)$$

after substituting the Planck expression for  $n_A$  in the last step. Consequently the lower limit of the fluorescence flux temperature  $\dot{E}_{\text{net}}/\dot{S}_{\text{net}}$  is the ambient temperature  $T_A$  as the pump power in Fig. 1 is reduced toward zero. Even if the laser pump is shut off, there remains some weak fluorescence stimulated by the absorption of ambient thermal radiation.

#### IV. LIMITING EFFICIENCIES OF OPTICAL CONVERTERS

The stage is set for a thermodynamic analysis of the efficiency with which the optical refrigerator in Fig. 1 converts heat into light. According to the first law (energy conservation),

$$E_{\text{out}} = E_{\text{in}} + Q. \quad (27)$$

The cooling coefficient of performance is defined in the usual way for a refrigerator as

$$\eta \equiv \frac{Q}{E_{\text{in}}}. \quad (28)$$

The maximum value of  $\eta$  is the Carnot limit,  $\eta_C$ , and is determined by the second law of thermodynamics. The entropy carried away by the fluorescence cannot be less than the sum of the entropy withdrawn from the cooling sample and the entropy transported in by the pump laser,

$$\frac{E_{\text{out}}}{T_f} \geq \frac{Q}{T} + \frac{E_{\text{in}}}{T_p}, \quad (29)$$

where  $T$  is the steady-state operating temperature of the refrigerator, and  $T_f$  and  $T_p$  are the flux temperatures of the fluorescence and pump radiation, respectively. The reversible Carnot limit is obtained by choosing the equality sign in Eq. (29). If one substitutes Eqs. (27) and (28) into Eq. (29), we obtain

$$\eta_C = \frac{T - \Delta T}{T_f - T}, \quad (30)$$

where  $\Delta T \equiv TT_f/T_p$ . Equation (30) can be interpreted as the Carnot coefficient of performance of the engine-driven refrigerator depicted in Fig. 5. The engine extracts heat  $E_{\text{in}}$  from a hot reservoir at temperature  $T_p$  and rejects heat  $E_{\text{out}}^{\text{engine}}$  to a cold reservoir at temperature  $T_f$ . It thus produces work  $W$  with a Carnot efficiency of

$$\eta_{\text{engine}} = \frac{T_p - T_f}{T_p}. \quad (31)$$

This work then drives a refrigerator to draw heat  $Q$  out of a cold reservoir at temperature  $T$  and to dump waste heat  $E_{\text{out}}^{\text{fridge}}$  into a hot reservoir at temperature  $T_f$ , with a Carnot coefficient of performance given by

$$\eta_{\text{fridge}} = \frac{T}{T_f - T}. \quad (32)$$

Note that as the pump power  $\dot{E}_{\text{in}}$  is reduced,  $T_f$  decreases—with a lower limit of  $T_A \geq T$  according to Eq. (26)—and hence  $\eta_{\text{fridge}}$  increases, but at the expense of an overall decreased cooling power  $\eta_C \dot{E}_{\text{in}}$ . The product of Eqs. (31) and (32) equals Eq. (30), and the net input and output energy fluxes of Fig. 5 reproduce those of Fig. 1 if one identifies  $E_{\text{out}} = E_{\text{out}}^{\text{engine}} + E_{\text{out}}^{\text{fridge}}$ . This scheme prompts the realization that the engine is required only because the pump radiation is not “pure work” for a real laser. If the pump laser were ideal, then  $T_p \rightarrow \infty$  so that  $\Delta T = 0$  and Eq. (30) would directly reduce to Eq. (32). In contrast, if one were to try to pump an optical cooler with fluorescence radiation (say by recycling the light), then  $T_p \rightarrow T_f$  and Eq. (30) indicates that its coef-

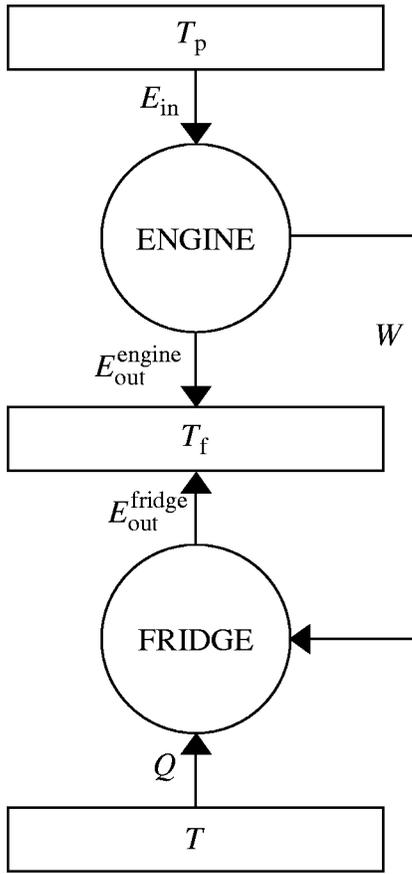


Fig. 5. A heat engine operating between thermal reservoirs with a high temperature of  $T_p$  and a cold temperature of  $T_f$  which is moderately larger than ambient, whose output work  $W$  is used to drive a refrigerator, which cools a load at temperature  $T$  somewhat less than ambient and rejects heat into the reservoir at intermediate temperature  $T_f$ .

efficient of performance would fall to zero. Thermodynamics thus explains why optical cooling requires a laser pump for efficient operation: The loss in entropy of the cooling load must, at minimum, be compensated by the gain in entropy of the radiation as it is converted from the input pump to the output fluorescence. Therefore, it is not surprising that Landau<sup>24</sup> dismissed the practicality of fluorescent cooling in 1946.

Consider an example using actual values<sup>22</sup> relevant to laser cooling of  $\text{Yb}^{3+}:\text{ZBLAN}$ . The laser pump is narrowband and very bright, so that Eq. (23) is applicable,

$$T_p = \frac{hc}{k\lambda_p} \frac{\bar{n}_p}{\ln \bar{n}_p}, \quad (33)$$

where

$$\bar{n}_p = \frac{\lambda_p^3}{2hc} \frac{\dot{E}_p}{\pi R_p^2 \Delta \nu_p \pi \delta_p^2}, \quad (34)$$

from Eq. (3). Suppose that the pump laser has a power of  $\dot{E}_p \approx 40$  W, a beam radius of  $R_p = 0.5$  mm, a bandwidth of  $\Delta \nu_p = 40$  GHz, a divergence of  $\delta_p = 1$  mrad, and a wavelength of  $\lambda_p = 1030$  nm. Then  $\bar{n}_p \approx 10^9$ , which justifies the use of Eq. (23), and  $T_p \approx 7 \times 10^{11}$  K. In contrast, the fluorescence is only moderately bright, so that Eq. (20) is called for,

$$T_f = \frac{hc}{k\lambda_{0f}} \frac{1}{1 + \ln(1/\bar{n}_f)}, \quad (35)$$

with

$$\bar{n}_f = \frac{\lambda_{0f}^3}{2hc} \frac{\dot{E}_f}{A_{\text{sample}} \Delta \nu_f \pi}. \quad (36)$$

The fluorescence spectrum has a peak wavelength of  $\lambda_{0f} = 975$  nm and a full-width at half-maximum of about  $\Delta \lambda_f = 35$  nm  $\approx c^{-1} \lambda_{0f}^2 \Delta \nu_f$  at room temperature. The fluorescence is assumed to be emitted homogeneously and hemispherically from the surface of the cooling sample (of area  $A_{\text{sample}}$ ), which is taken to be a cylinder whose height and diameter are each 3.0 cm, with a total power of  $\dot{E}_f = 40$  W. [Note that  $\dot{E}_p = \dot{E}_f / (1 + \eta)$  according to Eqs. (27) and (28), thus explaining the approximation in the specification of the pump power.] Hence  $\bar{n}_f = 6.4 \times 10^{-4}$  and  $T_f = 1760$  K. (An exact calculation using a measured fluorescence spectrum<sup>17</sup> gives a similar value of 1530 K, thereby justifying the approximations used here.) Clearly  $T_p$  is so much larger than  $T_f$  that Eq. (32) can be taken to be the Carnot coefficient of performance of this optical refrigerator. Consequently  $\eta_c = 20\%$  at room temperature, and it diminishes approximately linearly to zero as  $T \rightarrow 0$ .

In contrast, the actual cooling coefficient of performance is

$$\eta_{\text{actual}} = (\bar{\nu}_f - \nu_p) / \nu_p = (\lambda_p - \bar{\lambda}_f) / \bar{\lambda}_f, \quad (37)$$

according to the discussion in Sec. II, assuming that the pump beam is entirely absorbed by the sample. In the case<sup>8</sup> of  $\text{Yb}^{3+}:\text{ZBLAN}$ ,  $\bar{\lambda}_f = 995$  nm so that  $\eta_{\text{actual}} = 3.5\%$  at the pump wavelength of 1030 nm used in Eqs. (33) and (34). Although it might appear that one could increase the cooling performance by tuning the pump laser to longer wavelengths, in practice the measured value of  $\eta$  is found to roll off because (heat producing) trace impurity absorption begins to dominate the rapidly decreasing ytterbium absorption. Furthermore, the cooling coefficient decreases faster than linearly as the operating temperature is decreased,<sup>22</sup> because the long-wavelength absorption coefficient decreases with decreasing temperature (so that  $\lambda_p$  has to be reduced), while simultaneously the average fluorescence wavelength  $\bar{\lambda}_f$  increases (due to diminishing phonon absorption within the ground and excited bands of energy levels).

An alternative to pumping the refrigerator in Fig. 1 optically is to pump a semiconductor diode electrically.<sup>25</sup> The key to this electroluminescent cooling is to choose an applied forward bias  $V$  which is sufficiently smaller than the average bandgap emission energy  $h\bar{\nu}$  divided by the electron charge  $e$ . As in the case of laser cooling, one factor that determines how much smaller the pump energy  $eV$  must be than  $h\bar{\nu}$  to achieve net cooling is the fluorescence quantum efficiency. In particular, Auger processes and surface recombination limit this efficiency. If we ignore such limitations, the Carnot coefficient of performance is again given by Eq. (32), because the electrical pump in this case is a source of “pure” work. In practice, however, the required electrical current creates Joule heating of the diode, which is the primary reason that optical pumping is preferred.<sup>13</sup> Unfortunately, the large refractive index of semiconductors (e.g., 3.6 for GaAs) leads to trapping of the fluorescence photons, which de-

creases the external fluorescence quantum efficiency, as the photons rattle around inside the sample and are thus more likely to encounter a nonradiative decay channel. Berdahl<sup>26</sup> has suggested that this total internal reflection could be frustrated by bringing an external absorber to within a few microns of the semiconductor surface. Another suggestion by Berdahl is to reverse bias the diode so that it cools a neighboring sample by absorbing its thermal emission, a phenomenon known as negative luminescence.

The preceding thermodynamic analysis of photoluminescent cooling can be similarly applied to the optically pumped laser in Fig. 3. We define its efficiency as

$$\eta \equiv \frac{E_{\text{out}}}{E_{\text{in}}}. \quad (38)$$

An analysis similar to the derivation of Eq. (30) can be used to deduce a Carnot limit of

$$\eta_C = \frac{T_p - T}{T_p - \Delta T}, \quad (39)$$

where  $\Delta T \equiv TT_p/T_l$ . In this case,  $T$  is the steady-state operating temperature of the laser medium, and  $T_l$  and  $T_p$  are the flux temperatures of the output laser and input pump radiation, respectively. As one might have guessed,  $\eta_C$  increases as the ratio  $T_p/T_l$  of these two flux temperatures increases.<sup>27</sup> In fact, the thermodynamic limit on the efficiency of the laser becomes 100% as this ratio approaches unity, which is a big advantage of diode-pumped laser systems. (In practice, however,  $T_p$  will always be less than  $T_l$  because a set of mutually incoherent diode laser beams is essentially being combined into a single, high quality output beam.)

The idea of pumping one laser with another suggests an intriguing concept. One can connect the refrigerator of Fig. 1 to the laser of Fig. 3, and simultaneously pump both in such a manner that all of the waste heat generated by the latter is removed by the former. Remarkably, this scheme can be implemented using the same active material for both devices,<sup>28</sup> by choosing the pump frequency  $\nu_p$  to be intermediate between the mean spontaneous emission frequency  $\bar{\nu}_f$  and the stimulated emission frequency  $\nu_l$ . In effect, one is balancing the anti-Stokes cooling from the refrigerator against the Stokes heating from the laser, resulting in what is known as athermal or radiation-balanced lasing. A simultaneous optical and thermodynamic analysis of a model athermal laser based on an ytterbium-doped crystalline system can be found in Ref. 29.

## V. CONCLUDING REMARKS

Laser cooling is now a well established laboratory technique with myriad practical applications. For atom cooling, the concept frequently is explained from the point of view of momentum, in which large numbers of photons are fired like high-speed ping-pong balls against an oncoming bowling-ball-like atom to translationally slow it down.<sup>30</sup> Another explanation, appropriate both to the idea of the anti-Stokes cooling of solids and Doppler cooling of atoms, is to use energy conservation to balance the heat loss from the target against the energy gain of the radiation. As good as these explanations are, neither is able to explain why a laser pump is required rather than say a collimated beam of narrowband light from a high-intensity lamp. The simplest explanation of this last issue invokes entropy as discussed in this paper.

In particular, a statistical equation for radiation entropy can be deduced from the Shannon information-theoretical definition. The advantage of this approach over the traditional radiative heat transfer viewpoint is that it encompasses nonequilibrium photon distributions. Once the entropy is defined, two effective temperatures of radiation can be introduced to parametrize it. A Carnot limit on the performance of an optical engine or refrigerator is then easily derived from the first and second laws of thermodynamics. As usual, this limit would only be attainable for reversible operation of the device. The calculations in this article indicate, for example, that an ytterbium based solid-state laser cooler has a thermodynamic limit on its coefficient of performance of 20% for a typical set of operating conditions. However, experimental measurements to date have indicated that at best 3.5% of the pump power has actually been converted into cooling using this material. This discrepancy is indicative of the inefficiencies resulting from impurities in the samples and irreversibilities in the fluorescent emission. On the positive side, this shortfall implies that the ultimate limits on optical cooling of bulk matter have not been approached and that plenty of room remains for new scientific and engineering developments in this emerging field of research.<sup>17</sup>

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## APPENDIX: THE ENTROPY OF RADIATION

A recent derivation of Eq. (1) can be found in Ref. 21, but it is beyond the understanding of undergraduate students. A more accessible treatment is in Ref. 18 and is outlined here. Filling in the details in the derivation makes for a reasonable student exercise. We begin from the Shannon definition of the entropy,<sup>31</sup>

$$S = -k \sum_{\text{states}} P_{\text{state}} \ln P_{\text{state}}, \quad (A1)$$

where the summation is over all possible states of the system, each of which has probability  $P_{\text{state}}$ . In the present context, a particular state is defined by the occupation of  $N_1$  photons in optical mode 1,  $N_2$  photons in optical mode 2, and so on. Now assume that the probability  $p_i(N_i)$  of finding  $N_i$  photons in mode  $i$  is independent of the probability  $p_j(N_j)$  of finding  $N_j$  photons in mode  $j$  whenever  $i \neq j$ . Then

$$P_{\text{state}} = \prod_{i=1}^{\infty} p_i(N_i), \quad (A2)$$

where

$$\sum_{N_i=0}^{\infty} p_i(N_i) = 1 \quad \text{for any } i. \quad (A3)$$

Straightforward algebra leads to

$$S = -k \sum_{i=1}^{\infty} \sum_{N_i=0}^{\infty} p_i(N_i) \ln p_i(N_i). \quad (A4)$$

Next assume that the probability of finding one additional photon in any mode is independent of the number already occupying that mode. This assumption implies that

$$p_i(N_i) \propto q_i^{N_i}, \quad (\text{A5})$$

where  $0 \leq q_i < 1$ . If we normalize  $p_i(N_i)$  in Eq. (A5) according to Eq. (A3), we obtain

$$p_i(N_i) = (1 - q_i) q_i^{N_i}. \quad (\text{A6})$$

But the definition of the mean occupation number is

$$n_i \equiv \sum_{N_i=0}^{\infty} N_i p_i(N_i). \quad (\text{A7})$$

We substitute Eq. (A6) into Eq. (A7) and evaluate the sum to obtain

$$n_i = \frac{q_i}{1 - q_i} \Rightarrow q_i = \frac{n_i}{1 + n_i}. \quad (\text{A8})$$

Equation (A8) is consistent with the fact that  $q_i$  can be interpreted from Eq. (A6) as either the relative probability of there being one more photon in mode  $i$ , or the probability of finding a nonzero number of photons in that mode. (Consider for example the cases where  $n_i = 0, 1$ , or  $\infty$ .) We next substitute Eq. (A8) into Eq. (A6) to obtain

$$p_i(N_i) = \frac{n_i^{N_i}}{(1 + n_i)^{N_i + 1}}, \quad (\text{A9})$$

and then substitute Eq. (A9) into (A4) to finally find

$$S = k \sum_{i=1}^{\infty} [(1 + n_i) \ln(1 + n_i) - n_i \ln n_i]. \quad (\text{A10})$$

The density of photon states<sup>20</sup> per polarization mode in a frequency interval  $d\nu$  and element of solid angle  $d\Omega$  is  $c^{-3} \nu^2 d\nu d\Omega$ . We multiply this result by two because of the two independent transverse directions of the polarization of light, and by the speed  $c$  to obtain the entropy per unit time rather than per unit distance. We also multiply by the element of surface area  $dA$ , as well as by  $\cos \theta$  to project this area element perpendicularly to the direction of photon propagation. We thereby convert the sum in Eq. (A10) into the integral given by Eq. (1).

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