Advances in Laser Cooling of Solids

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We review the progress on laser cooling of solids. Laser cooling of ion-doped solids and semiconductors is based on the anti-Stokes fluorescence, where the emitted photons have a mean energy higher than that of the absorbed photons. The thermodynamic analysis shows that this cooling process does not violate the second law, and that the achieved efficiency is much lower than the theoretical limit. Laser cooling has experienced rapid progress in rare-earth-ion doped solids in the last decade, with the temperature difference increasing from 0.3 *to* 92 K. *Further improvements can be explored from the perspectives of materials and structures. Also, theories need to be developed, to provide guidance for searching enhanced cooling performance. Theoretical predictions show that semiconductors may be cooled more than ion-doped solids, but no success in bulk cooling has been achieved yet after a few attempts (due to the fluorescence trapping and nonradiative recombination). Possible solutions are discussed, and net cooling is expected to be realized in the near future.* [DOI: 10.1115/1.2360596]

1 Introduction

Cooling is a process in which thermal energy is absorbed from a lower temperature reservoir and deposited to a higher temperature reservoir, by consuming a small amount of higher grade energy. Mechanical, electrical, and optical energies are among the high grade energies, and are expected to be used for cooling purposes. Gas compression refrigerators, which consume mechanical energy, and thermoelectric coolers, which consume electrical energy, are matured techniques which have found very broad applications. High grade optical energies like lasers are, however, well-known for their heating effects rather than cooling capabilities. In fact, gases have been cooled to the order of nano-Kelvin by lasers, and this technique resulted in the 1997 and 2001 Nobel Prizes in Physics [1–4]. Its counterpart, laser cooling of solids, has also attracted great interests recently, for the potential to develop an optical cryocooler for a variety of important applications such as the cooling of sensors.

The concept of laser cooling (optical refrigeration) of solids dates back to 1929, when Pringsheim recognized that thermal vibrational energy (phonon) can be removed by the anti-Stokes fluorescence, i.e., the photons emitted by an optical material have a mean energy higher than that of the absorbed photons [5]. Initially, it was believed that optical cooling by the anti-Stokes fluorescence contradicted the second law of thermodynamics. Predictions suggested that the cycle of excitation and fluorescence was reversible, and hence the optical cooling would be equivalent to the complete transformation of heat to work [6,7]. This issue was cleared by Landau by assigning entropy to radiation [8]. It was shown that the entropy of a radiation field increases with its frequency bandwidth and also the solid angle through which it propagates. Since the incident laser light has a very small bandwidth and propagates in a well-defined direction, it has almost zero entropy. On the other hand, the fluorescence is relatively broadband and is emitted in all directions and, therefore, it has a comparatively larger entropy. In this way, the second law of thermodynamics is satisfied.

Many attempts have been made to realize radiative refrigeration experimentally, and the associated theoretical interpretations have been discussed. The earliest experiment was performed by Kushida and Geusic on Nd: YAG [9]. Reduced heating other than net cooling was observed, which was conjectured to be a result of the impurities in the crystal. Later Djeu and Whitney laser cooled low-pressure CO_2 by 1 K from 600 K by using a CO_2 laser for pumping [10]. In 1995, Epstein et al. [11] reported the first successful experiment of laser cooling in solids. Since then, various Yb or Tm doped glasses and crystals have been cooled [12–21]. Particularly, bulk solids have been cooled from room temperature to 208 K (creating a temperature difference $\Delta T = 92$ K) [21]. Continuous progress has been made [19,22,23] towards achieving cryogenic temperatures. For semiconductors, theoretical predictions have shown their potential to be cooled to as low as 50 K starting from room temperature [24], but experimental success is yet to be achieved due to some serious challenges to be overcome.

These progresses, as well as the recent success in laser cooling of gases and the subsequent achievement of Bose-Einstein condensation [1–4], again stimulated interest in optical cooling of solids.

2 Principles of Laser Cooling of Solids

Laser cooling can be viewed as the inverse cycle of lasers, and laser materials are in principle also good candidates for laser cooling. Common laser materials in the solid state include ion-doped solids and semiconductors, which are currently being studied for laser cooling.

In Fig. 1(*a*), the fundamental energy carriers involved in the laser cooling of rare-earth-ion doped solids are shown. There is a host crystal lattice, idealized as transparent to the pumping laser. Some of its atoms are replaced by optically active, doped ions (e.g., Yb^{3+}). The ion is represented by an effective transition dipole moment, which is the matrix element of the dipole operator $e_e\boldsymbol{r}$ [25], i.e.,

$$\boldsymbol{\mu}_e = \int \psi_f^* e_e \boldsymbol{r} \psi_i d^3 r \qquad (1)$$

where, e_e is the electron charge, $\boldsymbol{r}$ is the position vector, and ψ_i and ψ_f are the initial and final state wave functions of the two level system. One can think about this matrix element as coupling states ψ_i and ψ_f, which have different parity, creating or absorbing a photon. The electromagnetic field, which has a polarization vector $\boldsymbol{e}_\alpha$, may interact with the ion if the coupling factor $\boldsymbol{e}_\alpha \cdot \boldsymbol{\mu}_e$ is nonzero (i.e., they are not orthogonal).

Shown in Fig. 1(*b*) are the principles of the photon-electron-phonon interactions which result in the cooling effect in the solid. As the pumping wavelength is tuned to the red side of the reso-

Contributed by the Heat Transfer Division of ASME for publication in the JOURNAL OF HEAT TRANSFER. Manuscript received January 26, 2006; final manuscript received June 18, 2006. Review conducted by Zhuomin M. Zhang.

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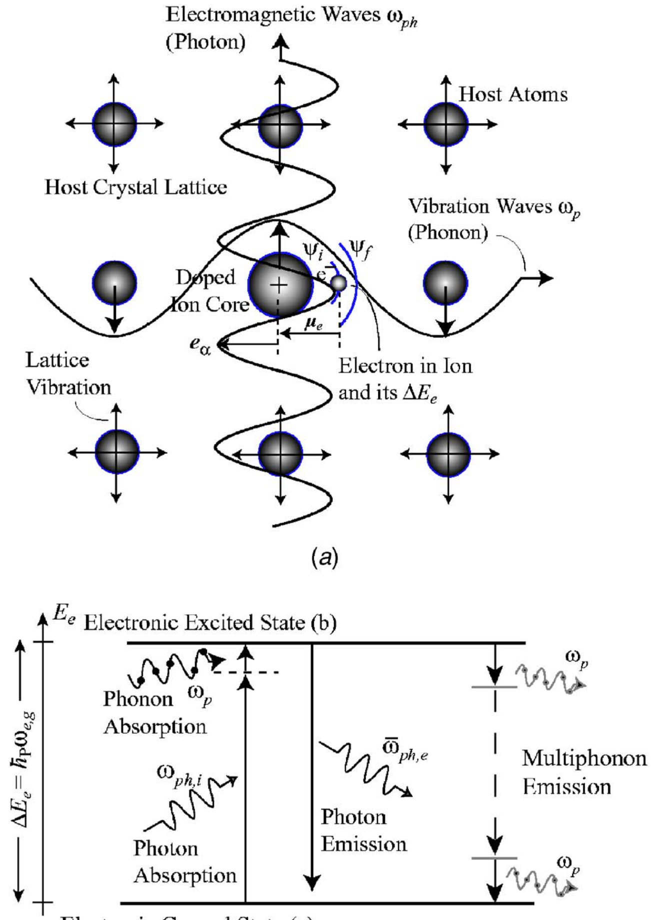

Fig. 1 (*a*) Three fundamental energy carriers in rare-earth ion doped solids irradiated by laser light: Photons from the pumping fields, phonons from the host crystal, and electrons of the doped ions. (*b*) Principles of laser cooling in rare-earth ion doped crystal. The electron is excited by absorbing a photon and one or more lattice phonons, and then decays by emitting a higher energy photon.

nance, the probability of a purely electronic transition between electronic sublevels, a first-order process, becomes smaller. On the other hand, the phonon-assisted transition, a second-order process, starts to contribute significantly to absorption. As a result, the absorption turns out to be a combination of the first- and second-order transitions. Since a much longer pumping wavelength than the resonance is used in laser cooling, the total transition is believed to be dominated by the second-order process. In such a process, the medium is irradiated by laser light with a frequency $\omega_{ph,i}$ that is below the resonance frequency $\omega_{e,g}$ for the energy gap (10,250 cm^{-1} for Yb^{3+} ion in Y_2O_3), the electron may still be excited by absorbing a photon from the pumping field and a phonon with a frequency ω_p from the host, such that $\omega_{ph,i}+\omega_p = \omega_{e,g}$. The electron then undergoes a radiative decay by emitting a photon with a frequency $\omega_{ph,e}$, or undergoes a nonradiative decay by emitting a few phonons (multiphonon relaxation), leading to internal heating of the system. The energy magnitudes of these carriers involved in the laser cooling process in $Yb^{3+}:Y_2O_3$ are shown in Fig. 2 [26].

The quantum efficiency $\eta_{e\text{-}ph}$ is defined as the ratio of the radiative decay rate to the total decay rate, or, in this case, the ratio of the number of emitted photons to that of absorbed photons [26]. The net cooling power per absorbed photon P_c is given by

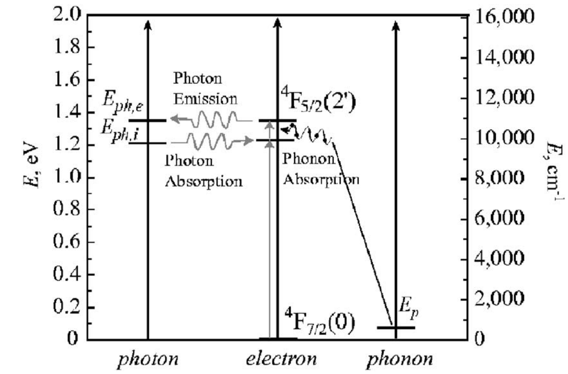

Fig. 2 Energy spectra of all three carriers in irradiated $Yb^{3+}:Y_2O_3$. Yb^{3+} has only two main electronic levels: $^4F_{7/2}$ and $^4F_{5/2}$. Carriers may interact with each other as energy and momentum conservations are met [26].

$$P_c = \eta_{e\text{-}ph}\hbar\bar{\omega}_{ph,e} - \hbar\omega_{ph,i} = \hbar\omega_{ph,i}\left(\eta_{e\text{-}ph}\frac{\bar{\omega}_{ph,e}}{\omega_{ph,i}} - 1\right) \tag{2}$$

where $\bar{\omega}_{ph,e}$ is the mean frequency of emitted photons. The cooling efficiency is defined as the ratio of the net cooling power and the absorbed power, i.e.,

$$\eta_c = \frac{P_c}{\hbar\omega_{ph,i}} = \eta_{e\text{-}ph}\frac{\bar{\omega}_{ph,e}}{\omega_{ph,i}} - 1 \tag{3}$$

Cooling is achieved as η_c is positive. Evidently, high quantum efficiency $\eta_{e\text{-}ph}$ is desirable.

The cooling process in semiconductors, as shown in Fig. 3, is similar to that of ion-doped solids. An electron originally in the valence band absorbs a laser photon and is excited to the conduction band, leaving a hole in the valence band. It then gains some energy by absorbing a phonon (intraband absorption) and climbs to a higher position in the conduction band. The electron then decays back to the valence band via either radiative recombination, or nonradiative recombination, which includes the multiphonon process and Auger process. The net cooling power is in the same form as Eq. (2).

3 Macroscopic Role of Laser Cooling

In Fig. 4 a macroscopic energy diagram is shown for a solid that is cooled by laser, where various energy flows are shown. To minimize the external thermal load to achieve the most cooling effect, in most of the existing experiments the solid was supported by very thin wires and was placed in a vacuum, to eliminate the

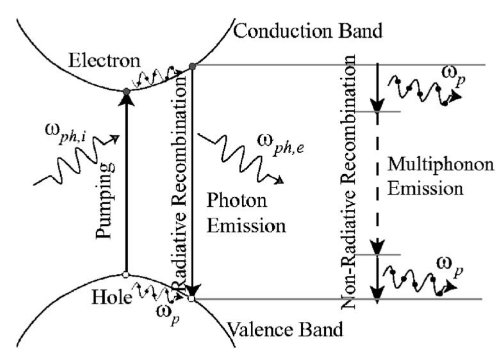

Fig. 3 Process for laser cooling of a semiconductor in which a laser photon with frequency $\omega_{ph,i}$ is absorbed followed by emission of an up-converted fluorescence photon with frequency $\omega_{ph,e}$

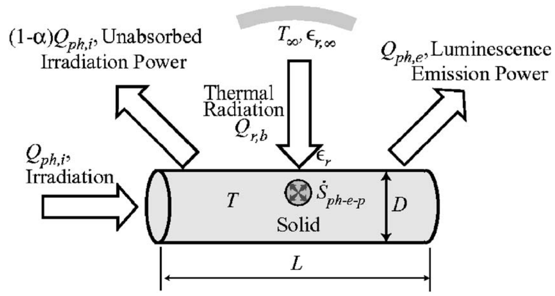

Fig. 4 The energy diagram for laser cooling of a solid, where radiation is the only external thermal load

conduction and convection. The only external load is then the thermal radiation from its surroundings, as shown in Fig. 4.

Using Eq. (3), the total cooling power $\dot{S}_{ph\text{-}e\text{-}p}$ is given by

$$\dot{S}_{ph\text{-}e\text{-}p} = P_c \frac{Q_{ph,a}}{\hbar\omega_{ph,i}} = \eta_c Q_{ph,a} = \eta_c \alpha Q_{ph,i} \tag{4}$$

where $Q_{ph,a}$ is the absorbed power, $Q_{ph,i}$ is the irradiation power, and α is the absorptivity. This equation links the macroscopic cooling behavior and the atomic level parameters. The result also indicates that the net cooling power $\dot{S}_{ph\text{-}e\text{-}p}$ is proportional to the absorbed power $Q_{ph,a}$.

The steady-state, integral-volume energy equation is

$$\dot{S}_{ph\text{-}e\text{-}p} - Q_{r,b} = 0 \tag{5}$$

Assuming that the surface area of the surrounding is much larger than that of the sample, the thermal radiation load $Q_{r,b}$ is given by [27]

$$Q_{r,b} = A_r \epsilon_r \sigma_{SB}(T_\infty^4 - T^4) \tag{6}$$

where A_r is the surface area of the solid, σ_{SB} is the Stefan-Boltzmann constant, and T_∞ is the temperature of the ambient radiation field (or the effective temperature in case the ambient field does not have a thermal spectra). Apparently the amount of cooling power $\dot{S}_{ph\text{-}e\text{-}p}$ governs how much the sample temperature can be lowered from the surrounding temperature.

4 Applications of Laser Cooling

Since the process of anti-Stokes fluorescence does not require any mechanical movement, such a solid-state cooler is likely to have a longer lifetime than other coolers. This is particularly useful in space, where reliability and lifetime are crucial considerations.

Despite the low cooling efficiency (2% to 3% at room temperature so far, and lower efficiency at lower temperatures), laser cooling is a promising candidate for cryocoolers, with the potential to cool ion-doped dielectrics and semiconductors to 50 K or even 10 K starting from room temperature [28]. Predictions have shown that at these cryogenic temperatures, thermoelectric coolers become ineffective or incapable, compared to laser coolers [29].

Another application is in the heat-balanced laser system. This process would employ fluorescence cooling to offset the heat produced in the generation of laser radiation [30–35].

5 Thermodynamics of Laser Cooling of Solids

As mentioned in the Introduction, there was a debate which stimulated the birth of radiation thermodynamics, that is, whether laser cooling is thermodynamically possible. Detailed discussions of the thermodynamics of optical cooling have been given by Mungan and Gosnell [36,37]. The following is based on their papers, with the inclusion of some more recent ideas from other researchers.

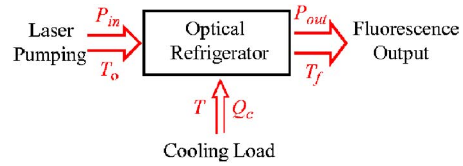

Fig. 5 A control volume showing various energies in and out in the laser cooling of a solid [37]

A simple control volume is shown in Fig. 5, in which the power flowing in and out (the pump laser, the external thermal load, and the luminescence emission) are marked. For a narrowband radiation which is independent of the angular directions θ and ϕ over a circular cone of half-angle δ, the energy flux density is given by [37]

$$I_{\dot{E}}(\omega) = \frac{1}{4\pi^2}\hbar c^{-2}\bar{f}_{ph}\omega_0^3 \Delta\omega \sin^2\delta \tag{7}$$

where c is the speed of light, $\bar{f}_{ph}$ is the average photon distribution function over the frequency range, ω_0 is the central frequency, and $\Delta\omega$ is the bandwidth of the beam. The entropy flux density is

$$I_S = \frac{1}{4\pi^2}k_B c^{-2}[(1+\bar{f}_{ph})\ln(1+\bar{f}_{ph}) - \bar{f}_{ph}\ln\bar{f}_{ph}]\omega_0^2\Delta\omega\sin^2\delta \tag{8}$$

If either the bandwidth $\Delta\omega$ or the divergence δ of the source collapses to zero, Eq. (7) implies that $\bar{f}_{ph}\to\infty$, to ensure that the energy flux density remains finite. In this limit, one can show that Eq. (8) yields $I_S\to 0$, i.e., 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.

To analyze the limiting efficiency of laser cooling of solids, it is useful to define the flux temperature T_F of the radiation, which is given by

$$T_F \equiv \frac{I_{\dot{E}}}{I_S} = \frac{\hbar\omega_0}{k_B}\frac{\bar{f}_{ph}}{(1+\bar{f}_{ph})\ln(1+\bar{f}_{ph}) - \bar{f}_{ph}\ln\bar{f}_{ph}} \tag{9}$$

Again, for an ideal laser, we have $T_F\to\infty$, which is consistent with the zero entropy at a finite irradiance. It can be further deduced that the flux temperature of narrowband radiation propagating in a well-defined direction is higher than that of broadband radiation propagating in all directions.

In laser cooling of solids, according to the first law of thermodynamics, we have $P_{out}=P_{in}+Q_c$. The cooling coefficient of performance is defined in the usual way for a refrigerator as $\eta = Q_c/P_{in}$. The maximum value of η is the Carnot limit, η_C, and is determined by the second law of thermodynamics. The entropy carried 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, i.e.,

$$\frac{P_{out}}{T_f} \geqslant \frac{P_{in}}{T_o} + \frac{Q_c}{T} \tag{10}$$

where T is the steady-state operating temperature of the refrigerator, and T_f and T_o are the flux temperatures of the fluorescence and pump radiation fields, respectively. The reversible Carnot limit is obtained by choosing the equality sign in Eq. (10). Finally we have

$$\eta_C = \frac{T-\Delta T}{T_f - T} \tag{11}$$

where $\Delta T = TT_f/T_o$. In the limit of $T_o \gg T_f$, the efficiency (11) would reduce to the Carnot form.

Consider an example using actual values relevant to laser cooling of Yb^{3+}:ZBLANP [37]. The temperatures of the pump laser and the fluorescence are calculated, using Eq. (9), to be $T_o=7$

$\times 10^{11}$ K and $T_f = 1760$ K, respectively. Thus the Carnot efficiency of this optical cooler is about 20% at room temperature, and it diminishes approximately linearly to zero as $T \to 0$. However, the actual cooling efficiency achieved to date is only around 3%, which indicates that much irreversibility has been produced in the process. One might use a longer pumping wavelength to obtain a higher cooling efficiency, but the absorption coefficient of Yb^{3+} would become too small. As a result, the trace impurity absorption will dominate over the Yb^{3+} absorption, and the cooling efficiency η decreases. To reduce these irreversibilities introduced into this process, the sample should be purified to suppress the trace absorption.

If the system is pumped hard, so that the threshold is reached, the emission will become stimulated rather than spontaneous. Although this would accelerate the cycling, the thermodynamic efficiency limit will become too small, due to the low entropy of the stimulated emission fields [38]. Anyway, one cannot expect to get cooling and laser output simultaneously by using a laser input.

6 Advance in Laser Cooling of Rare-Earth Ion-Doped Solids

Ion-doped solids were the first class of materials on which laser actions were demonstrated [39], and were attempted early for laser cooling.

6.1 Experimental Investigations. The earliest experiment was performed on Nd^{3+}:YAG by Geusic et al. at Bell Laboratories in 1968 [9], just a few years after he demonstrated the first laser action in this transition-metal doped crystal [40]. This focused on flash-lamp-pumped crystals of Nd^{3+}:YAG, with the fluorescence from one crystal being used for the cooling of another. When compared with an undoped reference sample, the neodymium-doped sample showed reduced heating, but net cooling was not observed. This was conjectured to be due to impurities in the crystals, which offset the cooling effects. A simple model yielded the rate of temperature change, and the results agreed with the experiment. Later rare-earth-ion doped solids were demonstrated for laser emission, and these materials immediately became attractive for laser cooling purpose, since the optical $4f$ levels are shielded from the surrounding by the filled $5s$ and $5p$ shells, leading to the suppression of the multiphonon relaxation. In 1995, Epstein et al. reported the first experimental success of laser cooling in solids [11]. The absorption and fluorescence spectra were measured, as shown in Fig. 6(a), where the mean fluorescence wavelength is marked. As the pumping wavelength was tuned longer than the mean fluorescence wave-length, a maximum local temperature decrease of 0.3 K was detected, as shown in Fig. 6(b). However, the cooling effect diminishes when the pumping wavelength is tuned further away, since the off-resonance absorptivity becomes too small. The cooling efficiencies achieved were up to 2%. Later, ytterbium-doped glasses have been cooled by nearly 70 K below room temperature and have reportedly cooled at temperatures as low as 77 K [16]. Edwards et al. [14] demonstrated a prototypical cryogenic refrigerator based on Yb^{3+}:ZBLANP pumped with a 1.6 W Ti:sapphire laser and measured a temperature decrease of 48 K from room temperature. Cooling from a low starting temperature in various Yb^{3+}-doped glasses has also been observed, suggesting that a cryogenic refrigerator with an extended dynamic range can be built. Mungan et al. [12] observed local cooling in a Yb^{3+}:ZBLANP sample at temperatures between 100 and 300 K, maintaining a cooling efficiency of about 0.01 through this range. Local cooling between 77 K and room temperature by photothermal deflection and spectroscopic techniques in a fluorochloride glass (Yb^{3+}:CNBZn) and a fluoride glass (Yb^{3+}:BIG) were reported by Fernandez et al. [16]. The cooling efficiency was shown to change with temperature, varying between 0.02 and 0.006 in the two materials. Gosnell cooled a Yb^{3+}-doped fiber by the amount $\Delta T = 65$ K from room temperature [15]. His experimental apparatus was very carefully

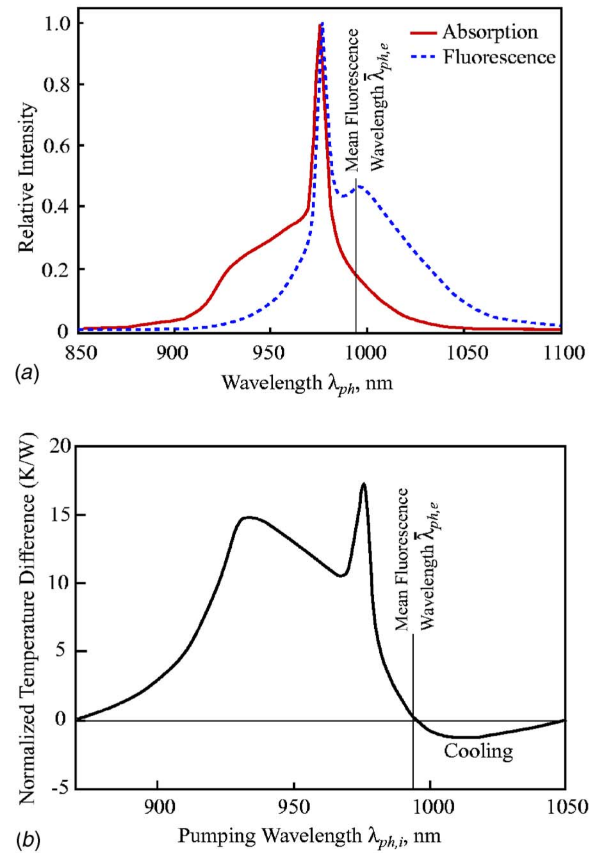

Fig. 6 (a) The absorption and fluorescence spectra of Yb^{3+}:ZBLANP, with the mean fluorescence wavelength marked [11]. (b) The normalized temperature difference with respect to the pumping wavelength. Cooling is detected as the pumping wavelength is tuned longer than the mean fluorescence wavelength [11].

designed, as shown in Fig. 7. This record was again pushed to 92 K below room temperature by Epstein et al. in 2005 [21]. Continuous progress has been made [19,22,23] towards achieving cryogenic temperatures.

In these experiments, accurate temperature measurement is of great importance. Temperature can be measured by contact or noncontact methods. A noncontact method, fluorescence thermometry, has been used in many of the existing experiments [11,15]. It is based on the fact that the fluorescence spectrum of the glass is independent of the pump laser wavelength but only dependent on temperature. A spectrum-temperature relation can be calibrated over a wide range of temperatures using a thermostat. Then the observed spectrum is compared to the reference spectra and the temperature is determined. This method is capable of measuring temperature without disturbing the original system, and is very suitable for systems as fine as laser cooling. Thermocouple is another obvious choice for its simplicity. However, it introduces an external thermal load to the cooling element and may reduce or even eliminate the cooling effect. This method is thus usually used for a rough examination of whether or not the system can be cooled, but not for an accurate temperature measurement [14].

To enhance laser cooling performance, one perspective is finding new materials, including dopants and hosts. Thulium-doped glasses are good candidates, since thulium has a transition resonance at 1.8 μm, whereas the ytterbium transition is near 1 μm. Then the thulium system is capable of obtaining the same amount of cooling power with a much smaller pumping photon energy. Hoyt et al. showed cooling of a sample of Tm^{3+}-doped ZBLANP by 1.2 K from room temperature, under vacuum, when approxi-

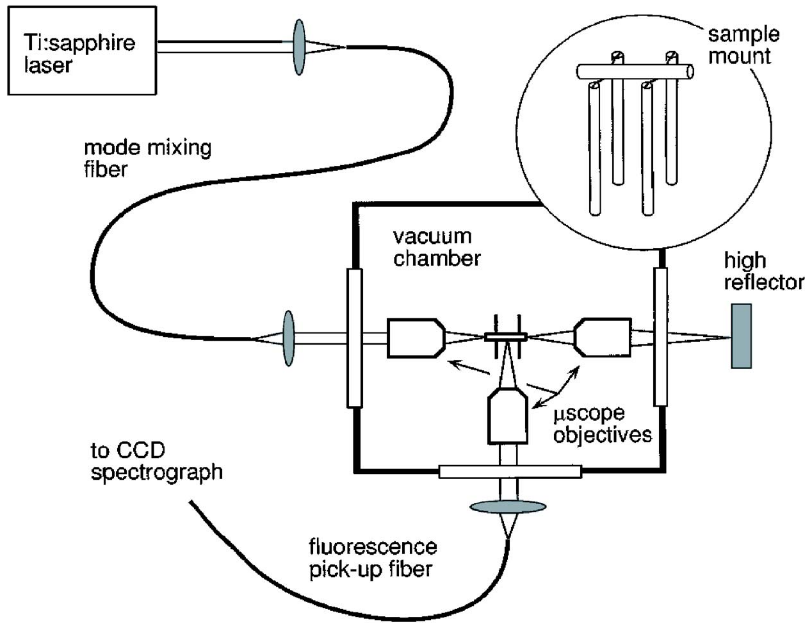

Fig. 7 Experimental apparatus used for observing laser cooling of Yb:ZBLANP [15]. Pump radiation from a cw Ti:sapphire laser undergoes mode scrambling within an external multimode silica fiber before injection into the sample fiber, which is positioned upon a sample mount (inset), imposing an extremely low conductive thermal load. Unabsorbed pump radiation is collected from the output end of the sample fiber and reinjected into the sample with the help of an external high reflector. Finally, emitted fluorescence is collected with a third internal optic and is spectrally resolved to determine the temperature.

mately 3 W of laser power at 1.9 μm was incident on the sample from a periodically poles lithium-niobate-based optical parametric oscillator in turn pumped by a 20 W cw Nd:YAG laser. Their results indicated a cooling efficiency of 3.4%, which compared favorably with the efficiencies achieved for ytterbium-doped glasses. To date, Yb^{3+} and Tm^{3+} are the only two rare-earth elements on which laser cooling has been demonstrated, although some other elements are considered to be good candidates.

A number of ytterbium-doped crystal hosts were also studied by Bowman and Mungan [22], to determine which of them can be cooled. Besides a number that did not exhibit net cooling, they found that crystals of ytterbium-doped $KGd(WO_4)_2$ can indeed be cooled. This was the first demonstration of anti-Stokes laser cooling of a crystal. Fernandez et al. achieved internal cooling of other ytterbium doped glasses (CNBZn and BIG) [16,41]. Recently, another set of results showing cooling of crystals was reported by Epstein et al. [17]. These results show cooling of 2.3% ytterbium-doped YAG crystals and 3% ytterbium-doped Y_2SiO_5. A comparison is made with ytterbium-doped ZBLAN, which shows that the cooling efficiency is slightly larger in ZBLAN, but the thermal and mechanical properties of YAG may be advantageous for some applications. Several other groups have tried to find other materials suitable for cooling, but even materials that have shown promise from an analysis of their absorption and emission spectra have had either too low a quantum efficiency or excessive absorption due to impurities for cooling to be achieved. Using the experimental data for lifetimes, Hoyt studied the possibility of laser cooling of a few Tm-doped solids, and identified those which have a potential for cooling due to their high quantum efficiencies [42].

Another perspective to enhance the cooling performance is modifying the structure. Gosnell used a long, thin optical fiber as the host, in order to increase the optical pathlength for a larger absorptivity, and to reduce the external thermal load [15]. To further enhance the absorptivity, some researchers [21,42] placed the sample between two dielectric mirrors of high reflectance at the pumping wavelength only, as shown in Fig. 8(*a*). In this way the laser pumping is reflected back and forth, while the fluorescence can escape. Recognizing that the mirrors may bring in extra loss, Heeg et al. proposed an alternative approach [20,43]. By locating the cooling medium inside a laser cavity, it was efficiently pumped by the inherent multipassing and high circulating power of the laser resonator. Recently, Ruan et al. attempted this problem from a new perspective: Nanostructure [44]. The medium in their model is a Yb^{3+} doped Y_2O_3 nanopowder, as shown in Fig. 8(*b*). They predicted that the absorption can be significantly enhanced due to the optimized dopant concentration, the size effect of the phonon density of states, and the multiple scattering of the pumping photons.

6.2 Theoretical Analysis. The basic principles of laser cooling and its thermodynamic validity provided motivation for the above mentioned experiments. Except for these, more detailed theoretical analysis achieved very limited progress, compared to the rapid improvements of laser cooling experiments. This is mainly due to very complicated physical mechanisms under the laser cooling process. Lamouche considered the temperature dependence of cooling efficiency [45]. By analyzing the temperature dependence of fluorescence and absorption spectra of Yb^{3+}:ZBLAN, they concluded that cooling would decrease with decreasing temperature, as shown in Fig. 9. Fernandez et al. used the Fermi's Golden Rule to interpret their experimental results, by assuming that the absorption is dominated by the phonon-assisted process [16]. The absorption rate $\dot{\gamma}_{e,a}$ is given by

$$\dot{\gamma}_{e,a} = \sum_f \dot{\gamma}_{e,i-f} = \frac{2\pi}{\hbar_P} \sum_f |M_{fi}|^2 \delta(\hbar_P \omega_{ph,i} + \hbar_P \omega_p - \hbar_P \omega_{e,g}) \tag{12}$$

where $\hbar_P \omega_{e,g}$ is the energy difference of the two electronic levels, and M_{fi} is the interaction matrix element. The δ function guaran-

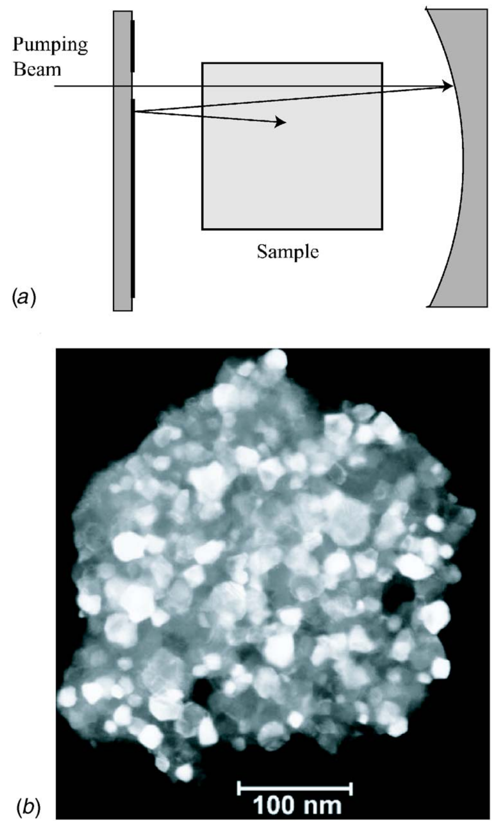

Fig. 8 Modified structures for enhanced laser cooling performance: (*a*) Cavity arrangement for multiple passes [42], (*b*) A micrograph of $Yb^{3+}:Y_2O_3$ nanopowder [51].

tees the energy conservation, that the sum of the pumping photon energy and the phonon energy should be equal to the electronic gap energy. In this theory, the decreasing cooling efficiency with decreasing temperature is due to the fact that the phonon distribution function decreases. Heeg et al. investigated the effect of another important factor, the fluorescence reabsorption, on the cooling performance [46,47]. The random process of fluorescence reabsorption and trapping were analyzed in solid-state optical materials. Using the absorption and fluorescence spectra of Yb^{3+}:ZBLAN as input data, they employed a random-walk model to test analytical approximations of the fluorescence escape efficiency and cooling efficiency, including reflections at the boundary. They concluded that moderate concentration and sample size should be used to avoid the dominance of fluorescence reabsorption.

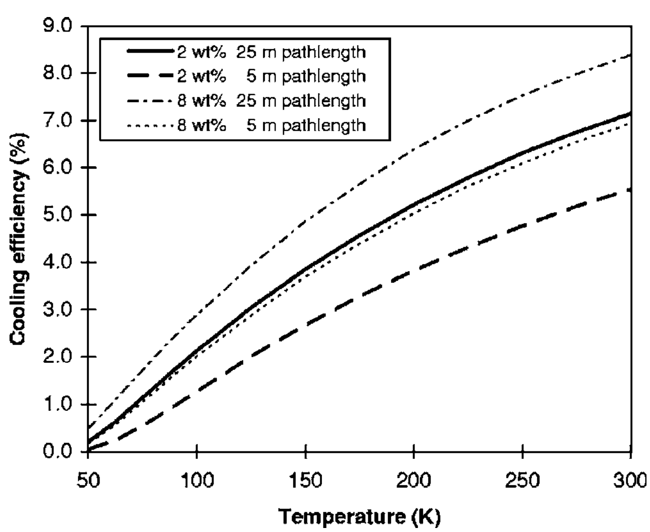

Fig. 9 Predicted cooling efficiency as a function of temperature, for the 2 and 8 wt % $Yb^{3+}:Y_2O_3$ in the linear regime for two optical pathlengths [45].

Despite these interesting theoretical studies, a more important issue, if not the most important, is to develop a theory that can provide the criterion of the material selection. Although there are many factors limiting the laser cooling performance, the materials properties, including dopants and hosts, are essential. An ideal dopant-host pair should allow for an effective ion-phonon coupling, which otherwise should not be large enough to result in multiphonon relaxation. Ruan et al. recently used the Fermi's Golden Rule to decouple various limiting factors, aiming to develop an atomic level of understanding [44]. They found that the cooling performance is limited by the population of the three carriers and their couplings. The nanostructure was proposed to be capable of enhancing the carrier populations, and the understanding of the ion-phonon coupling mechanism is to be established and is crucial for engineering the materials for laser cooling.

7 Advance of Laser Cooling in Semiconductors

Given the recent advance in fabrication of semiconductor devices and their use as lasers, it is not surprising that a number of researchers have considered these candidates for optical cooling. A GaAs/GaInP heterostructure was studied for possible cooling by Gauck et al. [48]. They observed blue-shifted luminescence but did not see net cooling because coupling inefficiency caused luminescence reabsorption. Finkeiben et al. [49] detected local cooling in the area of the pumping beam spot that was due to anti-stokes photoluminescence in a GaAs quantum-well structure, recording a temperature drop of 7 K from liquid-nitrogen temperature. Sheik-Bahae [24] et al. performed a theoretical analysis considering nonradiative decay and luminescence reabsorption, and proposed the feasibility of laser cooling in semiconductors. Nevertheless, no bulk cooling of semiconductors has yet been realized experimentally, where the luminescence trapping due to total internal reflection remains a major obstacle.

One possible solution is to use an index-matched dome lens attached to the cooling element [48], as shown in Fig. 10(*a*). Since the dome lens is much larger than the heterostructure, the fluorescence emitted at any angle would become nearly normal to the dome surface, making the extraction much more efficiently. The drawback is that the dome lens introduces more external thermal load and increases the system size. Another possible solution is to use a nanogap structure [50], as shown in Fig. 10(*b*). For the onset of the total internal reflection, there exists an evanescent wave on the other side of the surface which has an exponentially decaying amplitude and does not transfer any energy. If another surface is brought closely to the first one at a distance shorter than the wavelength, the evanescent waves will be coupled to the second surface and become propagating waves. In this way, the originally trapped fluorescence can be efficiently coupled out of the cooling element.

8 Conclusions and Outlook

Over the past decade, substantial progress has been made in the laser cooling of solids. Cooling of rare-earth-ion doped solids has been demonstrated and improved, and cooling in semiconductors are anticipated in the near future. Current work is focused on finding new materials and structures that can enhance the cooling performance, or that can be cooled (for semiconductors). Ways are also being explored in which a practical optical cooler might be engineered.

Further research into laser cooling of solids should firstly explore new materials and structures for enhanced cooling performance. Material properties determine the maximum cooling effi-

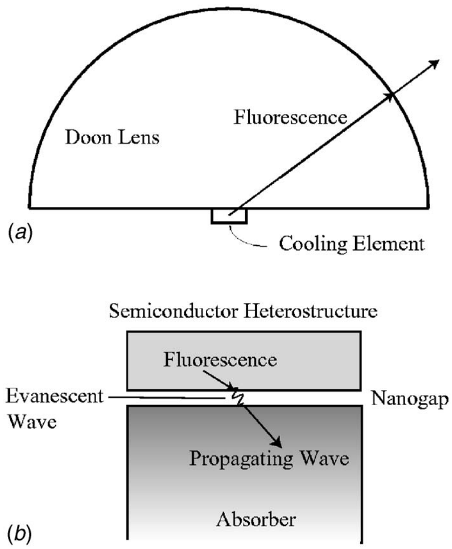

Fig. 10 Possible structures for extracting the fluorescence from the high refractive-index semiconductors: (*a*) An index-matching dome lens attached on the cooling element [48]. (*b*) A nanogap structure to couple the evanescent waves out of the cooling element [50].

ciency that can be achieved, so further advances could result from investigation of other dopants, such as thulium and others, to replace ytterbium. Consideration should also be given to other host materials, including crystals and glasses. New structures such as nanostructure are promising due to the quantum size effect, and should be considered as a major direction. Given the rate of advancement of semiconductor manufacturing techniques, they may be ideal candidates for laser cooling in the near future, since the electronic structures could be engineered to optimize the cooling efficiency for specific applications. Equally important to the experimental investigations is the development of a theory that can provide a criterion for the material selection. Although this is very challenging, considering the complexity of the electron-phonon coupling, useful results are expected to be obtained in the near future, which can then provide guidance for searching new materials and structures.

Acknowledgment

We are thankful to Professor Stephen Rand for many discussions/suggestions.

Nomenclature

A = surface area (m^2)
c_o = speed of light in vacuum (m/s)
E = complex electric field (V/m)
E = energy (J)
e_e = electron charge (C)
f_p, f_{ph} = phonon, photon distribution function
I = intensity (W/m^2)
M = interaction matrix
P = power (W)
Q = power (W)
r = interatomic distance (m)
$\dot{S}$ = energy conversion (W)
T = temperature (K)

Greek Symbols

α = absorptivity
δ = divergence
$\dot{\gamma}$ = transition rate (1/s)
$\dot{\gamma}_{e,a}$ = transition probability per unit time per unit energy density [$1/s/(J/m^3)$])
η_c = cooling efficiency
$\eta_{e\text{-}ph}$ = quantum efficiency
μ = magnetic permeability (H/m)
$\boldsymbol{\mu}_e$ = dipole moment vector (C-m)
ψ = wave function
ω = angular frequency (rad/s)

Subscripts

a = absorption
c = cooling
e = emission, electron
F = flux
f = fluid, final state, fluorescence
g = gap
i = incident, initial state
o = free space
p = phonon
ph = photon
r = radiative, real part of a complex number
S = entropy

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