Mechanism of refrigeration cycle on laser cooling of solids

Youhua Jia (贾佑华)[1,2*], Biao Zhong (钟 标)[2], and Jianping Yin (印建平)[2]

[1] *Science College, Shanghai Second Polytechnic University, Shanghai 201209, China*

[2] *State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200062, China*

[*] *Corresponding author: yhjia@sf.sspu.cn*

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A simple model is developed to study the laser cooling of solids. The condition of laser cooling of a solid is developed. By using some parameters of the Yb^{3+} ion, which is most widely used in laser cooling, we then calculate the cooling power and the cooling efficiency. In order to make a more precise analysis, the effect of fluorescent reabsorption, which is unavoidable in the cooling process, is discussed using the random walk model. Taking Tm^{3+} ion as an example, we derive the average number of absorption events and determine the change in quantum efficiency due to reabsorption. Finally, we obtain the red-shift of the fluorescent wavelength and the requirement of sample dimension.

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As early as 1929, Pringsheim proposed an idea to cool a solid material by using anti-Stokes fluorescence. In this process, the material absorbs a pumping photon with a longer wavelength and soon afterwards emits a fluorescence photon with a shorter wavelength; the energy difference between the two photons, which results from the internal energy of the material, is removed by the fluorescence radiation, and the material is cooled. Many researchers have initially opposed this idea as they cannot relate laser to refrigeration. However, in 1995, the first demonstration of net laser cooling of a solid was reported[1], and a Yb^{3+}-doped fluorozirconate glass ZBLANP was cooled to 0.3 K below room temperature. Thereafter, people have carried out a series of theoretical and experimental studies for laser cooling of solids and obtained great progress[2−12].

Since anti-Stokes fluorescence cooling has been successfully achieved, explanations about the experimental result have been continually explored. Lamouche *et al.* found a theoretical model[12] and derived the cooling efficiency in arbitrary temperature by evaluating the experimental spectroscopy. The relationship between the mean fluorescent wavelength and the temperature was also determined. As the model of Lamouche is very complex, we propose a simple two-level system to analyze the micro course of laser cooling and then calculate the cooling power and cooling efficiency. We then discuss several main parameters that influence cooling power and determine the relationship between temperature and time.

In order to obtain efficient laser cooling of solids, the key is to choose the proper fluorescent center and its level structure, as well as the appropriate energy gap. Taking Tm^{3+} ions as example, their energy manifolds are shown in Fig. 1. Each level in the several inhomogeneous broadened levels corresponds to a Stark-split manifold. For example, the $^3F_4$ level has 9 Stark-split levels, while the $^3H_6$ level has 13. Clearly, it is suitable to choose the $^3H_6$ and $^3F_4$ manifolds as the levels of laser cooling, wherein $^3H_6$ is the upper level and $^3F_4$ is the lower level of the laser pumping. In the cooling cycle, $^3H_6$ is the ground state and $^3F_4$ is the excited state; the corresponding frequency of the pumping laser is ν, while the mean fluorescent frequency is ν_f, and they must satisfy the condition of anti-Stokes fluorescence cooling: $h\nu_f > h\nu$.

The cooling cycle in anti-Stokes fluorescence cooling, as shown in Fig. 1, can be described as follows. The Tm^{3+} ion absorbs one pump photon $h\nu$, and the doped ion is excited from position 1 of the ground state $^3H_6$ to position 2 of the excited state $^3F_4$; the excitation is then thermodynamically relaxed (i.e., the non-radiation transition) from position 2 of the state $^3F_4$ to its position 3 by absorbing vibrant energy from the host. Afterwards, the atomic ion is spontaneously decayed from position 3 of the state $^3F_4$ to position 4 of the state $^3H_6$ by emitting a fluorescent photon $h\nu_f$. Finally, it is thermodynamically relaxed (i.e., the non-radiation transition) to the position of the state $^3H_6$, thereby forming a closed and repeatable "laser pumping-fluorescent radiation" cooling cycle in which the average energy, $h\nu_f - h\nu$, is removed from the solid material. Because the two thermodynamics decay processes mentioned earlier are in the same direction, the Tm^{3+} level structure relative to laser cooling in Fig. 1 can be simplified as a two-level system with excited-state splitting, as shown in Fig. 2, where $|1\rangle$ is the ground state and $|2\rangle$ is the excited state. This simplified two-level system (see Fig. 2) is then used to analyze the process of laser pumping-fluorescent radiation in anti-Stokes fluorescence cooling.

In Fig. 2, supposing that the energy gap between the ground state $|1\rangle$ and the lower level of the excited state $2\rangle$ is ΔE, and the gap formed by Stark splitting in the excited state $|2\rangle$ is δ, the number of nonradiative deexcitations per second can be defined as

$$W_{\rm nr} = N_2 W_0 \exp(-\gamma \Delta E), \tag{1}$$

where W_0 is the phenomenological parameter that depends strongly on the host material, N_2 is the excited-state population, and γ is inversely proportional to the characteristic phonon energy for a given material and is also host dependent. For the ZBLAN, the values of W_0 and γ are 1.99×10^5 s^{-1} and 2.1×10^{-3} cm^{-1}, respectively[13]. Assuming that only radiative and

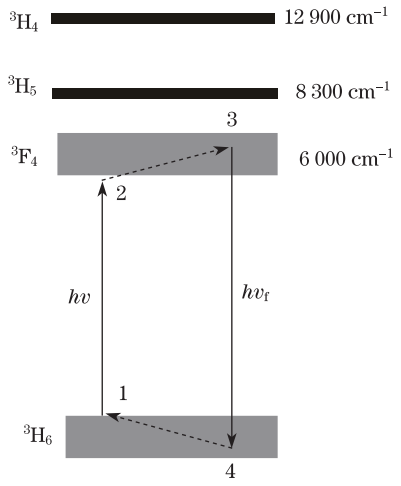

Fig. 1. Energy-level structure of Tm^{3+} ion.

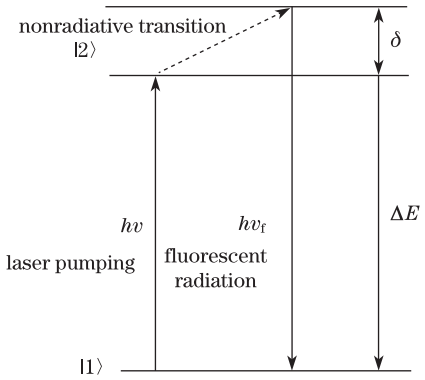

Fig. 2. Simplified energy-level structure of laser cooling.

nonradiative transitions exist in the cooling cycle; the impurity ions, the fluorescent reabsorption, and the process of energy transfer can be neglected; and the vibration energy of phonon is about equal to δ, we can obtain the number of phonons generated by the nonradiative transition per unit interval as $N_{\rm nr} = N_2 W_0 \exp(-\gamma\Delta E)\Delta E/\delta$. When the anti-Stokes fluorescent radiation occurs, each transition will carry away one phonon. The number of phonons absorbed by the radiative transition per unit interval (that is, the particle population excited to the excited state per unit interval) is given by $N_{\rm r} = [W_{\rm pump} - N_2 W_0 \exp(-\gamma\Delta E)]\exp(-\delta/kT)$, where k is the Boltzmann constant and $W_{\rm pump}$ is the pumping rate, which is a parameter relative to the laser pumping power and the absorption coefficient of the material; this can be defined as

$$W_{\rm pump} = \frac{P_{\rm pump}}{h\nu} = \frac{P_{\rm inc}(1-{\rm e}^{-\alpha L})}{h\nu} = \frac{N_2}{\tau_{\rm f}}, \qquad (2)$$

where α is the absorption coefficient, L is the length of the solid sample, and $\tau_{\rm f}$ is the fluorescence lifetime. When $N_{\rm r} \geqslant N_{\rm nr}$, there is cooling effect in the solid material. Supposing T= 300 K and δ = 230 cm^{-1}, our calculation shows that the condition $N_{\rm r} \geqslant N_{\rm nr}$ is converted into $\Delta E \geqslant 4\,800$ cm^{-1}. This condition is well satisfied by Yb^{3+} and Tm^{3+}, indicating that our simple model is feasible.

The power of anti-Stokes fluorescence cooling can be defined as[1]

$$P_{\rm cool} \equiv P_{\rm flu} - P_{\rm abs} = P_{\rm abs}\Big(\frac{\lambda}{\lambda_f} - 1\Big), \qquad (3)$$

where $P_{\rm abs}$ and $P_{\rm flu}$ are the pumping powers absorbed by the material and fluorescent-radiation power, respectively, and λ and $\lambda_{\rm f}$ are the pumping laser and fluorescent-radiation wavelengths, respectively. On the other hand, the cooling power can also be defined as

$$\begin{aligned} P_{\rm cool} &\equiv (N_{\rm r} - N_{\rm nr})\delta \\ &= (W_{\rm pump} - W_{\rm nr})\delta \exp\Big(-\frac{\delta}{kT}\Big) - W_{\rm nr}\Delta E \\ &= [W_{\rm pump} - N_2 W_0 \exp(-\gamma\Delta E)]\delta \exp\Big(-\frac{\delta}{kT}\Big) \\ &\quad - N_2 W_0 \exp(-\gamma\Delta E)\Delta E, \end{aligned} \qquad (4)$$

where the first item shows the power of Yb^{3+} ions, which transit to the excited state from the ground state, and the second item represents the power of phonons generated by multi-phonon relaxation. From Eq. (4), when the fluorescence lifetime of ytterbium is 1.9 ms, we can calculate the dependence of the energy gap on the cooling power under different pumping rates, and the results are shown in Fig. 3. It can be seen from Fig. 3 that the higher the pumping rate is, the greater the cooling power will be; with increasing the energy gap, the cooling power increases continuously and finally reaches the maximum constant value of $W_{\rm pump}\delta\exp(-\delta/kT)$. This is because the quantum efficiency of the transition between the ground and excited states will be increased to one when the energy gap is increased. In addition, if the pumping rate is increased by five times, the maximum cooling power will also be increased by five times.

In general, the relationship between the laser pumping energy and the quantity of heat released from the material can be characterized by the heat-light converting efficiency, which can also be called the cooling efficiency and defined as[4]

$$\Omega \equiv \frac{P_{\rm cool}}{P_{\rm abs}} = \frac{P_{\rm flu} - P_{\rm abs}}{P_{\rm abs}}. \qquad (5)$$

Substituting Eq. (4) into Eq. (5), we can obtain the average heat-light converting efficiency by

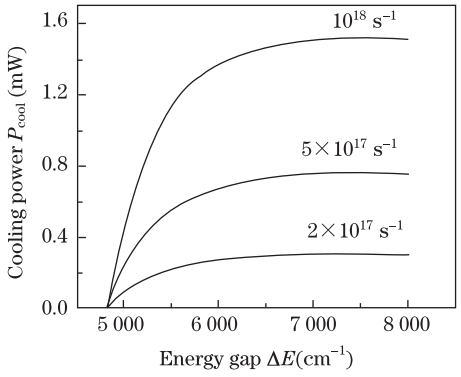

Fig. 3. Relationship between the cooling power and the energy gap for different pumping rates.

$$\begin{aligned}\Omega &= \frac{P_{\rm cool}}{W_{\rm pump}\Delta E} \\ &= \frac{(W_{\rm pump}-W_{\rm nr})\delta\exp(-\frac{\delta}{kT})}{W_{\rm pump}\Delta E} - \frac{W_{\rm nr}}{W_{\rm pump}} \\ &= [\delta\exp\Big(-\frac{\delta}{kT}\Big) - \tau_{\rm f}\delta\exp\Big(-\frac{\delta}{kT}\Big)W_0\exp(-\gamma\Delta E) \\ &\quad - \tau_{\rm f}W_0\exp(-\gamma\Delta E)\Delta E]/\Delta E, \end{aligned} \tag{6}$$

where $W_{\rm pump}\Delta E$ represents the pump power absorbed by the cooling material per unit interval. From Eq. (6), we can calculate the dependence of the heat-light converting efficiency on the energy gap, and the results are shown in Fig. 4. As seen in Fig. 4, the maximum heat-light converting efficiency and the corresponding optimal energy gap exist, indicating that under this optimal energy gap, the utilization efficiency of the pumping power will reach the maximal value. Therefore, even if the reasonable selection of the energy gap contributed to the increase in cooling efficiency, it still depends on new findings of efficiently cooling materials. From Eq. (6), it can be seen that the pumping rate has no influence on the cooling efficiency, which depends strongly on the cooling material.

Thus far, we have discussed the cooling power and cooling efficiency, neglecting the influence of fluorescent reabsorption. However, fluorescent reabsorption exists in many condensed-phase phenomena, which include photoactive centers with broad absorption and fluorescence spectra. Due to the overlap between absorption and fluorescence, the fluorescent reabsorption is unavoidable in laser cooling of solids.

The effect of fluorescent reabsorption on optical cooling in solids is calculated using the stochastic model[14]. This is a semi-analytical approach to this problem. This model has recently been used in studies on reabsorption of fluorescent emission[13]. Using the absorption and fluorescent spectra of Tm^{3+}:ZBLANP as input data, we analyzed the random process of fluorescent reabsorption in solid-state optical materials.

Figure 5 shows the absorptivity and fluorescence spectra of Tm^{3+}:ZBLANP[3]. The reabsorption coefficient is defined as the integrated result of the absorption spectrum and the normalized fluorescence intensity. We define

$$\alpha_{\rm R} = \frac{\int \alpha F_0 {\rm d}\lambda}{\int F_0 {\rm d}\lambda}, \tag{7}$$

where α is the absorption efficient and F_0 is the normalized intensity of the natural fluorescence. From the spectrum of Tm^{3+}:ZBLANP, using Eq. (7), the reabsorption coefficient is about 0.08.

The random walk process can be described as follows. A random number ξ is generated, and the fluorescence quanta travelling a distance l corresponding to a probability for the reabsorption of ξ is considered. According to the Beer's law, $l = -\ln\xi/\alpha_{\rm R}$. Two more random numbers, $0 < \gamma < 1$ and $0 < \phi < 1$, are generated to determine the new position of the photon when it is reabsorbed. The

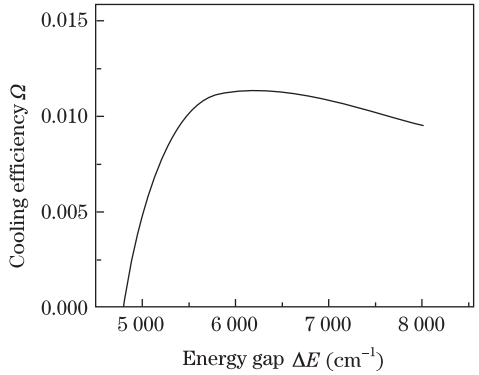

Fig. 4. Relationship between the cooling efficiency and the energy gap.

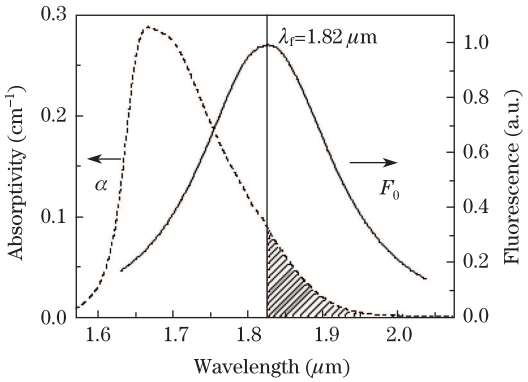

Fig. 5. Absorptivity and fluorescence spectra of Tm^{3+}: ZBLANP. The dotted curve is the absorptivity data and the solid curve is the fluorescence data.

new position can be given by

$$r(x_{\rm new}, y_{\rm new}, z_{\rm new}) = r(x_{\rm old}, y_{\rm old}, z_{\rm old}) + \Delta x + \Delta y + \Delta z, \tag{8}$$

where $\Delta x = l\cos(2\pi\phi)\cos(2\pi\gamma)$, $\Delta y = l\cos(2\pi\phi)\sin(2\pi\gamma)$, $\Delta z = l\sin(2\pi\phi)$. The decision of accepting or rejecting a random step will simply depend on whether the new position falls within the boundaries of the sample cell. If it does, reabsorption will likely occur. We can then take another step until the new position is outside the boundaries of the sample. Finally, the average number of absorption events N is determined, which can be used to analyze other phenomena in laser cooling of solids.

For a certain material, there is internal quantum efficiency η_0, which is the exciton-to-photon conversion efficiency. The quantum efficiency discussed here is only η_0. For a material to be laser cooled, it should have high quantum efficiency. When fluorescent reabsorption is considered, the quantum efficiency will decrease and can be described as

$$\eta = (\eta_0)^N, \tag{9}$$

where N is the average number of absorption events. Moreover, the change in quantum efficiency will result in the red-shift of fluorescent wavelength. The wavelength shift can be calculated by

$$\Delta\lambda = \frac{\lambda_{\rm F}}{\eta} - \frac{\lambda_{\rm F}}{\eta_0}, \tag{10}$$

where λ_F is the mean fluorescent wavelength. Taking Tm^{3+}:ZBLANP for example, the results are shown in Fig. 6. The fluorescent reabsorption can increase the fluorescent wavelength; when it comes to the edge of the absorption spectra, cooling will not easily occur.

Since the sample dimension has a significant impact on the average number of reabsorption, it will also greatly influence the cooling effect. Figure 7 shows the quantum efficiency as a function of the sample width. Large sample scale can lead to the decrease in quantum efficiency, although this is not clear for Tm^{3+}:ZBLANP. Similar calculations were carried out on Yb^{3+}:ZBLANP, and the influence is quite remarkable; cooling large materials will be very hard.

In conclusion, we present a simple theoretical model to analyze the condition of laser cooling of solids, especially focusing on the energy gap of the doped ions. We choose a proper energy gap, which is at least 5 000 cm^{-1}. Furthermore, there is an appropriate energy gap in Ho^{3+} or Er^{3+}[15] or Dy^{3+} ions, which may be used to realize the laser cooling of new solid materials[16,17]. In addition, the cooling power is calculated. Meanwhile, we analyze the fluorescent reabsorption problem in the laser cooling of solids. The random walk process shows the change in fluorescent wavelength and the cooling efficiency, and sample dimension was also discussed. Moreover, considering the efficient absorption and fluorescent reabsorption problems simultaneously, sample materials must be well fabricated. These experiment results are valuable as they can pave the way for the development of practical optical refrigerators[18].

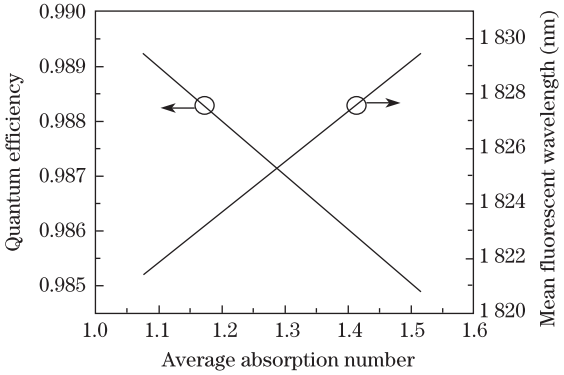

Fig. 6. Quantum efficiency and the mean fluorescent wavelength as a function of the average absorption number.

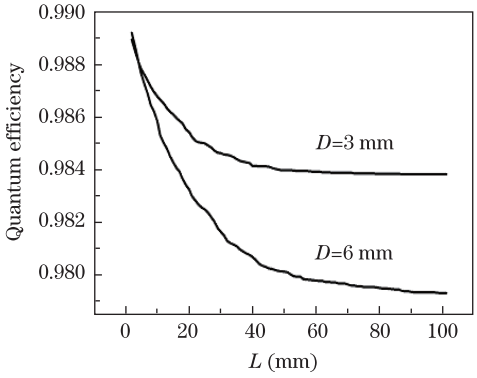

Fig. 7. Relationship between the quantum efficiency and the sample size.

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