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Improving laser cooling efficiencies of Yb-doped yttrium aluminum garnet by utilizing non-resonant anti-Stokes emission at high temperatures

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Abstract: Laser cooling in solids has been demonstrated, and cooling from room temperature to cryogenic temperatures has so far been the main focus of research. However, operation at high temperatures can be advantageous. We propose that laser cooling of Yb-doped yttrium aluminum garnet ceramic discs is enhanced above 300 K. Photoluminescence (PL) spectra is evidence that the phonon-assisted energy transfer from resonant states, which exhibit a narrow PL peak, to inhomogeneously distributed energy states becomes stronger at higher temperatures. This results in an enhanced non-resonant anti-Stokes PL at high temperatures. The ideal cooling efficiency determined from the PL spectrum at 470 K is 1.7 times higher than that at 300 K.

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1. Introduction

Cooling via efficient emission of anti-Stokes photoluminescence (PL) [1,2] (or so-called laser cooling) can be utilized to realize novel cooling devices with high reliability, compactness, and absence of mechanical vibrations [3]. While there exist several materials where laser cooling has been actually observed, it has been shown that rare earth (RE)-doped materials are good candidates for solid-state laser cooling because of their narrow emission spectra and discrete energy levels [4,5]. For example, resonant excitation of a Nd-doped yttrium aluminum garnet (YAG) crystal with intense 1064-nm laser light can result in reduction of heat [6]. The temperature does not necessarily decrease, because the background absorption and the non-radiative decay were not suppressed enough. Net cooling, that is, the actual decrease of the crystal temperature, has been observed for the first time in Yb-doped heavy-metal-fluoride glass (ZBLANP) [7]. The solid material with the best net cooling performance that has so far been reported, is Yb-doped yttrium lithium fluoride (YLF:Yb), which has enabled demonstration of optical refrigeration down to cryogenic temperatures below 100 K [8]. Recently, a HgCdTe sensor was cooled from room temperature to 135 K using solid-state optical refrigeration based on YLF:Yb [9]. However, other materials may be also suited for realizing optical refrigeration; (Y,Yb)₃Al₅O₁₂ (YAG:Yb) [10–12], Yb-doped fluorozirconate glass [13–15], and YLF:Yb [8,16,17] have been extensively studied. In particular, YAG:Yb, which was used to demonstrate net cooling by 8.9 K in air at atmospheric pressure [11], has a higher thermal conductivity, chemical stability, and mechanical stiffness compared to YLF:Yb.

Regarding the performance of such devices, a low background absorption coefficient α_B and a long non-radiative lifetime of the employed material are essential for efficient net cooling. The former parameter has to be low to avoid parasitic losses [17,18], and the latter has to be low to improve the anti-Stokes PL yield. Because the radiative lifetime of the intra-orbital $f-f$ transition is on the order of milliseconds, non-radiative recombination is relatively difficult to suppress when using RE-doped materials [15]. It has been shown that low non-radiative decay rates can be

realized in a trivalent RE ion with a large energy difference between ground state and first excited state (referred to as E_p) [19,20] and a host material with a low maximum phonon energy $\hbar\omega_{\max}$. Note that the trivalent Yb ion has a large energy difference of $E_p \approx 1.24$ eV [21]. Net cooling has been achieved in RE-doped materials that satisfy the empirical condition $\hbar\omega_{\max} < E_p/8$ [15].

Thus far, reports on laser cooling in Yb-doped low-phonon energy host materials have focused on the cooling of solids from room temperature to lower temperatures. On the other hand, cooling in order to maintain a stable temperature above room temperature is another promising application area of laser cooling. For instance, light-emitting diodes with high output powers generate a huge amount of heat, which needs to be quickly dissipated to prevent malfunction. According to the temperature dependence of multiphonon absorption in Er^{3+} in LaF_3 [22], the probability of multiphonon absorption obeys the Bose–Einstein distribution. Therefore, a strong anti-Stokes PL and high laser cooling efficiency can be expected at high temperatures. In this study, we reveal the temperature dependence of the anti-Stokes PL and relative cooling power of (Yb:Y)AG, where Yb is stable at the Y-site of YAG up to 470 K. The presentation of (Yb:Y)AG is used to identify which site is nominally substituted by Yb ion. The observed Stokes and anti-Stokes PL spectra are used to estimate the heat that can be carried away from the sample in an ideal case. Our results suggest that a high laser-cooling efficiency of (Yb:Y)AG can be realized above 300 K owing to the enhanced non-resonant anti-Stokes PL at high temperatures. The predicted ideal cooling efficiency at 470 K (2.2%) is significantly higher than that at 300 K (1.3%).

2. Sample fabrication and experimental setup

The (Yb:Y)AG samples used in this study were prepared by the solid-state reaction method explained in the following. First, high-purity (99.99%) raw powder materials of Y_2O_3 , Yb_2O_3 , and Al_2O_3 were mixed with a pestle for 1 hour. The total weight of the mixed powder was 1 g. After annealing the mixed powder in an electronic furnace at 1700 °C in air, the obtained crystal was crushed into a fine powder (particle diameter ≈ 40 μm). Several $(\text{Y}_{1-x}\text{Yb}_x)_3\text{Al}_5\text{O}_{12}$ crystal powders with Yb_2O_3 mole fractions x in the range from 0.02 to 0.13 were fabricated. For the fabrication of each (Yb:Y)AG ceramic discs, polyvinyl alcohol was added to the powder, and the powder was mechanically pressed at 200 MPa to obtain a disc with a diameter of 13 mm and a thickness of 2 mm. After annealing the disc in the electronic furnace at 1700 °C in air, the obtained (Yb:Y)AG ceramic disc was polished to a thickness of 100 μm and attached to a copper plate with a Ag/epoxy paste. The copper plate was mechanically fixed onto a sample holder in a cryostat system including a heater.

We performed four types of experiments: PL, PL excitation (PLE), anti-Stokes PL, and laser Raman spectroscopy measurements. To obtain monochromatic excitation light for the PL and PLE measurements, a supercontinuum laser with a repetition frequency of 20 MHz and a pulse duration of 6 ps was combined with a 220-mm double monochromator containing a grating with a line spacing of 300 gr/mm and a blaze wavelength of 1000 nm. The full width at half-maximum of the monochromatic excitation light was less than 1.6 nm. The anti-Stokes PL of each fabricated (Yb:Y)AG ceramic discs was measured with a narrow linewidth (< 1.26 nm) tunable diode laser. The incident angle of this p -polarized excitation light at 1030 nm [resonant excitation of the E3–E5 transition in (Yb:Y)AG as shown in Section 4] was fixed at the Brewster angle. The PL signal was dispersed by a 140-mm monochromator that uses a 600-gr/mm grating with a blaze wavelength of 1000 nm, and for detection we employed a liquid-nitrogen-cooled InGaAs diode array. The spectral resolution of this setup was better than 0.3 nm. A crossed polarizer pair was used to suppress the intensity of excitation light (scattered from the sample surface) that enters the monochromator. Laser Raman spectroscopy was conducted at various temperatures using a 500-mm aberration-corrected monochromator equipped with a 2400-gr/mm grating. The excitation wavelength in this experiment was 532 nm.

3. The relative cooling power

The cooling efficiency η_c is defined as the ratio of cooling power P_c to absorbed power P_{abs} [23]. This definition considers the effects of external luminescence quantum efficiency η_{ext} and the background absorption coefficient α_B , and a positive value of η_c represents cooling of the system while a negative value represents heating:

$$\eta_c = \frac{P_c}{P_{\text{abs}}} = \eta_{\text{ext}} \left(\frac{1}{1 + \frac{\alpha_B}{\alpha(\lambda)}} \right) \frac{\lambda_{\text{exc}}}{\lambda_f} - 1, \quad (1)$$

where $\alpha(\lambda)$ is the absorption coefficient of the cooling material, λ_{exc} is the excitation wavelength, and λ_f is the mean fluorescence wavelength. The latter can be calculated as shown in Eq. (2).

$$\lambda_f = \frac{\int \lambda F(\lambda) d\lambda}{\int F(\lambda) d\lambda}, \quad (2)$$

Here, $F(\lambda)$ is the fluorescence intensity, which is difficult to measure due to the actual contribution of the scattered excitation light in the PL spectra. We estimated $F(\lambda)$ by subtracting the scattered light contribution obtained by fitting a Gaussian function to the data. The ideal cooling efficiency, which considers the condition $\eta_{\text{ext}} = 1$ and $\alpha_B = 0$, reads [23]

$$\eta_c^{\text{ideal}} = \frac{\lambda_{\text{exc}}}{\lambda_f} - 1. \quad (3)$$

Note that the expressions for the cooling efficiency in Eq. (1) or also the ideal cooling efficiency in Eq. (3) cannot be used to directly evaluate the cooling power per single absorbed photon, because these terms represent the ratio P_c/P_{abs} only. Generally, when we compare the cooling efficiency, P_{abs} should be known. However, P_{abs} is not necessarily the same for each sample and is difficult to precisely determine the value. In this work, we newly defined the relative cooling power ξ in Eq. (4) as defined as a multiplying of the ideal cooling efficiency and the integrated luminescence intensity. Here, it is noted that the integrated luminescence intensity obtained under a given excitation intensity is proportional to the quantum efficiency. The cooling power P_c of the sample showing a strong luminescence becomes strong. Utilizing this relative cooling power, we can relatively compare the laser cooling property without using P_{abs} .

$$\xi = \left(\frac{\lambda_{\text{exc}}}{\lambda_f(\lambda_{\text{exc}})} - 1 \right) \int F(\lambda) d\lambda. \quad (4)$$

The mean fluorescence wavelength λ_f in Eq. (4) depends on λ_{exc} , because the assignment of photons to anti-Stokes or Stokes PL relies on the excitation wavelength λ_{exc} . A $\xi > 0$ indicates the possibility for cooling and the magnitude of ξ represents the power. In this work, ξ was used to identify the optimal sample and excitation conditions for laser cooling.

4. Optimization of laser cooling in (Yb:Y)AG

Before optimizing the excitation condition, we verified the optimum Yb_2O_3 mole fraction x in (Yb:Y)AG for laser cooling. Figure 1(a) shows a typical room-temperature PL spectrum of (Yb:Y)AG excited at 659 nm. We can confirm several strong peaks, which correspond to the intra-orbital $f-f$ transitions of (Yb:Y)AG. The fine-structure (splitting of the Stark levels) arises from the crystal field [24]. The inserted energy level diagram depicts the structure of the $4f$ electronic states in (Yb:Y)AG. The PL peaks appearing at 940, 968, 983, 1030, and 1048 nm are attributed to the $E6 \rightarrow E1$, $E5 \rightarrow E1$, $E7 \rightarrow E4$, $E5 \rightarrow E3$, and $E5 \rightarrow E4$ transitions, respectively. Here, we focus on the $E6 \rightarrow E1$, $E5 \rightarrow E1$, and $E5 \rightarrow E3$ transitions indicated with the diamond,

circle, and triangle, respectively, in Fig. 1(a). The x dependences of the PL peak intensities at 940 nm ($E6 \rightarrow E1$), 968 nm ($E5 \rightarrow E1$), and 1030 nm ($E5 \rightarrow E3$) are shown in Fig. 1(b). All three transitions exhibit their maximum PL intensity at $x = 0.06$. In the range from $x = 0.02$ to 0.06, the PL intensity increases with x owing to an increase in the emission cross section. However, a further increase in x leads to a reduction of the PL intensity because of concentration quenching. In the case of $\lambda_{\text{exc}} = 1030$ nm shown below, the re-absorption probability of the anti-Stokes PL by Yb ions is higher than that of the Stokes PL because there are strong absorption peaks at the wavelengths of 910, 940, and 968 nm [25]. Therefore, the external quantum efficiency of the anti-Stokes PL emitted from (Yb:Y)AG becomes lower than that of the Stokes PL in highly Yb-doped (Yb:Y)AG.

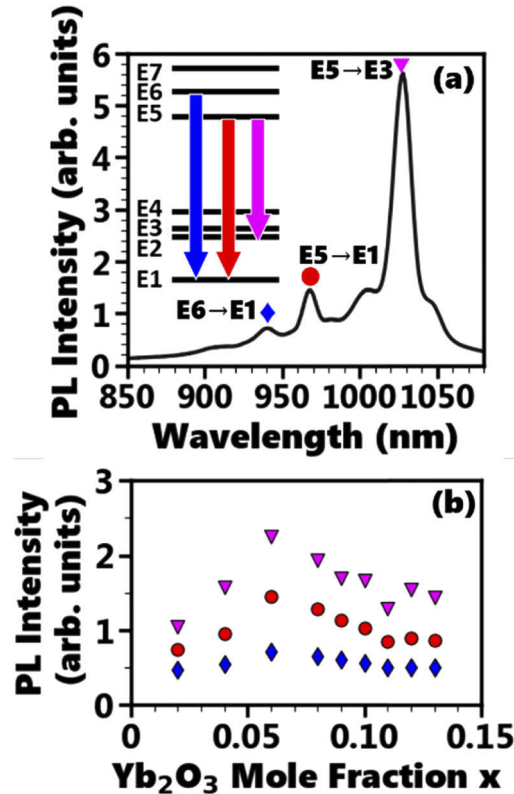


Fig. 1. (a) Room-temperature PL spectrum of (Yb:Y)AG with a Yb_2O_3 mole fraction $x = 0.06$. The employed excitation wavelength is $\lambda_{\text{exc}} = 659$ nm. (b) The dependences of the PL intensities on the Yb_2O_3 mole fraction obtained from the (Yb:Y)AG discs under the same conditions.

Figure 2 shows the excitation wavelength dependence of the relative cooling power ξ of the (Yb:Y)AG sample with $x = 0.06$ at the sample temperature $T = 300$ K. As mentioned in Section 3, a positive (negative) value of ξ indicates the possibility for cooling (heating) and $|\xi|$ represents the relative amount of heat carried away (generated) per absorbed photon. Obviously, when the sample is excited at 1030 nm, the relative cooling power reaches its maximum. This indicates that strong absorption is indispensable for obtaining large net cooling. Figure 3 shows a typical room-temperature PL spectrum of (Yb:Y)AG resonantly excited at 1030 nm (which induces the transition $E3 \rightarrow E5$). The anti-Stokes PL peak at 968 nm ($E5 \rightarrow E1$) is clearly observed. It is considered that multiphonon absorption corresponding to the energy difference of 77.1 meV (that

is, the energy difference between E3 and E1) is responsible for the relatively strong anti-Stokes PL peak at 968 nm.

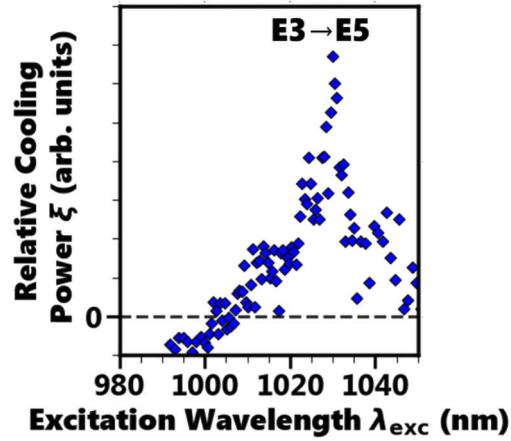


Fig. 2. Excitation wavelength dependence of the relative cooling power ξ of the (Yb:Y)AG sample with $x = 0.06$.

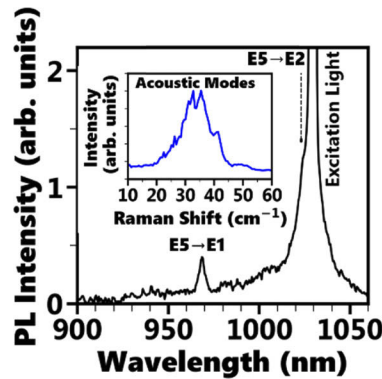


Fig. 3. Typical room-temperature PL spectrum of (Yb:Y)AG resonantly excited at $\lambda_{\text{exc}} = 1030$ nm (resonant excitation of the E3–E5 transition). The two anti-Stokes PL peaks at 968 and 1023 nm are assigned to the transitions E5→E1 and E5→E2, respectively. The inset shows the Raman signal of (Yb:Y)AG with $x = 0.06$ in the region of acoustic modes.

The inset of Fig. 3 plots the Raman scattering spectrum of (Yb:Y)AG with $x = 0.06$ in the region of acoustic phonon modes at 300 K. As the phonon density of states in undoped $\text{Yb}_3\text{Al}_5\text{O}_{12}$ [26] does not show any signal near 35 cm^{-1} (4.34 meV), we infer that the signal appearing in the acoustic phonon mode region arises from Yb doping. The energy of the phonon mode near 4.34 meV agrees well with the energy difference between the E2 and E3 energy levels [25], and thus phonon absorption is likely to cause the stronger anti-Stokes signal at 1025 nm in Fig. 3.

5. Temperature dependence of anti-Stokes PL

The PLE images in Fig. 4(a) and 4(b) provide the PL spectra of the (Yb:Y)AG sample with $x = 0.06$ measured as a function of the excitation wavelength λ_{exc} . Figures 4(a) and 4(b) are the results measured at $T = 300$ and 450 K, respectively. The color bar represents the logarithm

of the spectral intensity, and the intensities of both data sets can be directly compared. The saturated linear line in the image comes from the scattered excitation light, and the PL signals that appear at wavelengths λ shorter than the excitation wavelength λ_{exc} (on the left side of the saturated line) are the anti-Stokes signals. In Fig. 4(a), the integrated anti-Stokes signal for a single excitation wavelength surpasses the integrated Stokes signal for λ_{exc} longer than 1008 nm. In particular, the intensity of the anti-Stokes PL peak at 968 nm ($E5 \rightarrow E1$) becomes strong when $\lambda_{\text{exc}} = 1030$ nm (resonant excitation of the $E3 \rightarrow E5$ transition), which is the absorption peak wavelength of (Yb:Y)AG. This result is consistent with the relative cooling power curve in Fig. 2.

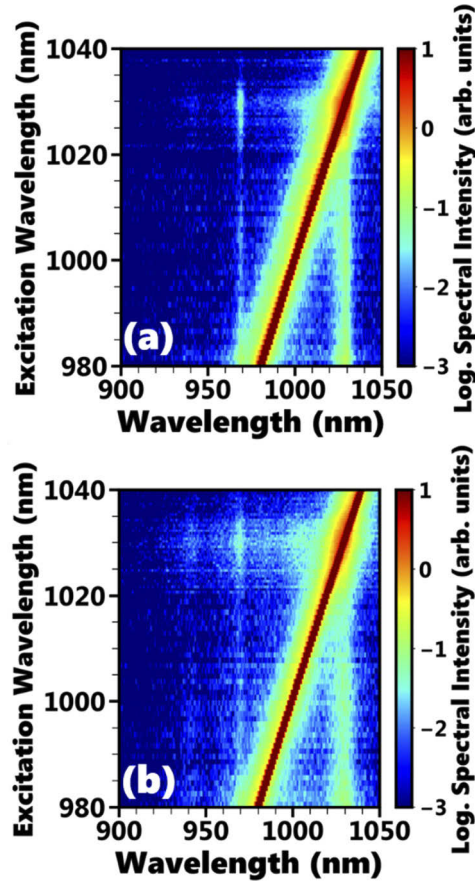


Fig. 4. PLE images of the (Yb:Y)AG sample with $x = 0.06$ at (a) 300 K and (b) 470 K. The horizontal axis, vertical axis, and color bar represent the PL wavelength, excitation wavelength, and logarithm of the spectral intensity, respectively. The strong signal following the linear function $\lambda_{\text{exc}} = \lambda$ corresponds to the detected (saturated) intensity of the scattered excitation light.

As expected from the temperature dependence of multiphonon absorption, the intensities of the non-resonant, broad anti-Stokes and Stokes PL signals observed at $T = 470$ K [Fig. 4(b)] are stronger than those at 300 K. To clarify the differences between the PL spectra measured at $T = 300$ and 470 K, the spectral intensity difference Δ between those two data sets is shown in Fig. 5. The enhancement (attenuation) of the PL intensity that occurs as the temperature is changed from 300 to 470 K, is indicated with positive (negative) values of Δ shown in blue (red). The non-resonant anti-Stokes and Stokes PL signals clearly become stronger with temperature,

while the resonant Stokes PL peak intensity at 1030 nm ($E5 \rightarrow E3$) and the resonant anti-Stokes PL peak intensity at 968 nm ($E5 \rightarrow E1$) decrease. Note that the term “resonant” in combination with (anti-)Stokes PL peaks is used to refer to the intra-orbital f-f transitions between the ideal energy states, which exhibit relatively narrow PL signals. Figure 6 shows the temperature dependence of the PL spectra of the (Yb:Y)AG sample under excitation with $\lambda_{\text{exc}} = 659$ nm. At low temperatures of 20 K, broadly distributed non-resonant signals are observed in addition to the sharp peaks discussed in Fig. 1. The broadly distributed non-resonant signals arise from the Yb^{3+} ions surrounded by inhomogeneously distributed atomic structures. The temperature increase leads to an enhancement of the phonon-assisted energy transfer from ideal states to the inhomogeneously distributed energy states.

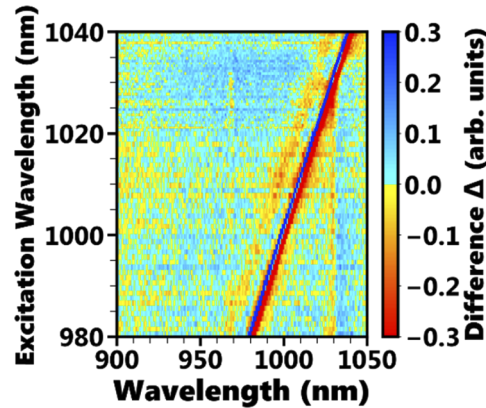


Fig. 5. Image of the PL intensity differences obtained by subtracting the spectral intensity at 300 K from that at 470 K (differences are shown on a linear scale).

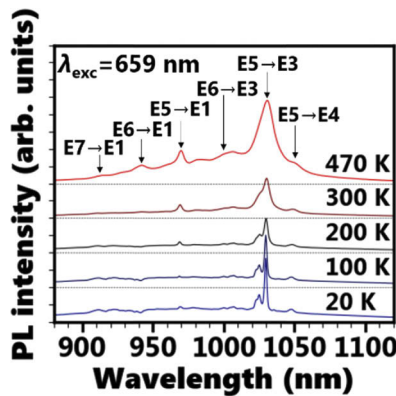


Fig. 6. PL spectra of the (Yb:Y)AG sample at $T = 20, 100, 200, 300,$ and 470 K under 659-nm excitation. Data are offset for clarity.

To clarify the detailed temperature dependence of the anti-Stokes PL spectra at elevated temperatures in the range from 100 to 470 K, the excitation wavelength was set to $\lambda_{\text{exc}} = 1030$ nm and the obtained spectra are provided in Fig. 7. At 100 K, the sharp anti-Stokes PL peak signal at 968 nm is not visible, but it can be clearly confirmed at $T = 125$ K. This peak's intensity and linewidth increase with temperature up to 300 K. Above 300 K, the intensity of the non-resonant anti-Stokes PL component significantly increases; the anti-Stokes PL peak at 968 nm is gradually buried under the signal of the non-resonant broad anti-Stokes PL as the temperature increases.

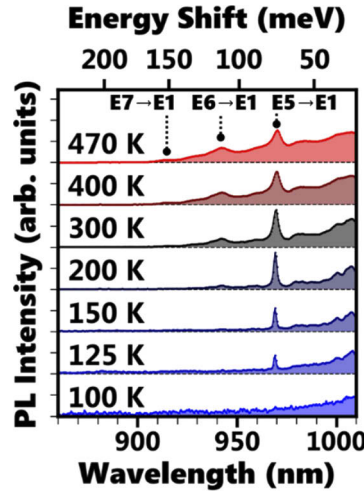


Fig. 7. Anti-Stokes PL spectra of the (Yb:Y)AG sample at $T = 100, 125, 150, 200, 300, 400$, and 470 K for $\lambda_{\text{exc}} = 1030$ nm (resonant excitation of the E3–E5 transition). The spectra are offset for clarity.

To discuss the temperature dependence of the intensity and spectral linewidth of the anti-Stokes PL, the resonant anti-Stokes PL peak at 968 nm was fitted by the following Lorentzian function:

$$A \left(\frac{\frac{1}{2}\sigma_h}{(E - E_0)^2 - \left(\frac{1}{2}\sigma_h\right)^2} \right) + B, \quad (5)$$

where A is the Lorentzian amplitude, B is the background intensity, σ_h is the full width at half-maximum representing the homogeneous linewidth, E is the energy, and E_0 is the central energy of the resonant anti-Stokes PL peak. As explained below, the temperature-dependent broadening of the anti-Stokes PL can be explained by a change in the homogeneous linewidth. The inhomogeneous linewidth is assumed to be negligible because the observed anti-Stokes PL signal is narrow.

Figure 8(a) plots the anti-Stokes PL spectrum observed at 300 K for $\lambda_{\text{exc}} = 1030$ nm and the fitting result. The temperature dependences of the integrated PL intensity of the peak at 968 nm and the background intensity of the anti-Stokes PL signal are shown in Fig. 8(b). As can be confirmed in Fig. 8(b), the integrated intensity of the resonant anti-Stokes PL peak at 968 nm initially increases with increasing temperature and reaches its maximum value near 300 K. This initial increase can be interpreted by considering the population of optical phonons. As shown in Fig. 7, the resonant E5–E1 anti-Stokes PL peak is observed for $T \geq 125$ K (which corresponds to thermal energies ≥ 10.8 meV). According to the lattice dynamics in RE aluminum garnets [27], Raman and infrared active optical modes are observed near 100 cm^{-1} (12.4 meV). This moderate agreement suggests that the optical phonon is excited in case of $T \geq 125$ K. Therefore, the optical phonon absorption process between E1 and E3 levels exhibits a threshold-like behavior at $T = 125$ K. On the other hand, the integrated intensity of the E5–E1 anti-Stokes PL decreases above $T = 300$ K despite the monotonically increasing phonon population at higher temperatures. To identify the origin of this behavior, we investigated the temperature dependence of the Raman signal corresponding to the high-frequency T_{2g} optical mode ($e_{7(12)1}$) in (Yb:Y)AG (data is shown in Fig. 9). The data evidences that the phonon energy decreases with increasing temperature, and thus the multiphonon absorption rate decreases at higher temperatures. In addition, as

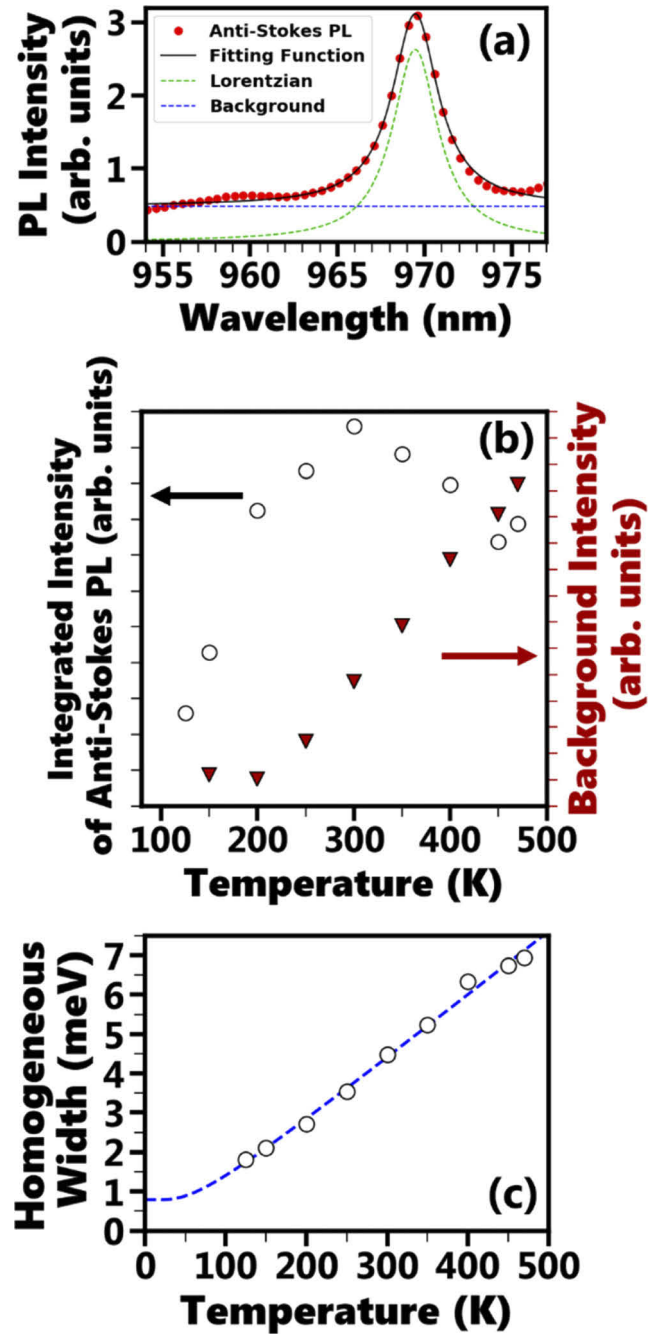


Fig. 8. (a) Fit of the anti-Stokes PL peak at 968 nm obtained from (Yb:Y)AG at 300 K for $\lambda_{\text{exc}} = 1030$ nm using a Lorentzian function and a constant for the background intensity. (b) Temperature dependence of the integrated intensity of the resonant anti-Stokes PL ($E_5 \rightarrow E_1$) and the background intensity of the non-resonant anti-Stokes PL in (Yb:Y)AG. (c) Temperature dependence of the homogeneous width of the anti-Stokes PL peak at 968 nm and the theoretical curve considering optical phonon scattering according to Eq. (6).

discussed above, the phonon-assisted energy transfer from ideal states (with narrow PL peaks) to inhomogeneously distributed energy states is enhanced as the temperature increases. These processes lead to a reduction in the integrated intensity of the resonant anti-Stokes PL peak at 968 nm. The trade-off relationship between phonon population and the energy transfer processes gives rise to the maximum at approximately 300 K.

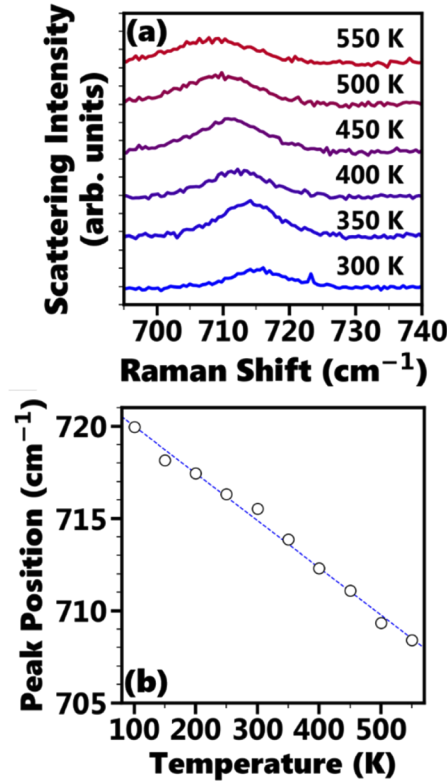


Fig. 9. (a) Temperature dependence of Raman spectrum of the high-frequency T_{2g} symmetry mode ($e_{7(12)1}$) in the (Yb:Y)AG ceramic. Data are offset for clarity. (b) The temperature dependence of the Raman peak position of the data in provided in (a). The blue dashed line is a linear fit to the data.

The second characteristic feature of the temperature dependence of the resonant anti-Stokes PL peak at 968 nm is that the peak width increases with temperature [Fig. 8(c)]. The blue dashed line in Fig. 8(c) represents the fitting result obtained using Eq. (6), which describes the temperature-dependent linewidth broadening by optical-phonon scattering,

$$\Gamma(T) = \gamma_0 + \frac{\gamma}{\exp\left(\frac{\hbar\omega_p}{k_B T}\right) - 1}, \quad (6)$$

The first term on the right side of Eq. (6), γ_0 , denotes the homogeneous linewidth at $T = 0$ K. Regarding the second term, γ is a coefficient for the electron–optical-phonon interaction, $\hbar$ is Dirac’s constant, $\hbar\omega_p$ is the optical-phonon energy, and k_B is the Boltzmann constant. The fitting result is $\gamma_0 = 0.79$ meV, $\gamma = 1.38 \times 10^{-3}$ meV, and $\hbar\omega_p = 14.6$ meV. We note that the Stokes PL peak of (Yb:Y)AG at 968 nm observed under resonant excitation with $\lambda_{\text{exc}} = 940$ nm (corresponding to the transition $E1 \rightarrow E6$), has a spectral linewidth of 1.65 meV at 4.4 K. Because the spectral linewidth consists of the inhomogeneous and homogeneous linewidth, the result

$\gamma_0 = 0.79$ meV is considered reasonable. Furthermore, the obtained optical-phonon energy of 14.6 meV coincides with the calculated and measured minimum transverse optical-phonon energy of 14.8 meV in YAG [28]. This excellent agreement for $\hbar\omega_p$ supports the model that explains the temperature dependence of the linewidth in Fig. 8(c) via homogeneous broadening due to the population of optical phonons.

6. High laser-cooling efficiencies above room temperature

To verify the effect of the competition between the enhancement of the non-resonant anti-Stokes PL and that of the Stokes PL, the evaluated spectra of ξ of the (Yb:Y)AG sample for $T = 300$, 350, and 470 K are shown in Fig. 10. We find that the maximum relative cooling power ξ is larger at higher temperatures. At 470 K, ξ is 1.8 times larger than that at 300 K because of the enhancement in the non-resonant anti-Stokes PL. The mean fluorescence wavelength of (Yb:Y)AG at $T = 470$ K is $\lambda_f = 1007$ nm, which is 9 nm shorter than $\lambda_f = 1016$ nm at 300 K. This value departs from previously reported values of approximately ~ 1009 nm [11,29]. The difference of the mean fluorescence wavelengths is caused by the decreased signal component of the shorter wavelength side of the PL spectrum of our ceramic disc sample, which can be interpreted by a relatively strong re-absorption in the ceramic disc. When we employ Eq. (3), we find that the predicted ideal cooling efficiency of (Yb:Y)AG for $\lambda_{exc} = 1030$ nm and $T = 470$ K is 2.2%, which is 1.7 times higher than the 1.4% at 300 K.

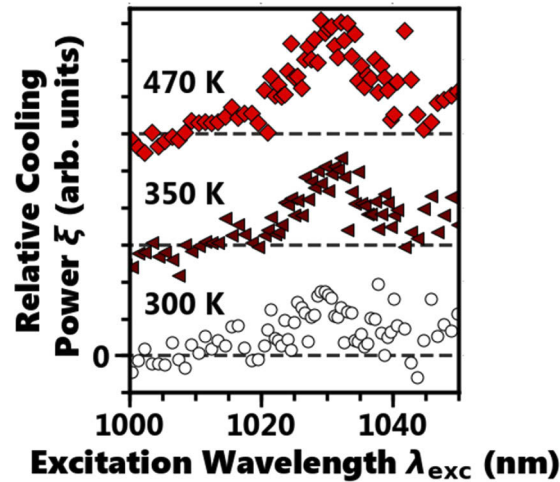


Fig. 10. Excitation wavelength dependences of the relative cooling power of (Yb:Y)AG at $T = 300$, 350, and 470 K. Data are offset for clarity.

From the practical perspective, it is helpful to indicate how much optical refrigeration is any better than passive cooling. The thermal conductivity of Yb:YAG decreases by 10% from $4.4 \text{ Wm}^{-1}\text{K}^{-1}$ at 300 K to $3.9 \text{ Wm}^{-1}\text{K}^{-1}$ at 470 K [30]. Contrarily, the ideal laser cooling efficiency is enhanced by 70% from 1.3% at 300 K to 2.2% at 470 K. Therefore, the enhancement rate of the ideal laser cooling efficiency is higher than the reduction rate of the thermal conductivity. On the other hand, the amount of heat transferred to the surroundings by radiative heat transfer strongly depends on the temperature gradient between the sample and ambient and the surface-volume ratio of the sample [31], and, therefore, the surface-volume ratio of a given substance determines a critical condition that the laser cooling surpasses the radiative heat transfer.

7. Conclusion

We propose that high laser-cooling efficiencies can be achieved by utilizing the significantly enhanced non-resonant anti-Stokes PL in (Yb:Y)AG ceramic discs at high temperatures. Our data clarifies that the non-resonant anti-Stokes PL intensity is enhanced above $T = 200$ K. In contrast, the integrated intensity of the resonant anti-Stokes PL decreases above 300 K. This is caused by two competing processes: the increased phonon population at higher temperatures (which increases the resonant anti-Stokes intensity) and the smaller phonon energy at higher temperatures (which decreases the intensity of the non-resonant anti-Stokes PL). We show that the relative cooling power ξ of (Yb:Y)AG with $x = 0.06$ at 470 K is 1.8 times larger than that at 300 K owing to the enhancement in the non-resonant anti-Stokes PL. The predicted ideal cooling efficiency η_c at 470 K is 2.2%, which is 1.7 times higher than that at 300 K.

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