Optically pumped infrared stimulated radiation in Pr^{3+}:Y_2SiO_5

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The infrared stimulated radiation of $^1D_2 \rightarrow {^3F_2}$ and $^1D_2 \rightarrow {^3H_6}$ transitions in Pr^{3+}:Y_2SiO_5 (YSO) via pulsed laser pumping has been observed. The threshold energy, temperature dependence and divergence angle for the stimulated radiation are also measured.

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Pr^{3+} (trivalent praseodymium), which has a large number of metastable multiplets ($^3P_{2,1,0}$, $^1D_2$, $^1G_4$), is an interesting laser material, and the generation of visible stimulated radiation from $^3P_0$ state in crystal and glass hosts was reported[1−4]. But up to now the observed stimulated radiation from $^1D_2$ multiplet is only related to $^1D_2 \rightarrow {^3F_3}$ and $^1D_2 \rightarrow {^3F_4}$ transitions. These are the stimulated radiations of $^1D_2 \rightarrow {^3F_3}$ transition in Pr^{3+}:$YAlO_3$ crystal and $^1D_2 \rightarrow {^3F_4}$ transition in Pr^{3+}:$KGd(WO_4)_2$ crystal[5−8].

In the study of promising laser materials, several papers dealing with the spectroscopy and the energy levels of Pr^{3+}:YSO as well as the visible stimulated radiation of $^3P_0 \rightarrow {^3H_5}$, $^3H_6$, $^3F_2$ transitions were reported[4,9,10]. But to our knowledge, there is no report on the stimulated radiation from $^1D_2$ multiplet up to now.

In this letter, we report the successful observation of the multiple stimulated radiation lines of $^1D_2 \rightarrow {^3F_2}$ and $^1D_2 \rightarrow {^3H_6}$ transitions in Pr^{3+}:YSO crystal via pulsed laser pumping. We have also measured the divergence angle, temperature dependence and threshold energy for the stimulated radiation.

The experimental setup is shown in Fig. 1. The sample temperature is varied between 10 and 300 K with a liquid helium cooled cryostat (ARS Displex GSW202N). The sample with a size of $5 \times 5 \times 3$ mm^3 and 0.05% Pr^{3+} in YSO is excited by a Q-switched Nd:YAG pumped R6G dye laser (pulses duration of 10 ns, line-width of 0.1 cm^{-1}, repetition rate of 10 Hz). The stimulated radiation signal in the forward direction is analyzed by a monochromator (spectral resolution of 0.05 nm, with a Hamamatsu R943 photon-multiplier), and treated by a boxcar (EG&G, Model 4152). The measurement of the fluorescence decay time at right angle to the direction of the laser beam is performed using a 500 MHz digital oscilloscope (HP54616C).

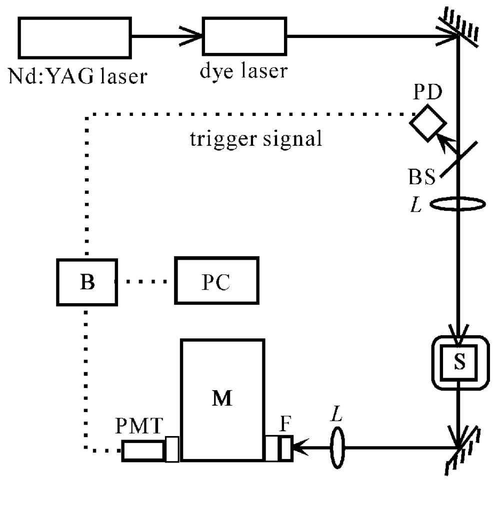

Fig. 1. Experimental set up infrared stimulated emission measurement. M: monocromator; B: boxcar; F: filter; L: lens; S: Pr^{3+}:YSO.

Under the action of the crystal field in YSO, states $^1D_2$, $^3F_2$ and $^3H_6$ are split into five, five and eight Stark components, respectively. A simplified energy-level diagram for of Pr^{3+} is shown in Fig. 2.

The infrared stimulated radiation lines observed from $^1D_2$ of Pr^{3+}:YSO are shown in Fig. 3. The wavelengths of the pumping laser at 578.0, 574.8 and 570.8 nm correspond to the $^3H_4(0) \rightarrow {^1D_2(5)}$, the $^3H_4^*(0) \rightarrow {^1D_2^*(4)}$ and the $^3H_4^*(0) \rightarrow {^1D_2^*(5)}$ transitions, respectively[10,11]. States $^3H_4^*$ and $^1D_2^*$ correspond to

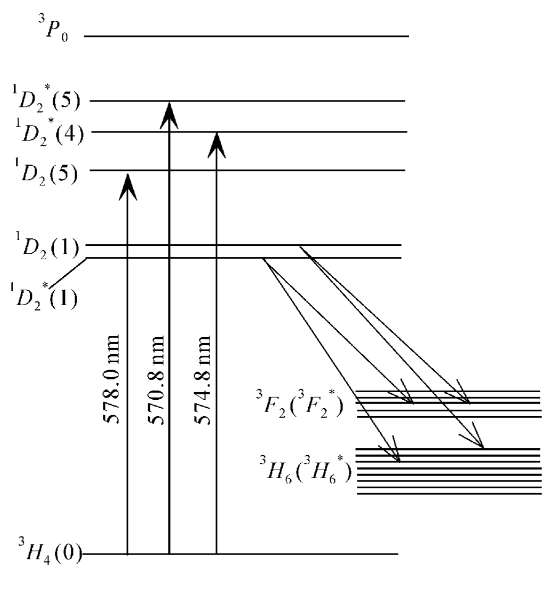

Fig. 2. Energy level diagram of Pr^{3+}:YSO.

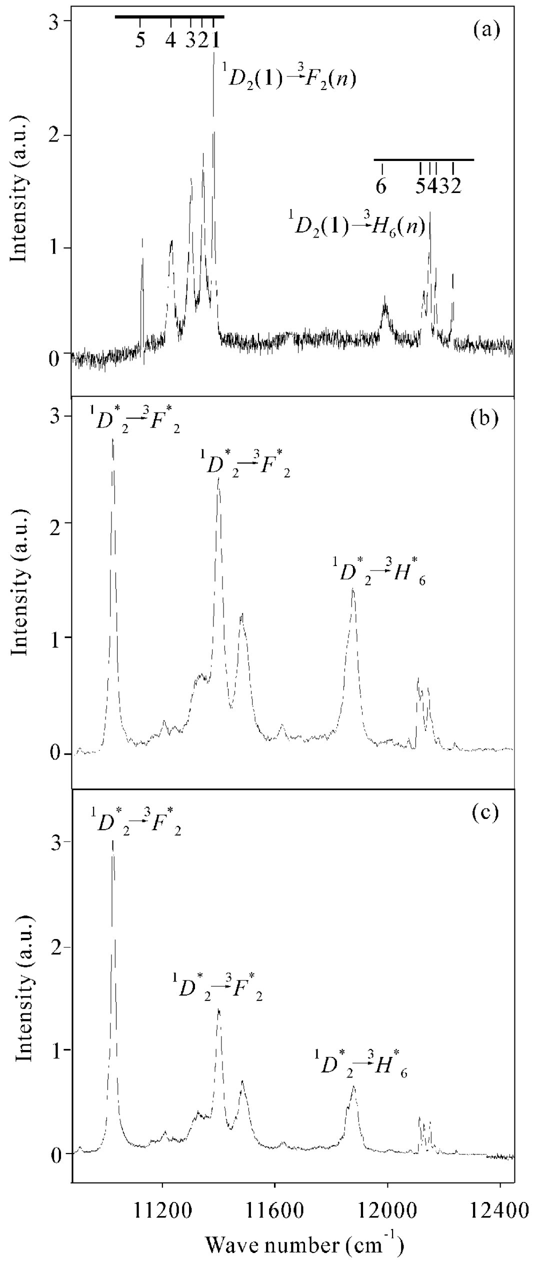

Fig. 3. Infrared stimulated radiation spectra of Pr^{3+}:YSO with the laser excitation wavelength of (a) 578.0, (b) 570.8 and (c) 574.8 nm.

the energy levels of another impurity center of Pr^{3+}:YSO (*represents another impurity center)[10,11]. At the liquid helium temperature, the population of the higher Stark splitting components of state $^1D_2$ will quickly transfered to the lowest one[11,12]. Using the results in Refs. [4] and [9], the measured infrared stimulated radiation lines shown in Fig. 3(a) can be identified. Up to now, for the Stark splittings of states $^3F^*_{2,3,5}$ and $^3H^*_{5,6}$ have not been determined, we can not determine the n value of Stark splittings in the stimulated radiation spectra corresponding to $^1D^*_2(1) \rightarrow {}^3F^*_2(n)$ and $^1D^*_2(1) \rightarrow {}^3H^*_6(n)$ transitions in Fig. 3(b) and (c).

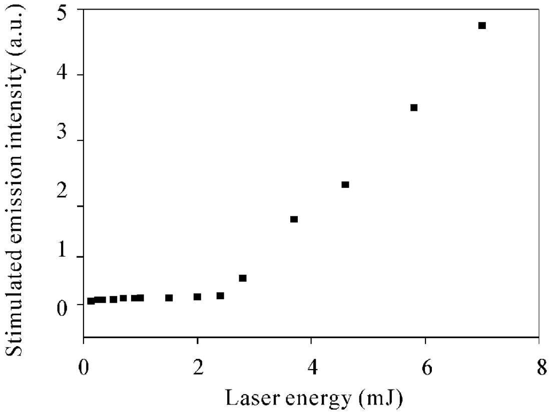

Fig. 4. Intensity of $^1D_2(1) \rightarrow {}^3F_2(2)$ stimulated emission versus laser energy.

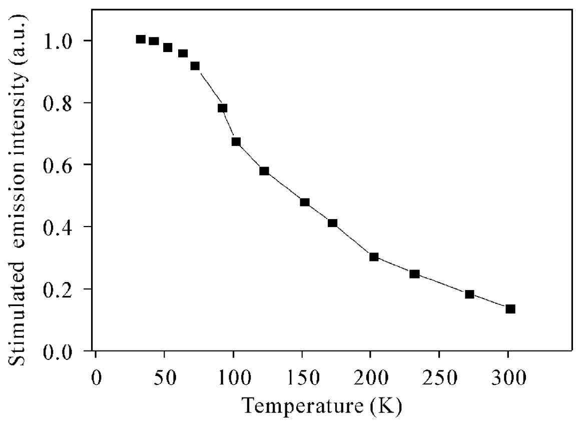

Fig. 5. Stimulated radiation intensity versus temperature in Pr^{3+}:YSO.

Figure 4 shows the change of stimulated radiation intensity at 11372 cm^{-1} of $^1D_2 \rightarrow {}^3F_2$ transition with laser energy. We can see that the stimulated radiation intensity rapidly increases when the laser energy is above the threshold energy of about 2.4 mJ. The divergence angle of 0.36 mrad is also measured. The threshold energy and the divergence angle for the other stimulated radiation lines are listed in Table 1.

Figure 5 shows the temperature dependence of the stimulated radiation intensity at 11372 cm^{-1}. The intensity becomes weaker with the raising temperature, but the stimulated radiation can be observed at room temperature. In addition, the temperature quenching of the population in $^1D_2$ is observed by measuring the decreased decay time and intensity of $^1D_2$ fluorescence with the raising temperature.

To our knowledge, this is the first time for the multiple stimulated radiation lines of $^1D_2 \rightarrow {}^3F_2$ and $^1D_2 \rightarrow {}^3H_6$ transitions in Pr^{3+}:YSO crystal to be observed.

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Table 1. The Measured Divergence Angle, Threshold Energy and Identified Stimulated Emission Transitions from $^1D_2$ Multiplet in Pr^{3+}:YSO

Laser Wavelength (nm)	Emission Transition	Wave No. (cm^{-1})	Threshold Energy (mJ)	Divergence Angle (mrad)
578.0	$^1D_2(1) \rightarrow {}^3F_2(1)$	11372	2.4	0.36
	$^1D_2(1) \rightarrow {}^3F_2(2)$	11316	2.4	0.36
	$^1D_2(1) \rightarrow {}^3F_2(3)$	11296	2.4	0.36
	$^1D_2(1) \rightarrow {}^3F_2(4)$	11223	2.4	0.36
	$^1D_2(1) \rightarrow {}^3F_2(5)$	11138	2.4	0.36
	$^1D_2(1) \rightarrow {}^3H_6(2)$	12222	3.0	0.36
	$^1D_2(1) \rightarrow {}^3H_6(3)$	12150	3.0	0.36
	$^1D_2(1) \rightarrow {}^3H_6(4)$	12172	3.0	0.36
	$^1D_2(1) \rightarrow {}^3H_6(5)$	12125	3.0	0.36
574.8	$^1D_2^* \rightarrow {}^3F_2^*$	11022 11391 11475	3.2	0.40
574.8	$^1D_2^* \rightarrow {}^3H_6^*$	11863	4.0	0.40
570.8	$^1D_2^* \rightarrow {}^3F_2^*$	11022 11391 11475	2.7	0.38
570.8	$^1D_2^* \rightarrow {}^3H_6^*$	11863	3.5	0.38

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