Solid-state green-light-emitting upconversion coherent random laser in macroporous $LiNbO_3$:Er

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A solid-state green-light-emitting upconversion coherent random laser was realized by pumping macroporous erbium-doped lithium niobate with a 980 nm laser. The lasing threshold was determined to be about 40 kW/cm^2. Above the threshold, the emission intensity increased sharply with the increasing pump intensity. Moreover, a narrow multi-peaks structure was observed in the green-light-emission band, and the positions of lasing lines were various at different angles. The results were the direct evidences of coherent random lasing emission from macroporous erbium-doped lithium niobate. These phenomena were attributed to the coexistence of upconversion emission and a multiple scattering feedback mechanism.

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Conventional lasers achieve feedback by an optical cavity with mirrors, while the cavity of random lasers results from multiple scattering and from light localization in a disordered gain medium that, above a threshold, leads to the sharp increase of the emission intensity. Since such random lasers were first proposed by Letokhov[1], they have recently attracted much interest and attention[2–11] and have been studied widely, because one can learn more about important scientific concepts, such as transport and localization in active random media, and because there are highly interesting technological applications, such as micro/nano lasers, laser paint, light-emitting devices, and display technology. There are two kinds of feedback mechanisms for random lasers: one is the intensity (energy) feedback mechanism; the other is the field (amplitude) feedback mechanism. The first one is incoherent, and the second one is coherent, so random lasing behaviors can be classified into two categories: (1) random lasing with an incoherent (or non-resonant) feedback mechanism, also called incoherent random lasing; (2) random lasers with a coherent (or resonant) feedback mechanism, also called coherent random lasing[2]. For any type of random lasers, in the emission spectrum, the dependence of integrated light-emitting intensities on excitation intensities exhibits obvious threshold behavior. For an incoherent random laser, when the pumping intensity exceeds a threshold, the emission spectrum narrows continuously toward a single peak, and the emission of such a laser does not exhibit spatial coherence. For a coherent random laser, because the feedback is supplied by a recurrent light current, it is coherent and resonant. For such lasers, when the pump intensity exceeds a threshold, a narrow multi-peaks structure emerges in the emission spectra, and the positions of these peaks depend on the observation angle[2].

In the early reports, random lasing merely exhibited incoherent emission, while random lasers with a coherent regime have been observed in various disordered systems in recent years. To date, coherent random lasing action has been demonstrated in the infrared[12], yellow[10], blue[13], and ultraviolet spectral range[2,14]. Considering that coherent random lasers are promising for various applications[2–4], achieving random lasers in the wavelength range where coherent random lasers were rarely investigated, especially in visible range, seems quite attractive[10,11,13,15]. In the green spectral range, a plasmonic random laser was demonstrated[11], but the narrow multi-peaks structure, which is one of the most important characteristics of coherent random lasers[2,13], was not observed in the green-light-emission band. This phenomenon indicated that it was an incoherent random laser. Using a special dye and TiO_2 nanoparticles, a coherent random laser emitting at 560 nm was achieved in the colloidal dye solution, but not in the solid-state system[15].

Since green is one of the primary colors and a solid-state laser is compact, portable, and stable, developing a solid-state coherent random laser in the green spectral region is of high interest for some practical applications. Moreover, most studies were based on down conversion emission, i.e., random lasers emitting photons of lower energy than the pump photon energy. An upconversion emission scheme can significantly improve the capabilities of random lasers. Not surprisingly, developing various new types of random lasers based on upconvesion emission is one of the hot topics in optics[16–19]. One advantage of upconversion random lasers is that the threshold is decreased[17], so the upconversion mechanism supplies an idea to achieve a green-light-emitting coherent random laser at relatively low pump intensities. These advantages make solid-state

upconversion coherent green-light-emitting random lasers attractive.

In this Letter, we report a solid-state green-light-emitting coherent random laser in macroporous erbium-doped lithium niobate (LN:Er) based on an upconversion mechanism. Lasing action from the green-light-emission band (from 520 to 570 nm) was induced by a 980 nm excitation laser in macroporous LN:Er and was quite different from the fluorescence behavior in bulk LN:Er.

The sample under study here was macroporous LN:Er. LN:Er is a quite important material for integrated optics applications, because it combines emission and gain properties of Er^{3+} with the excellent electro-optic, acousto-optic, and nonlinear optical properties of LN[20–22]. Its energy levels and favorable upconversion properties permit emission of green light under excitation at 980 nm. A further reason to prefer LN over other host materials for systems based on random scattering is stronger scattering due to its high refractive index (about 2.2–2.3)[23]. LN:Er is therefore the material of choice for our purpose.

In this work, an LN single crystal doped with 0.2 mol % Er^{3+} was grown by the Czochralski method and ground to small particles by a planetary ball mill. The LN:Er particles passed through a 220 nm membrane filter and were dispersed in deionized water. Then, the suspension was dried up at 60°C in a glass beaker. The thickness of the resulting sample was about 2.0 mm. The average size of the LN:Er particles was several hundred nanometers, as evaluated with an atomic force microscope (AFM) (see inset of Fig. 1). Although the LN:Er microcrystals are anisotropic, the macroporous LN:Er sample is apparently isotropic because the scatterers are randomly oriented[23–25].

Random lasing of the macroporous LN:Er sample was excited by a 980 nm picoseconds laser with 5 ps duration. The excited light impinged perpendicularly to the sample surface and was focused by a lens to a spot with a diameter of 120 μm. Emission spectra were collected by a spectrometer in a direction 30° from the front surface of the sample.

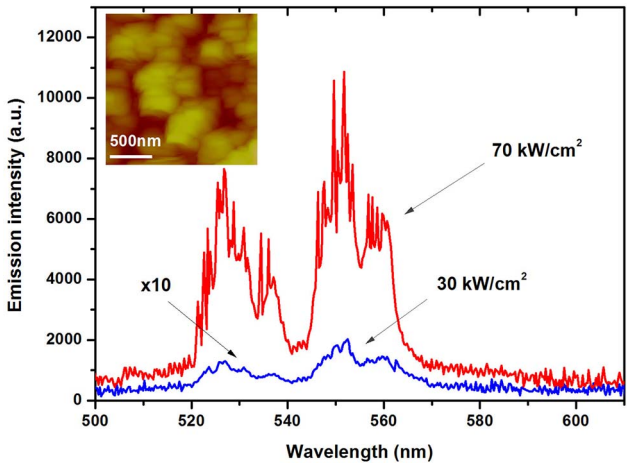

Fig. 1. Emission spectra of the macroporous LN:Er sample at different excitation intensities (30 and 70 kW/cm^2, inclination angle: 30°). The inset is an AFM image of the sample.

Figure 1 shows the photoexcited upconversion emission spectra at different pump intensities. The emission band was from 520 to 570 nm. At low intensity (30 kW/cm^2), only a broad emission band can be made out. However, obvious narrow emission peaks appeared at high excitation intensity (70 kW/cm^2). The integrated green-light-emission intensity was shown in Fig. 2(a) for the green-light-emission band as a function of excitation power. The lasing threshold was determined from this figure to be about 40 kW/cm^2. The slope above the threshold was larger than below. Below the threshold, the intensity of the green light increased slowly (slope: 1.73). Above the threshold, its intensity increased sharply (slope: 5.73). For comparison, we pumped bulk LN:Er in the same experimental configuration. In contrast to the distinguished onset of random lasing action marked by a change of the slope at the threshold intensity, there was no such threshold with increasing pump intensity [Fig. 2(b)]. The fluorescence emission simply increased smoothly with nearly quadratic intensity dependence.

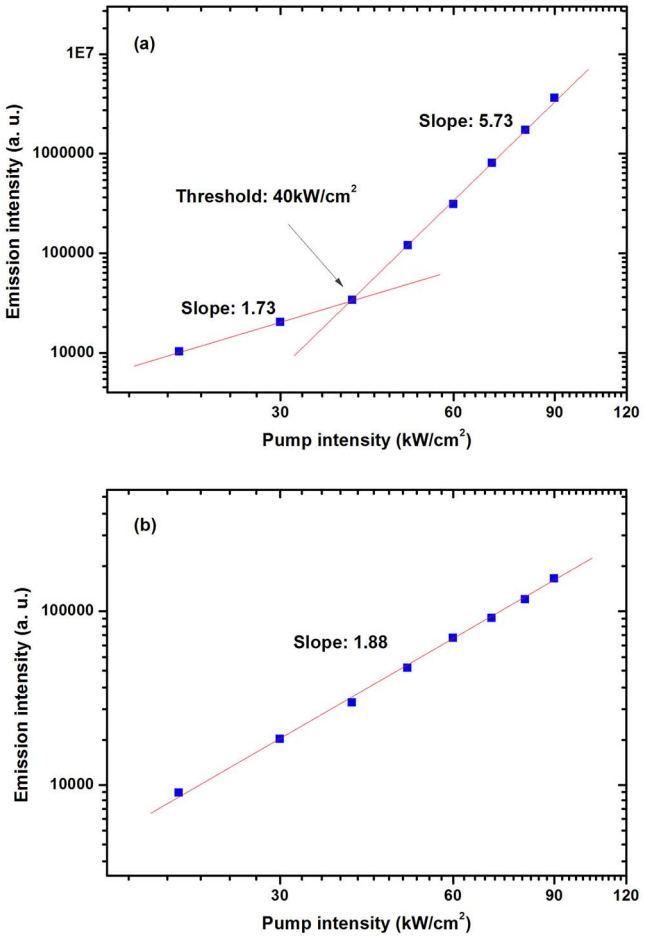

Fig. 2. Integrated emission intensity as a function of the excitation intensity on a log-log scale for (a) macroporous and (b) bulk LN:Er. The intensities were integrated from 520 to 570 nm (green-light-emission band). Solid lines are the pertinent linear fits.

Actually, the narrow multi-peaks structure in the emission spectrum is one of the most important characteristics of coherent random lasers, and another key characteristic is that the lasing emission spectra vary with the observation angle above the threshold. One can distinguish between a coherent regime and an incoherent regime according to both characteristics. As shown in Fig. 1, the narrow multi-peaks structure was demonstrated. We also measured spectra at 70 kW/cm^2 (above the threshold: 40 kW/cm^2) for directions at 20° (Fig. 3) and at 30° (Fig. 1) from the sample surface. The fact that the spectra were different further indicated that lasing was in the coherent feedback regime.

As shown in Figs. 1 and 3, the multi-peaks in the green-light-emission band can be classified into two bands centered at 550 and 527 nm, respectively. This phenomenon was attributed to the coexistence of upconversion emission and a multiple scattering feedback mechanism induced by multiple scattering. Upconversion emission of Er^{3+} results from excited-state absorption (ESA) as well as from energy transfer upconversion (ETU). The green emission is due to transitions from two Er^{3+} levels: the 527 and 550 nm emission bands are assigned to the $^2H_{11/2} \rightarrow {}^4I_{15/2}$ and $^4S_{3/2} \rightarrow {}^4I_{15/2}$ transitions, respectively. The processes are shown in Fig. 4. In the ESA process, the Er^{3+} is first excited from the ground state to the $^4I_{11/2}$ state (GSA) by pumping with 980 nm infrared light. Since the lifetime is sufficiently long, some of the ions in the $^4I_{11/2}$ level may absorb another 980 nm photon to populate the higher $^4F_{7/2}$ level. In the ETU process, after the ion has been excited to the $^4I_{11/2}$ level by the laser beam, a neighboring Er^{3+} ion that is also in the $^4I_{11/2}$ state transfers its energy to the initial ion, thereby exciting it to the $^4F_{7/2}$ level. In Er^{3+} ions-doped materials, upconversion for 520/540 nm emission can be excited by both ESA and ETU. Generally, the ESA process from dopant ions is the dominant one in samples with a concentration of 0.5 mol % or lower[26]. Because the sample used in this work is doped with 0.2 mol % Er^{3+}, the dominant upconversion

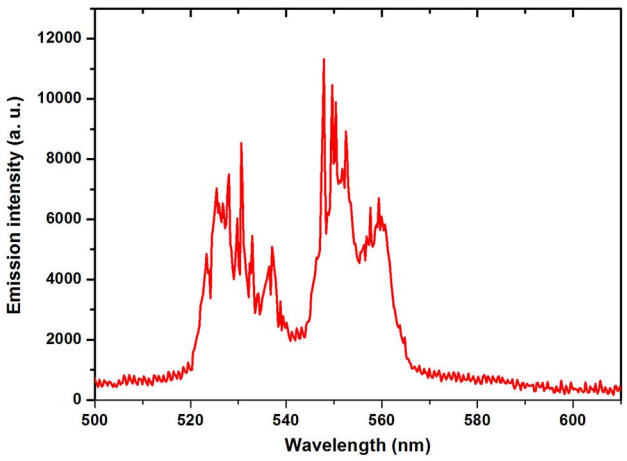

Fig. 3. Emission spectrum of macroporous LN:Er at 20° from the sample surface (pump intensity: 70 kW/cm^2).

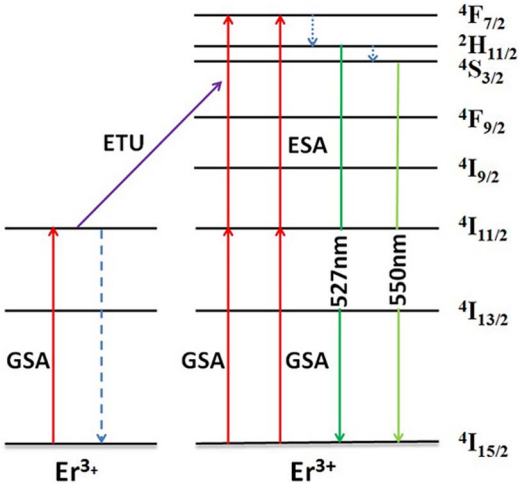

Fig. 4. Energy levels of Er^{3+} showing the ESA and ETU pathways under pumping by a 980 nm.

mechanism is the ESA process. In both cases, nonradiative relaxation from the $^4F_{7/2}$ level populates the levels $^2H_{11/2}$ and $^4S_{3/2}$. Finally, the transitions from $^2H_{11/2}$ and $^4S_{3/2}$ to the ground state generate the emission of green light at 527 and 550 nm. The reason that the intensity of the emission from $^4S_{3/2}$ is higher than from $^2H_{11/2}$ is because of the existence of efficient nonradiative relaxiation from $^2H_{11/2}$ to $^4S_{3/2}$. Figure 2 shows that the slope value above the threshold is much higher than the one below the threshold. It indicates that random lasing action occurred in the macroporous LN:Er sample. Because upconversion light is strongly scattered in the disorder medium, many closed light loop paths can be formed through multiple scattering, and these loops could serve as cavities for light. Below the threshold, the gain of emitted light is less than the loss, and it is the traditional upconversion fluorescence phenomenon. Therefore, the emission intensity rises up slowly with the increasing of the pump intensity. Above the threshold, the gain exceeds the loss in such cavities. Then, lasing behaviors occur and lead to rapid increasing of the emission light. Hence, the slope value above the threshold is much higher than the one below the threshold. Moreover, the thresholds reported in most publications were several hundred kW/cm^2[2,12,13] and thus higher than the threshold for macroporous LN:Er. This is caused by the high efficiency of exciting gain materials in the upconversion disordered medium. According to the scattering theory, the pumping light with the near-infrared wavelength underwent weaker scattering than the upconversion green-light-emitting emission band. Hence, efficient pumping of high-Q random cavities can be realized. As a result, a low threshold random laser is able to be achieved in an upconversion random gain medium[17,19].

In conclusion, a solid-state green-light-emitting upconversion coherent random laser was developed using macroporous LN:Er. The lasing emission band (from 520 to 570 nm) can be excited by a 980 nm laser. Moreover, the multi-peaks structure was observed, and the lasing

spectra fluctuated randomly with the emission angle. These phenomena were attributed to the coexistence of upconversion emission and a multiple scattering feedback mechanism. The results make macroporous LN:Er appear promising for an application as micro-sized active elements in photonic devices.

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