

Switching of emissivity and photoconductivity in highly doped $\text{Yb}^{3+}:\text{Y}_2\text{O}_3$ and Lu_2O_3 ceramics

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Reversible jump of thermal emission accompanied with jump of photocurrent is observed in highly doped $\text{Yb}^{3+}:\text{Y}_2\text{O}_3$ and Lu_2O_3 bulk ceramics pumped at 940 nm wavelength. In contrast, when these materials are heated with a CO_2 laser at 10.6 μm wavelength, only a gradual increase of thermal emission and photoconductivity are observed up to the melting point. These results are interpreted as a ytterbium mediated, photoassisted avalanche of thermal emission. © 2007 American Institute of Physics. [DOI: 10.1063/1.2739318]

Substantial efforts are being made to improve lighting sources for larger fractional output in the visible, tailored emission spectrum for improved object rendering, or higher wall-plug conversion efficiency. These include the development of suitable combination of light-emitting diodes and phosphors,^{1–3} or search for alternatives to the toxic mercury-based fluorescent tubes.^{4,5} The control of thermal emission from heated materials also attracts a lot of interest. Periodically microstructured surfaces have been shown to provide spectrally selective outputs.^{6–8} The control of thermal emission from photonic crystals has also been used to suppress unwanted infrared emission at the photonic band gap.^{9–11}

We report reversible switching of the emissivity in Yb^{3+} -doped sesquioxide, Y_2O_3 , and Lu_2O_3 . These are refractory materials with a melting point of ~ 2450 °C,¹² but they are highly transparent in the 0.2–8 μm range¹³ and they are electrical insulators. We found an avalanche of broadband emission, with a spectrum similar to the thermal radiation of a blackbody, with the simultaneous jump of electrical conductivity in these materials when pumped at an absorption resonance of the Yb^{3+} doping ion. No such phenomenon took place when heating the samples to temperatures well above the melting point with a CO_2 laser.

This phenomenon was discovered when studying the luminescence properties of highly Yb^{3+} -doped (10%) Lu_2O_3 and (15% and 20%) Y_2O_3 polycrystalline ceramics¹⁴ pumped at 940 nm wavelength. Millimeter size samples without special surface finish were used. The samples were pumped with a 200- μm -core fiber-coupled laser diode (LIMO25-F200-DL940) focused to a 200 μm spot at the edge of the sample using a pair of aspherical lenses ($f=8$ mm). The luminescence signal was collected perpendicular to the pump axis and analyzed with a spectrum analyzer (ANDO AQ-6315A). The sample transmission was simultaneously measured at 632.8 nm wavelength with a He–Ne laser probe coaxial with the pump. The transmitted beam was detected with a silicon detector, while the transmitted pump light and infrared luminescence were blocked with filters. The probe beam was

chopped at 213 Hz and sent to a lock-in amplifier in order to separate the probe light from the visible emission coming out from the sample.

The Lu_2O_3 sample was pumped with about 4.5 W (~ 14 kW/cm²) for about 8 s with 50% duty cycle. The luminescence, measured at $\lambda=1030$ nm with 5 nm resolution, rose rapidly and then decreased to a small fraction of its maximum value (Fig. 1). Then, the luminescence abruptly dropped and a jump of broadband thermal emission was observed. A drop of transmission at 632.8 nm of about 40%, synchronous with the drop of Yb^{3+} luminescence, was observed (Fig. 1) which indicates the jump of absorption at $\lambda=632.8$ nm when thermal emission took place. The sample recovered its original transmission value when the pump was cut.

The sequence of Yb^{3+} luminescence, its quenching, and strong thermal emission was reproducible over the duration of the experiment of about 1 h. The pumping power, required to achieve bright broadband emission (BBE), increased when

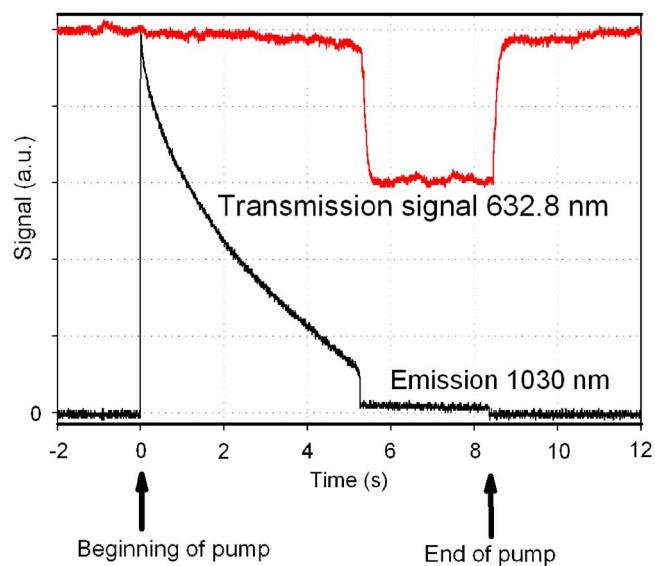


FIG. 1. (Color online) Quenching of luminescence in 10% $\text{Yb}:\text{Lu}_2\text{O}_3$ pumped at 940 nm and transmitted signal at 632.8 nm. Delay of the transmission signal is due to the time constant of the lock-in amplifier, 100 ms.

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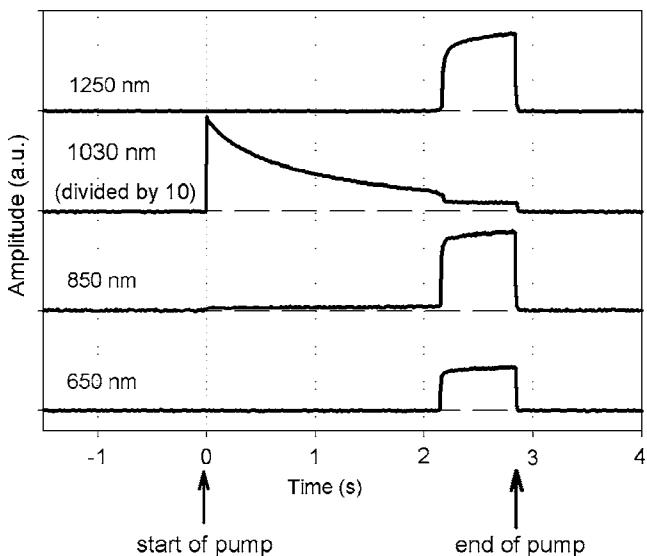


FIG. 2. Oscilloscopes obtained for 10% Yb:Lu₂O₃ at 650, 850, 1030, and 1250 nm. The curves are vertically shifted for clarity.

shorter pumping cycles were used. Signals centered at 650, 850, 1030, and 1250 nm with more than 5.5 W, 2.8 s long pulses are shown in Fig. 2. No detectable signal existed at 650 nm before the onset of BBE, at which point the signal jumped up by two orders of magnitude above the noise level within 3 ms and then rapidly flattened. Similar results were obtained at other wavelengths.

The quenching of luminescence has been reported in nanocrystalline Yb³⁺, Er³⁺:Y₂O₃ powders.^{15,16} The authors attributed their observations to nonradiative decay by a multiphonon relaxation process. However, from the evolution of the signal at 850 nm, which arises from thermal emission, we found that the temperature increases very fast at the beginning of the pulse and then barely increases during the rest of the pulse, while the Yb³⁺ luminescence at 1030 nm steadily decreases (Fig. 2). This result suggests that the quenching of Yb³⁺ luminescence is not entirely due to multiphonon relaxation process to the ground state.

The emission spectra taken at different pumping powers with cw pumping are shown in Fig. 3 for the 15% Yb:Y₂O₃

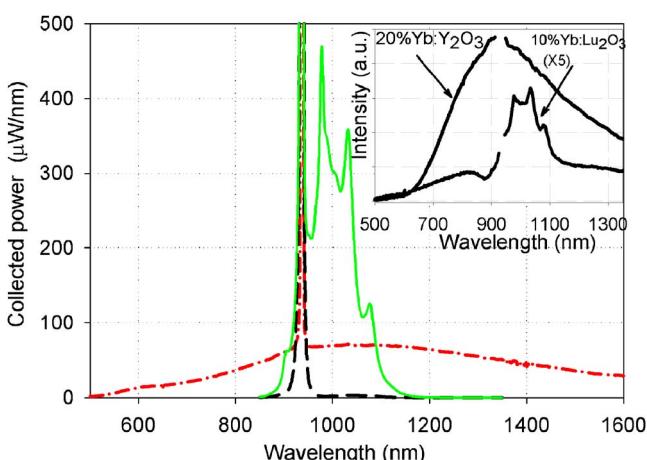


FIG. 3. (Color online) Emission spectrum from 15% Yb:Y₂O₃ taken at lower power (2 W) at higher power below (dashed) and above (dotted dash) the threshold of broadband emission at about 4 W pump power. The peak at around 940 nm is the pump. Inset: spectra obtained in 10% Yb:Lu₂O₃ (5.5 W power) and 20% Yb:Y₂O₃ (3.5 W).

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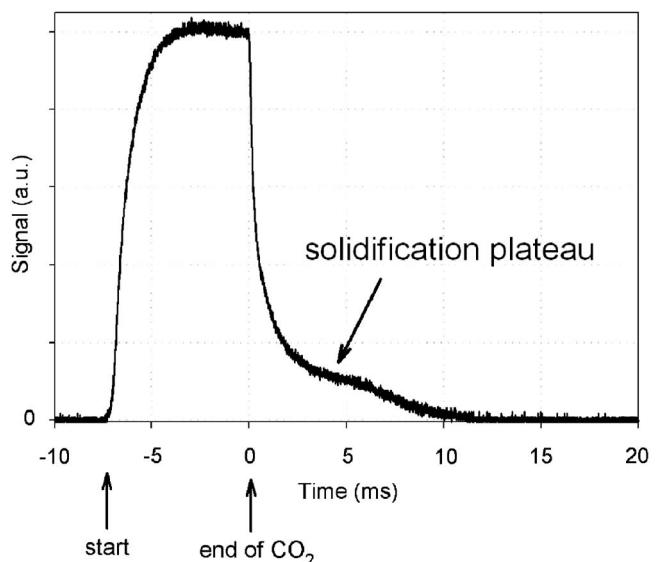


FIG. 4. Pyrometric signal at 850 nm from 10% Yb:Lu₂O₃ irradiated at 10.6 μm .

sample. Well below the threshold of BBE, the luminescence spectrum from ytterbium ions is dominant (solid line). As the pumping power is increased, the luminescence from ytterbium disappears and thermal emission is barely detected (dashed curve). Increasing only slightly the pumping power leads to a jump of broadband emission (dash dotted), which remained when the pump power was reduced by several percents, indicative of a bistable phenomenon, similar to that reported with rare-earth-doped yttria nanopowders.¹⁷ Emission spectra of the 20% Yb:Y₂O₃ and 10% Yb:Lu₂O₃ samples, taken at the threshold of BBE, are shown in the inset of Fig. 3. The former displayed the strongest signal. The sample showed traces of melting after the experiment. The spectrum from Lu₂O₃ displayed emission from Yb³⁺ because the heating was less severe. Samples with lower concentration of ytterbium were also tested, but the jump of emission described herewith could not be observed.

To determine whether the jump of emission is connected to the pumping of ytterbium, the sample was heated using a CO₂ laser ($\lambda=10.6 \mu\text{m}$) focused to a diameter of about 100 μm . In contrast to Yb³⁺ pumping, a gradual increase of pyrometric signal with pumping time was observed, when similar power was used, up to the melting point. When the sample surface melted, the solidification plateau was observed at the cool down, without sudden drop of emission (Fig. 4). This result suggests that the jump of emission (Fig. 2) is not caused by the melting of the sample, although melting was found to occur in the 20% Yb:Y₂O₃ sample, possibly due to a jump of absorption at the pump wavelength.

Time-resolved photoconductivity measurements were carried out to detect the generation of charge carriers, when pumping at 940 nm. A piece of 10% Yb:Lu₂O₃ of about 200 μm thickness was pressed between copper electrodes. The sample was placed in series with a 100 k Ω resistor. The circuit was driven with a 30 V dc power supply. The voltage at the resistor was used to monitor the current through the sample when pumped at 940 nm. At low pump densities, neither BBE nor photocurrent was detected. When more than 6 W pump power was focused on the sample, jumps of thermal emission and photocurrent simultaneously took place, indicating that charge carriers are produced (Fig. 5).¹⁸ Both

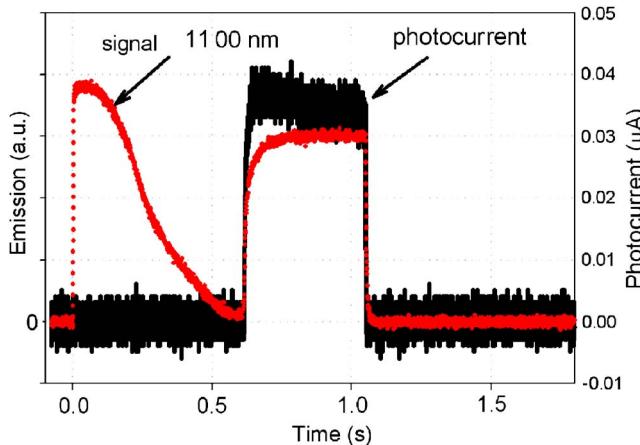


FIG. 5. (Color online) Signal at $\lambda=1100$ nm and photocurrent from 10% Yb-doped Lu_2O_3 sample pumped at 940 nm.

signals dropped when the pump pulse was turned off. The photoconductivity signal was stable from pulse to pulse over more than 1000 shots of the duration of the experiment. Observation of the sample with the optical microscope showed that the sample was fractured during the experiment, probably because of thermal stress arising from the heat flow through the copper electrodes. Photocurrent was also detected when irradiating with a CO_2 laser, but the photocurrent increased slowly with time, over a 0.1 s time scale.

In summary, a reproducible avalanche of thermal emission was observed in highly doped Yb^{+3} -doped Lu_2O_3 and Y_2O_3 bulk ceramics when pumped at an absorption resonance of Yb^{+3} . Reversible jumps of emission coinciding with the jump of photocurrent were observed. These features were absent when the samples were heated with a CO_2 laser, which indicates that the avalanche process is mediated by ytterbium ions. More work is under way to elucidate the mechanisms and possible applications of this phenomenon.

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- ¹⁸See EPAPS Document No. E-APPLAB-90-017720 for a video of the luminescence, its quenching, and the broadband emission of $\text{Yb}^{3+}:\text{Lu}_2\text{O}_3$ sample taking place during the photoconductivity measurements. This document can be reached via a direct link in the online article's HTML reference section or via the EPAPS homepage (<http://www.aip.org/pubservs/epaps.html>).