Laser induced anti-Stokes white emission nature from Yb-doped YAG nanopowders

Chaika M., Tomala R., Strek W.
m.chaika@intibs.pl

INTiBS PAN WROCŁAW

Introduction

The laser Induced Broadband Emission (LIBE) has recently been detected by several groups as a source of sun-like emission spectra [1-4]. LIBE is a characteristic threshold phenomenon that occurs under a focused beam of continuous infrared laser. This phenomenon was detected in vacuum ambient; a decrease in ambient pressure below a certain point leads to disappearance of LIBE. Several possible mechanisms of LIBE in different materials have been proposed. Due to the lack of experimental data, the mechanism behind this process is still not entirely understood.

Methods

The laser induced broadband emission (LIBE) of $(Yb_xY_{1-x})_3Al_5O_{12}$ nanocrystals was investigated in vacuum using the focused infrared laser beam [1]. The samples were prepared via Pechini's technique. The different shapes of LIBE spectra were observed. The samples (Fig. 1) were excited by a focused laser (980 nm) with a laser cross section up to 0.175 mm, and maximum laser output of 2 W ($5.5 \cdot 10^3$ W/cm^2 on the sample surface). Emission spectra were measured using two types of detectors: AVS-USB2000 Avantes Spectrometer and Ocean Optics NIRQuest512-2.5 Spectrometer for the visible and near infrared spectra, respectively.

Fig. 1 The photo of LIBE under excitation at 980 nm of the $(Yb_xY_{1-x})_3Al_5O_{12}$ nanocrystals

Results

Emission intensity increases sharply with increasing excitation power after exceeding the threshold (Fig. 2). For Yb:YAG nanocrystals the order of the process is in the range of 4-10 for Vis and 2-4 for NIR. The emission is characterized by an enormously large Stokes shift in the range from 2 eV to 10 eV (from 16000 cm^{-1} to 80000 cm^{-1}). The time dependence of LIBE intensity I(t), is given by $I(t) = Ie^{(t/\tau)}$. The observed decay times taken in the Vis and NIR parts of the spectrum were 0.3±0.1 s and 0.3±0.2 s, respectively. The build-up times of Vis and NIR were 4±2 s and 4±2.5 s, respectively. No clear difference between decay and build-up times was observed for Vis and NIR. So, Anti-Stokes (Vis) and Stokes (NIR) parts of the emission spectra are caused by the similar phenomena [1].

The LIBE characteristics including an order of the process, rise and decay times, were independent on the type of emission spectra and concentration of Yb^{3+} ions. Intensity of the emission spectra decreased/increased with time after switching off/on the laser excitation, while the shape of the emission spectra remained approximately the same. This is consistent with the colour of the emission, which remains white even after significant decrease in intensity. The color of the sample in the emitting spot changes from white to dark blue caused by formation of Yb^{2+} ions. The generation of broadband emission begins after reaching a certain "time delay" in range 0-10 seconds (Fig. 3).

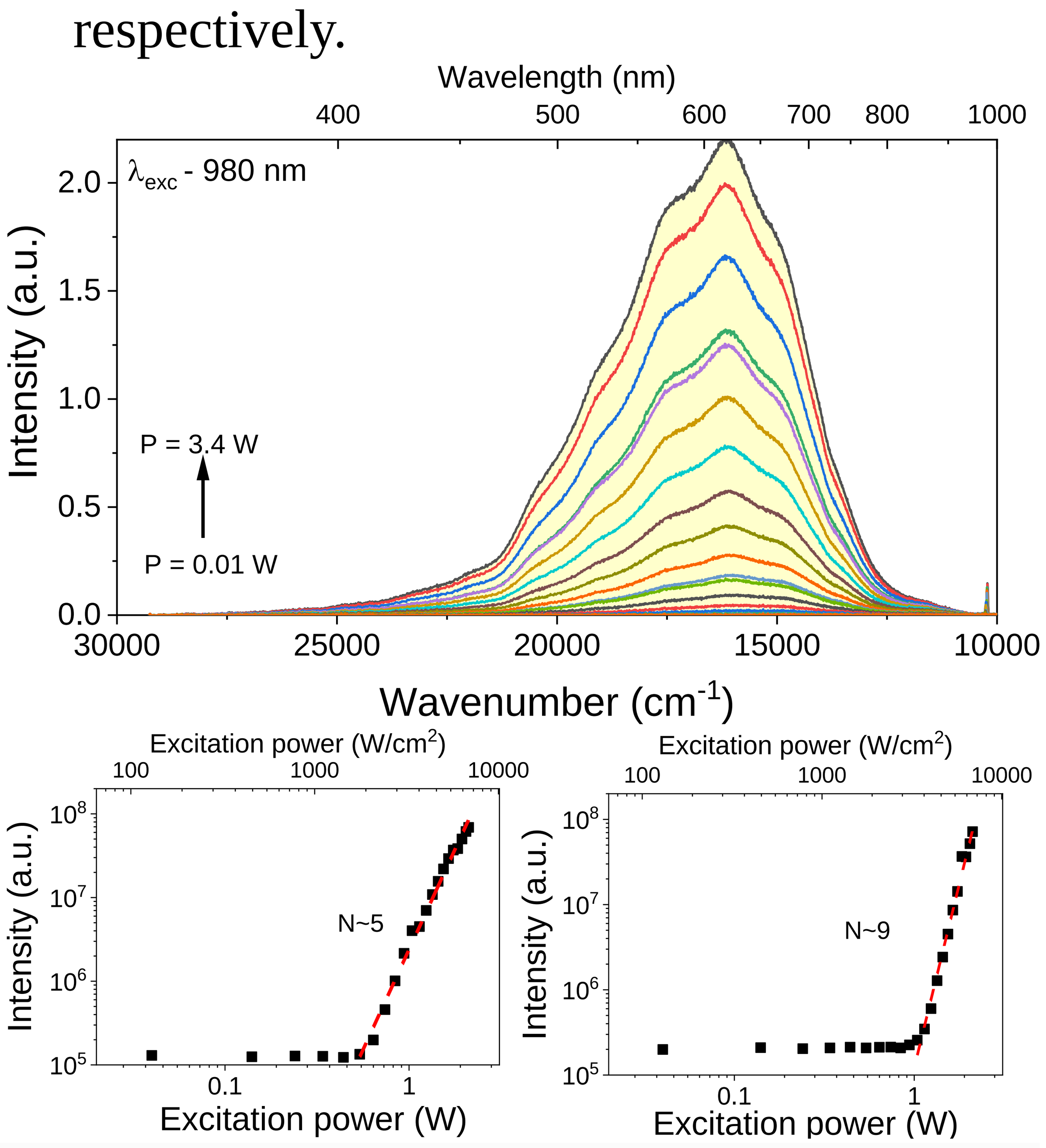

Fig. 2: The photo of LIBE under excitation at 980 nm of the $(Yb_xY_{1-x})_3Al_5O_{12}$ nanocrystals

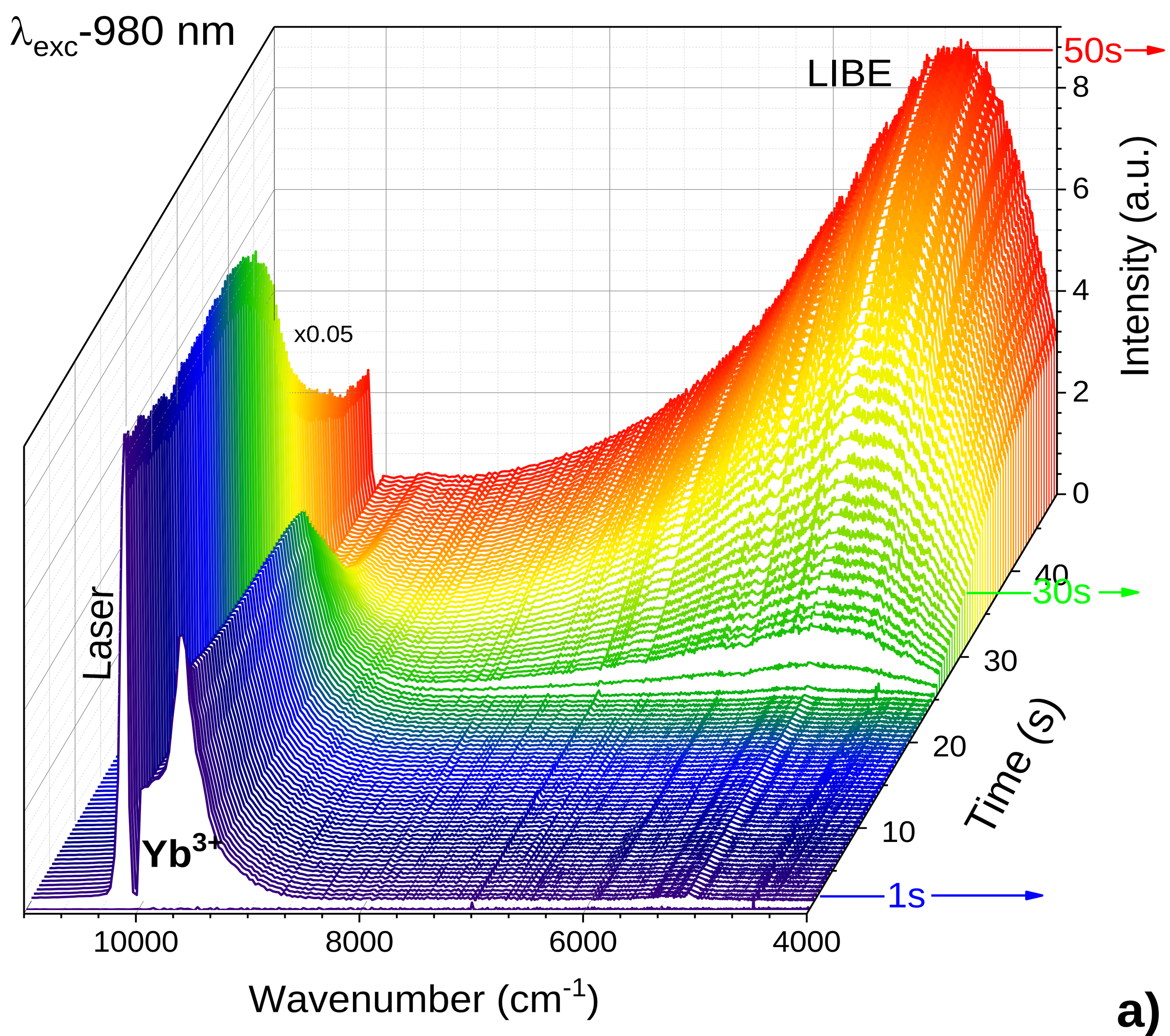

Fig. 3 Temporal evolution of the emission spectra of the $(Yb_xY_{1-x})_3Al_5O_{12}$ nanocrystals

Conclusion

Beforehand, several possible LIBE mechanisms have been proposed for different materials, however they don't cover all aspects of this phenomenon. All of these mechanisms are based on multiphoton absorptions with further propulsion of an electron into the conduction band/upper state where the electron emit high energy photons. The most popular explanation of LIWE in RE-doped materials is Inter Valence Charge Transfer (IVCT) process in Yb^{2+}/Yb^{3+} mixed valence pair for example (Fig. 4) [1-4]. IVCT mechanism may be responsible for multiphoton absorptions of laser light and, as result, promotion of electrons to the conduction band with further radiative recombination.

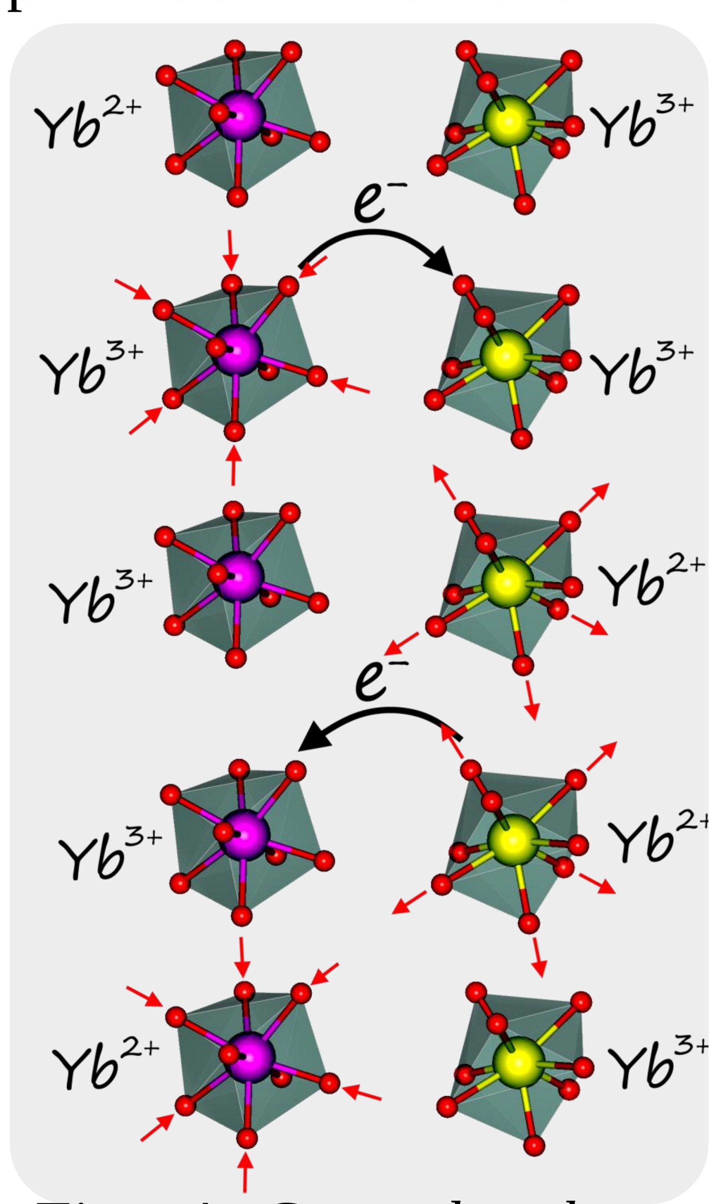

Fig. 4 General scheme related to IVCT

Literature

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[3] Chaika, M. A., Tomala, R., & Strek, W. (2021). Infrared laser stimulated broadband white emission of transparent Cr:YAG ceramics obtained by solid state reaction sintering. **Optical Materials**, 111, 110673.

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Contact information

Mykhailo Chaika, Institute of Low Temperature and Structure Research PAS. Poland. **Tel. +48 514 221 675**