Two-photon absorption and stimulated emission in poly-crystalline zinc selenide with femtosecond laser excitation

February 17 2022

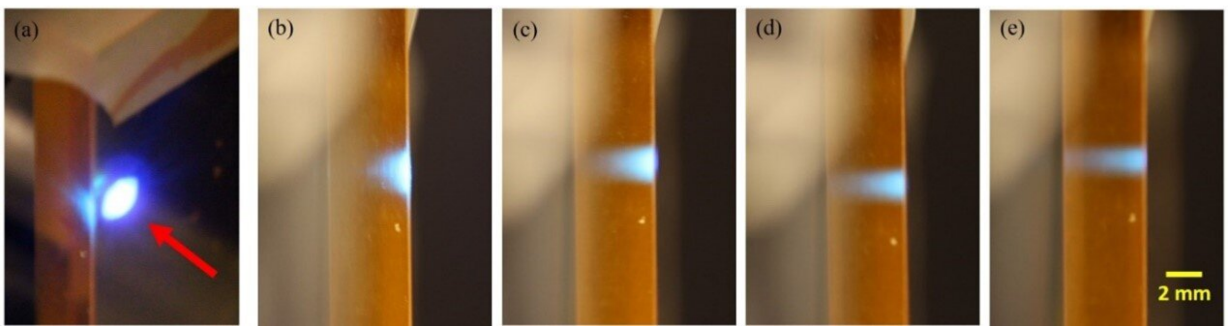

Fig. 1. Two-photon induced fluorescence in polycrystalline ZnSe while varying incident laser intensity at 775 nm (a) intense front face blue emission; (b) side view, I = 25 GW cm-2; (c) I = 4 GW cm-2; (d) I = 1.5 GW cm-2; (e) 1 GW cm-2. Fluorescence appears deeper in sample as incident intensity decreases. Credit: Compuscript Ltd

In a new publication from *Opto-Electronic Advances*, authors from the Laser Engineering Group in the School of Engineering at the University of Liverpool, Liverpool, United Kingdom, discuss two-photon absorption and stimulated emission in poly-crystalline zinc selenide with femtosecond laser excitation.

Zinc selenide (ZnSe) is a fascinating optical material, a semi-conductor which is transparent from the visible spectrum all the way to the far infra red where, for example it is used as windows in heat sensing thermal cameras. When doped, single crystal ZnSe is the basis of light emitting diodes. Its optical properties, however vary with incident light intensity—termed a non-linear response. This sensitivity has driven the current study where the exposure to 200 femtosecond (2×10^{-13} sec) laser pulses in the near infra red at 775 nm causes emission of intense, beautiful blue fluorescence—through a process known as 2-photon absorption where simultaneous absorption of two low energy photons at 775 nm exite electrons to higher levels, where, after a few nanoseconds, high energy blue fluorescent photons (460–500 nm) are emitted.

The material studied here is polycrystalline ZnSe—readily available, still very pure and much cheaper than single crystal material. The two-photon absorption coefficient (b) was also found to change with intensity and measured using a "Z-scan technique" where a thin sample of ZnSe is translated through a weakly focused laser beam while measuring the change in transmission. This variation in b also infers that further sequential photon absorption (or excited state absorption) takes place during laser exposure and termed reverse saturated absorption. At low peak intensities I^{-2}, we measured b = 3.5 cm GW-1 at 775 nm, in agreement with other research—reducing significantly as intensitiy is increased.

The intense blue fluorescence observed encouraged us to consider whether at ultrahigh intensity, stimulated emission could be induced in polycrystalline ZnSe by two-photon absorption at 775 nm. This has previously been observed in single crystal ZnSe. As the fluorescence lifetime was measured to be te ~ 3.3 ns, a thin 0.5 mm thick sample was mounted in a short (10 cm) optical cavity providing feedback. Stimulated emission was indeed confirmed by significant line narrowing from a bandwidth Dl = 11 nm (cavity blocked) to Dl = 2.8 nm at peak wavelength lp = 475 nm while the upper state lifetime also decreased. This is the first reported observation of stimulated emission in polycrystalline material. These results suggest that with more optimum pumping conditions and crystal cooling, polycrystalline ZnSe might reach lasing threshold via two-photon pumping at l = 775 nm.

The Laser Engineering Group in the School of Engineering at the University of Liverpool is headed by Professor Geoff Dearden, an expert in Lasers and Photonics. Over a number of years, ultrafast laser-materials interactions (using femtosecond and picosecond pulses) have been studied in detail using, for example laser ablation for production of periodic complex surface micro-structures (laser beam engineering on metals, polymers and semi-conductors. Such structures have applications in controlling surface hydrophobicity, anti-bacterial response, security marking and precision micro-structuring of high value components for sectors such as aerospace. With femtosecond pulses, transparent polymers (PMMA) and dielectrics such as Sapphire have been internally micro-structured at high speed through parallel beam, multi-photon absorption. The resulting periodic refractive index engineering can create hig- quality, high-efficiency volume Bragg gratings, useful in spectral analysis and creating high temperature sensors for extreme environments (aero-engines).

More information: Qianliang Li et al, Two-photon absorption and stimulated emission in poly-crystalline Zinc Selenide with femtosecond laser excitation, *Opto-Electronic Advances* (2022). DOI: 10.29026/oea.2022.210036

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