Generating ultra-violet lasers with near-infrared light through 'domino upconversion' of nanoparticles

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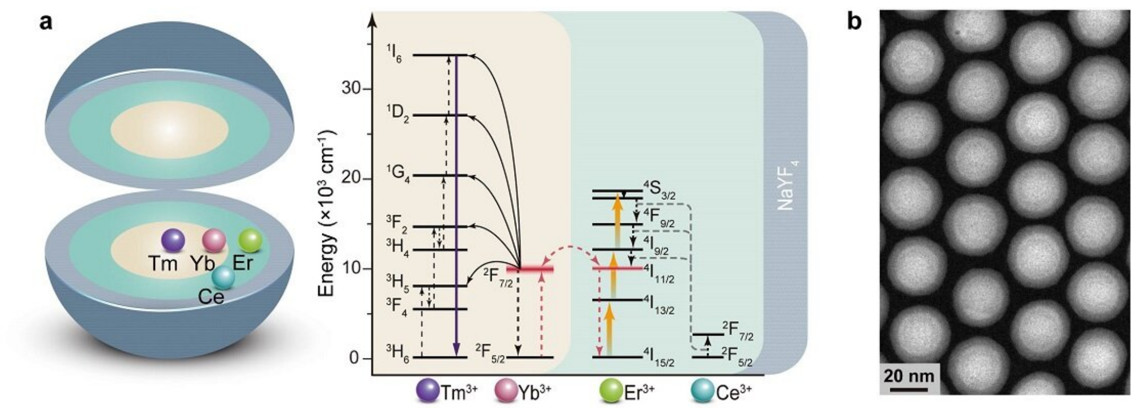

(a) Schematic design of a NaYF4:Yb/Tm@NaErF4:Ce@NaYF4 core–shell–shell nanoparticle for domino upconversion (left panel) and the proposed energy transfer mechanism in the nanoparticle. (b) A high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of the NaYF4:Yb/Tm@NaErF4:Ce@NaYF4 nanoparticles, highlighting the layered structure. Credit: City University of Hong Kong

Strong and coherent ultraviolet light emission devices have enormous medical and industrial application potential, but generating ultraviolet light emission in an effective way has been challenging. Recently, a collaborative research team co-led by researchers from City University of Hong Kong (CityU) developed a new approach to generate deep-ultraviolet lasing through a "domino upconversion" processing of nanoparticles using near-infrared light, which is commonly used in telecommunication devices. The findings provide a solution for constructing miniaturized high energy lasers for bio-detection and photonic devices.

In the world of nanomaterials, "photon upconversion" means that when nanomaterial is excited by light or photons with a long wavelength and low energy, it emits light with a shorter wavelength and higher energy, such as ultraviolet light.

Challenge in achieving photon upconversion

Photon upconversion characterized by high-energy emission upon excitation of lower-energy photons is of exceptional interest among scientists. This is because it holds potential for cost-effective construction of miniaturized deep-ultraviolet emission devices, which have enormous medical and industrial application potential, such as microbial sterilization and biomedical instrumentation. However, the photon upconversion process has limited flexibility, as it occurs mainly in special lanthanide ions comprising fixed sets of energy levels.

A research team co-led by Professor Wang Feng, from Department of Materials Science and Engineering, and Professor Chu Sai-tak, from Department of Physics at CityU, together with Dr. Jin Limin from the Harbin Institute of Technology (Shenzhen), overcame the obstacle by introducing a "domino upconversion" tactic.

Special structural design of nanoparticles

Domino upconversion is like a chain reaction, in which energy amassed in one upconversion course triggers another succeeding upconversion process. By using a doughnut-shaped microresonator, incorporated with specially designed "upconversion nanoparticles," the team successfully generated high-energy, deep-ultraviolet light emission at 290 nm by excitation of low-energy infrared photons at 1550nm.

"As the excitation wavelength was in the telecommunication wavelength range, the nanoparticles can be readily used and integrated into existing fiber-optic communication and photonic circuits without complicated modification or adaptation," said Professor Wang. The findings were published in the journal *Nature Communications*, titled "Ultralarge anti-Stokes lasing through tandem upconversion."

The idea of constructing "domino upconversion" was inspired by a previous study of energy transfer in core-shell nanoparticles by Professors Wang and Chu. The core-shell structure design of the nanoparticle allows the multiphoton luminescence process in erbium (Er^{3+}) ions. By adapting a similar synthetic protocol, the team successfully constructed "core-shell-shell" nanoparticles through a wet-chemistry method to explore the energy-transfer mechanism of lanthanide ions, including thulium (Tm^{3+}) ions.

Doughnut-shaped microresonator

Through the careful design of doping composition and concentration in different layers or shells of the upconversion nanoparticles, the team successfully achieved a tandem combination of Er^{3+} and Tm^{3+} ions-based upconversion processes (domino upconversion). In the experiment, the Er^{3+} ions contained in the outer shell responded to 1550 nm near-infrared photon excitation, a wavelength located in the telecommunication range. By incorporating the nanoparticles into a doughnut-shaped microresonator cavity, the team further generated a high-quality ultraviolet microlaser, demonstrating lasing action at 289 nm by 1550 nm excitation.

"The upconversion nanoparticles act as 'wavelength converters' to multiply the energy of incident infrared photons," explained Professor Wang. He expects the findings to pave the way for the construction of miniaturized short-wavelength lasers and says they may inspire new ideas for designing photonic circuits. He added that the miniaturized ultraviolet laser using this domino upconversion technology can provide a platform for sensitive bio-detection, such as the detection of cancer cell secretion, by monitoring the lasing intensity and threshold, which offers great biomedical application potential in the future.

More information: Tianying Sun et al, Ultralarge anti-Stokes lasing through tandem upconversion, *Nature Communications* (2022). DOI: 10.1038/s41467-022-28701-1

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