Towards extremely high-order optical nonlinearity at the nanoscale

Xiang Hao* and Cuifang Kuang*
Zhejiang University, College of Optical Science and Technology, State Key Laboratory of Modern Optical Instrumentation, Hangzhou, China

Nonlinear optics lie in the center of many optical technologies. As the superposition principle no longer holds, optical nonlinear response from materials frees many of the constraints in classical optics and has enabled a range of practical applications, from laser processing to quantum optics. Yet, irrespective of the advantages, the nonlinear response is typically observable at very high laser intensity. The photon avalanche (PA) effect is therefore exceptional, since it allows sufficiently weaker lasers for excitation. Unfortunately, PAs are mostly restricted to bulk materials and rely on cryogenic conditions,[1] hindering their wide application.

To resolve this dilemma, a group led by Prof. Qiuqiang Zhan from South China Normal University recently developed a strategy[2] to generate huge optical nonlinearities from various emitters localized in multilayer core/shell optical nanocrystals. This novel technique is rooted in the new concept of migrating photon avalanche (MPA) mechanism. More specifically, the avalanche looping cycles occur in the Yb^{3+}/Pr^{3+} co-doped core and it allows a 26th-order nonlinearity with a clear pumping threshold as low as 60 kW/cm^2 under continuous-wave (CW), near-infrared laser excitation. This dual-doping strategy successfully overcomes the concentration quenching in singly-doped system, which is mostly used in previous studies. Most importantly, these PA cores can serve as nano-engines, arousing huge nonlinearity from emitters distributed in spatially separated shells via the long-range sublattice energy migration. In combination with the upconversion process, the MPA mechanism can realize multiplied nonlinear luminescence. For example, by doping Tm^{3+} emitters into the outer shell layer, the authors further enhanced the optical nonlinear response order up to 46.

The most important advance of this work is that the proposed universal MPA is achievable at the nanoscale at room temperature. The established core-shell nanoparticles have a small diameter of 9.5 nm and a thickness of 3.5 nm, respectively. In addition, this strategy is universal with globally propagating optical nonlinear response, avoiding the sophisticated excitation schemes required in traditional PA mechanisms. These advantages render the authors' work an important step forward and expand the possibility for potential applications, including single-CW-beam driven diffraction-unlimited imaging/sensing, lithography, miniaturized laser and optical data storage.[3–5] For example, they exhibited ultralow-power, single-beam nanoscopy with a resolution of 62 nm ($\lambda/14$). Furthermore, they realized immunolabelling of subcellular filaments with PA nanoprobes and achieved nanoscopic bioimaging with multicellular field of view. Critically important for practical applications, the MPA enables fast PA kinetics with a very short response time (19.2 ms), whereas in traditional PA studies a rise time of seconds to minutes was required. Thereby, a 100 μs per pixel dwell

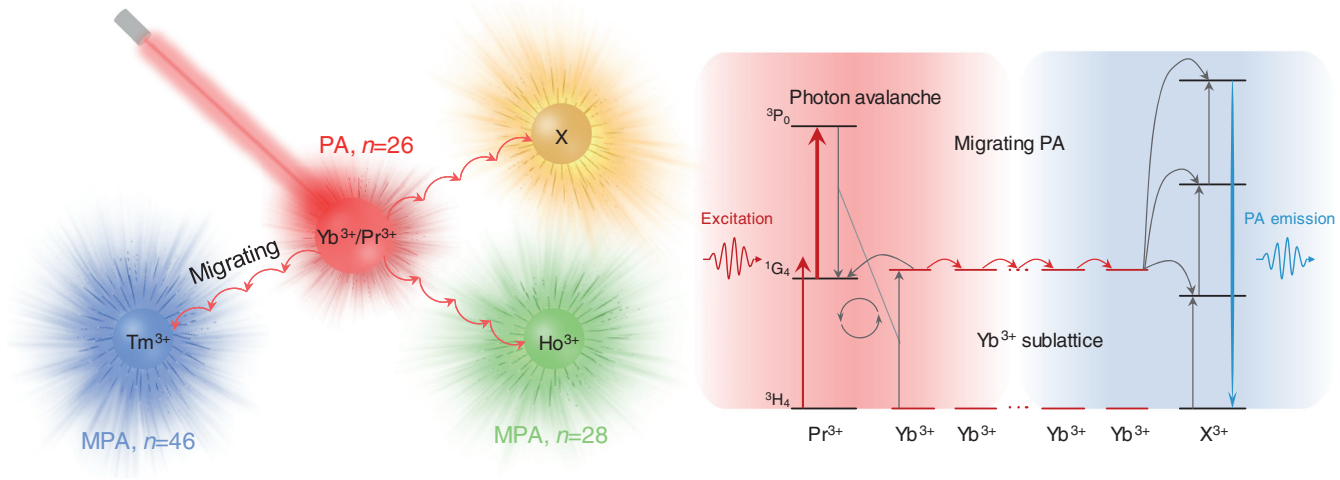

Fig. 1 Schematic illustration of the MPA mechanism with different emitters. For the nonlinear process, nonlinear order n is defined by $I_{em} \propto I_{ex}^{n}$, where I_{em} is the emission intensity and I_{ex} is the excitation intensity. The photon avalanche firstly occurs in Yb^{3+}/Pr^{3+} co-doped core and emissions from Yb^{3+} and Pr^{3+} all exhibit a nonlinear order of $n = 26$. The avalanching energy can migrate to the third ion (Tm^{3+}, Ho^{3+}, etc.) via Yb^{3+} sublattice, triggering multiplied nonlinear upconversion luminescence. Credit: Qiuqiang Zhan, South China Normal University.

*Address all correspondence to Xiang Hao, haox@zju.edu.cn; Cuifang Kuang, cfkuang@zju.edu.cn

time was realized in MPA nanoscopic imaging, which is compatible with a standard confocal/STED microscopic imaging setup.

The ultimate hope for this technique is that triggering optical nonlinearity at this unprecedented scale will yield unprecedented insights. This work represents significant progress towards extraordinary high-order optical nonlinearity at the nanoscale and its adaptation to an important application. However, as the proverb observes, Rome was not built in a day: while the high-order optically nonlinear response at the nanoscale proves possible, the full realization of its potential is still to come. The flexibility to tune the optical properties, in particular, the excitation and the emission spectra, will be a critical part of this process. Besides, for the bioimaging application demonstrated by the authors, their extension to imaging within living cells—capabilities that are developing rapidly—is another major concern. The naturally bleaching-free emitters presented in this work will be very promising for future live-cell imaging/tracking using nanoscopy and similar techniques (Fig. 1).

References

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