Surface functionalized microcavity boosts nonlinear optics

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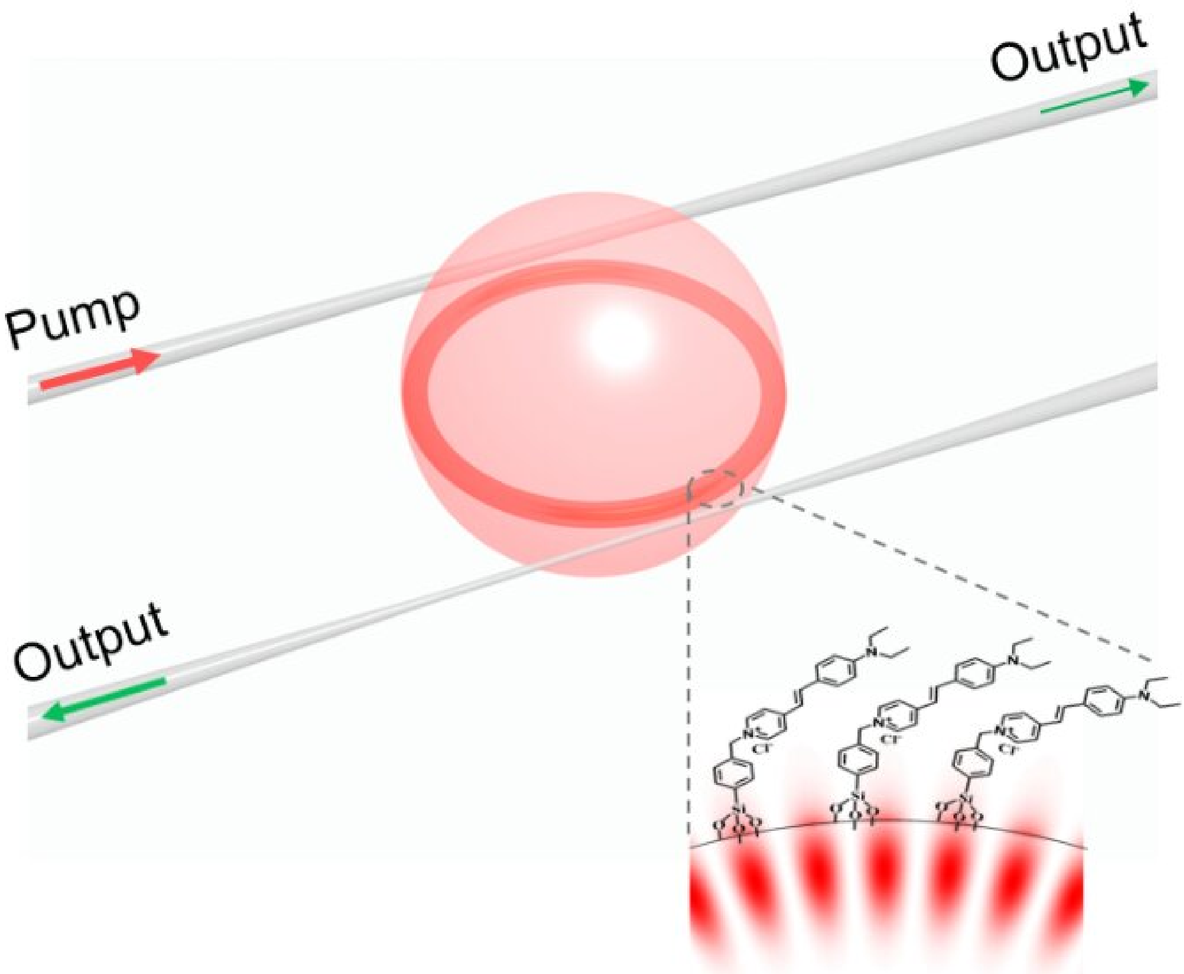

Figure 1: Schematic of the organically functionalized silica microcavity with dual fiber couplers. Credit: Peking University

Silica optical microcavities are mainstay photonic devices, valued for their intrinsically ultra-low loss in the broadband spectra and mature fabrication processes, but unfortunately, they suffer from low second- and third-order optical nonlinearity. A notable feature of the microcavity is the inherent leaking evanescent field at the surface, which opens the window for light-matter interactions at the surface.

Now, a research group led by Professor Yun-Feng Xiao at Peking University, in collaboration with Professor Xiaoqin Shen at Shanghai Tech University, has achieved record-high efficient third-harmonic generation (THG) in a surface functionalized silica microcavity. This work has been published online in *Physical Review Letters* titled "Microcavity nonlinear optics with an organically functionalized surface."

In this work, conjugated organic molecules are employed for the functionalization of the microcavity surface, which hold very large nonlinear optical response due to their large delocalized electron systems. Through a surface functionalization strategy, it is promising to bridge the high quality factor (Q) microcavities with the vast library of nonlinear molecules.

Given the geometry and material dispersion in a cavity, the optical frequency mismatch for pump light and third harmonic (TH) signal with their corresponding cavities modes can spoil the doubly resonant enhancement of TH output, especially in ultra-high-Q microcavities. "The surface enhanced third-order nonlinearity is one part of the story for efficient THG," said Jin-hui Chen, a "Boya" postdoctoral in the group of Professor Xiao. "We develop the dynamic-phase-matching method by leveraging the Kerr and thermal effects to tackle the challenging optical mode dispersion in ultra-high-Q microcavities."

These effects collaboratively introduce a frequency shift of the cavity modes, and lead to the dynamical compensation of both the pump and TH resonance mismatch. As a result, the bright TH signal is observed under a pump power of several milliwatt, with maximized conversion efficiency as high as 1,680 percent/W2, which is four orders of magnitude higher than that of the best reported pure silica microcavities. The ultra-high conversion efficiency is contributed by the strong nonlinearity of organic molecules and the ultra-high-Q-resonant enhancement of both pump light and TH signal.

To further identify the origins of the nonlinear signals, the researchers analyzed the pump polarization dependent TH or third-order sum frequency (TSF) output. They found that the output TH or TSF power with a transverse-electric pump polarization is about two orders of magnitude higher than that with a transverse-magnetic pump polarization due to the surface alignment of organic molecules.

"The experiment achieves the highest record of THG efficiency in silica photonics," said Professor Xiao. "Even more important, the work may open up the new horizon to improve properties and expand applications of microcavities, which is made of conventional bulk materials, such as silica and silicon nitride. The technology and the mechanism we learned and developed in this work, including the surface functionalization and dynamical-phase-matching method, will perform as the foundation for various applications, especially in broadband tunable nonlinear photonics."

More information: Jin-hui Chen et al. Microcavity Nonlinear Optics with an Organically Functionalized Surface, *Physical Review Letters* (2019). DOI: 10.1103/PhysRevLett.123.173902

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