Physicists develop miniature Raman laser sensors for single nanoparticle detection

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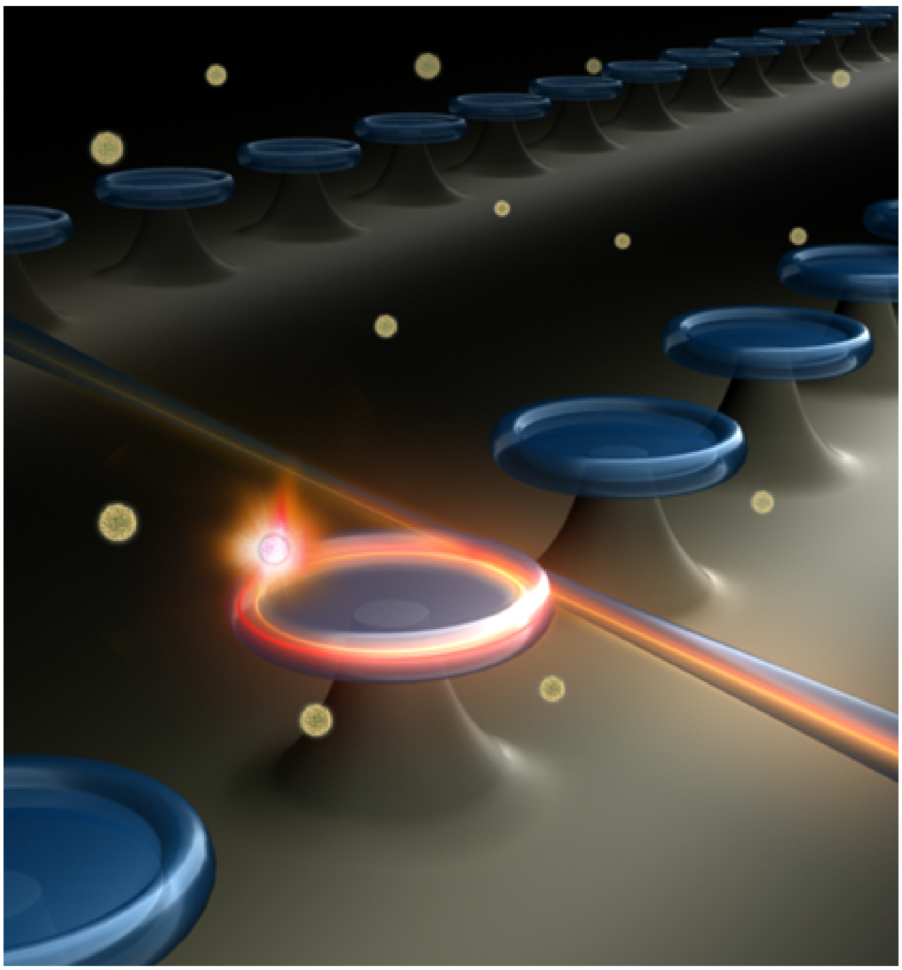

An array of microcavity Raman lasers for single nanoparticle detection. The pump laser light (in orange) is coupled into the microcavity via a fiber taper to generate the Raman laser (in red). When the Raman laser encounters a nanoparticle, it splits into two lasing lines due to backscattering, thus producing a beat note in the temporal Raman power. By measuring the beat frequency changes, real-time single nanoparticle detection can be performed.

Optical sensing of nanoscale objects with ultrahigh sensitivity is highly desirable for applications in various fields, such as in early-stage diagnosis of human diseases and in environmental monitoring, as well as in homeland security. The ultimate sensitivity of a nanoparticle sensor is expected to be at the level of a single nanoparticle response. Very recently, a group led by Professors Xiao Yun-Feng and Gong Qihuang from Peking University, has developed a novel microcavity Raman-laser sensor, and realized single nanoparticle detection in both air and in an aqueous environment. The smallest nanoparticle they detected in water was 20 nanometers in radius. Their new experimental results were published in *Proceedings of National Academy of Sciences* online Early Edition.

The microcavities they use are called whispering gallery mode (WGM) optical microcavities, which are analogous to the acoustic whispering gallery in St. Paul Cathedral in London and the echo wall in the Temple of Heaven in Beijing. Due to the low propagation loss and small size of such microcavities, photons can circulate inside for up to hundreds of thousands of times, thus dramatically increasing light-matter interactions. Therefore, WGM microcavities are ideal platforms for label-free, ultrasensitive detection of single nano-objects.

In this work, researchers detected nanoparticles using stimulated Raman scattering (also called Raman lasing) in the microcavities. Raman scattering is an inelastic light scattering process, whereby the energy of an incident photonis modified by an inelastic interaction with the cavity material. The cavity material changes its energy state and the photon loses an equivalent amount of energy. The scattered light has a lower frequency and is called the Stokes emission. The energy difference between the incoming photon and the scattered Stokes photon matches a molecular vibration in the cavity material.

Stimulated Raman scattering occurs in both counter-propagating traveling cavity modes (i.e., clockwise and counterclockwise modes) which are initially degenerate. When the Raman laser encounters a nanoscale object, it will split into two Raman lines, because the nano-object induces a coupling between the two counter-propagating modes through backscattering. When both split modes lase simultaneously, a beat note is produced, with the beat frequency corresponding to the Raman mode splitting. Thus, by monitoring the beat frequency of the split-mode Raman lasers, ultrasensitive nanoparticle detection can be realized.

"This mode splitting sensing mechanism is robust to many noise sources, such as thermal noise and laser frequency noise, since the two split modes share exactly the same noise," said Professor Xiao. "Moreover, Raman laser has much narrower line width compared to a passive microcavity, and thus to significantly lower the detection limit."

"In order to demonstrate the feasibility of this sensing mechanism, we first did a control experiment in air. We transferred single nanoparticles to and from the microcavity using a nanofiber, using a technology previously developed by our group, and measured the beat frequency changes after each transfer process," said Li Bei-Bei, a Ph.D. student in Professor Xiao's group. "As expected, every time a nanoparticle is transferred to the cavity surface, the beat frequency of the Raman laser experienced an abrupt change." Using this method, real-time single nanoparticle detection in an aqueous environment was then performed, with the smallest particle of 20 nanometers in radius detected.

"Another advantage of this sensing mechanism is that Raman gain is an inherent property of almost any material, so we do not need to dope the cavity with an additional gain medium. Furthermore, since Raman scattering occurs under practically any pump wavelength, this sensing method also loosens the requirement of specific wavelength bands for the pump laser," said Xiao. "Due to these advantages, this Raman-laser sensor represents a significant step towards practical microlaser sensors."

More information: Bei-Bei Li, William R. Clements, Xiao-Chong Yu, Kebin Shi, Qihuang Gong, and Yun-Feng Xiao. "Single nanoparticle detection using split-mode microcavity Raman lasers." *PNAS* 2014 ; published ahead of print September 29, 2014, DOI: 10.1073/pnas.1408453111

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