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Revealing low-radiative modes of nanoresonators with internal raman scattering

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The resonantly enhanced optical interaction of light with metallic, dielectric, or semiconductor nanoparticles, which are the basic components of various nanooptical devices, is at the forefront of modern nanophotonics [1–4]. The examples of vital applications include on-chip integrated photonic systems [5, 6], lab-on-chip technologies [7], medical biosensing [8] and others. However, understanding the modal content of resonant nanostructures is non-trivial problem due to their substantially subwavelength sizes and typically complicated geometries. This problem has very limited solution within the common optical spectroscopy methods as some of the modes can be low-radiative because of their weak coupling to freely propagating waves, and in this sense are often known as *dark-modes* [9–11]. Thus, studying the modal spectrum of optical nanostructures with account on dark modes immediately becomes a complicated problem.

The solution of this problem usually relies on sophisticated optical methods such as total internal reflectance microscopy (TIRM) [12] and its combination with atomic-force microscopy [13, 14]. The methods based on resonant mode excitation through accelerated electron scattering showed their efficiency in observing the dark-modes of nanostructures.

In this Letter, we suggest detecting an intrinsic Raman signal, which corresponds to inelastic photon scattering via optical phonons excitation in a crystalline lattice of a nanostructure to determine its electromagnetic modal structure [15]. The Raman signal generated by lattice vibrations is distributed over the volume of the nanoresonator and effectively couple to all resonant modes including dark ones. By exciting the optical phonons and their consequent spontaneous decay, the incoherent Stokes signal is generated inside the nanoresonator under study. The intensity of the Raman signal is governed by the optical density of states [16, 17] inside the nanoresonator at the frequency of the Raman emission, and additionally enhanced at the excitation frequency by resonant excitation of the eigenmodes. We reveal magnetic quadrupole mode in a rather complicated system of a silicon nanoparticle arranged on a gold substrate as well as an anapole state, demonstrating the applicability of our approach even when dark-field optical spectroscopy methods are not efficient. Silicon as one of the main materials used in all-dielectric nanophotonics [1, 18, 3] has a well pronounced peak in Raman spectrum at $\Omega \approx 520\,\mathrm{cm}^{-1}$, which is defined by the light coupling with optical phonons at the band central point [19–21]. The system under study consists of a subwavelength silicon nanoparticle (SiNP) placed over a gold substrate as show in Fig. 1. It should be stressed that approach can be applied for detecting any resonant mode excited in any structures that have inherent Raman scattering at visible range.

In conclusion, we have suggested an efficient approach for experimental determining the spectral content of nanoresonator basing on utilizing an incoherent Raman scattering, inherent to the resonator material. The approach was confirmed theoretically and experimentally for silicon nanoparticle resonators possessing a strong intrinsic Raman response. We have shown that the suggested method allows us to reveal the magnetic quadrupole resonance combining with the anapole state in the system of a silicon nanoparticle arranged on a gold substrate, demonstrating the applicability of our

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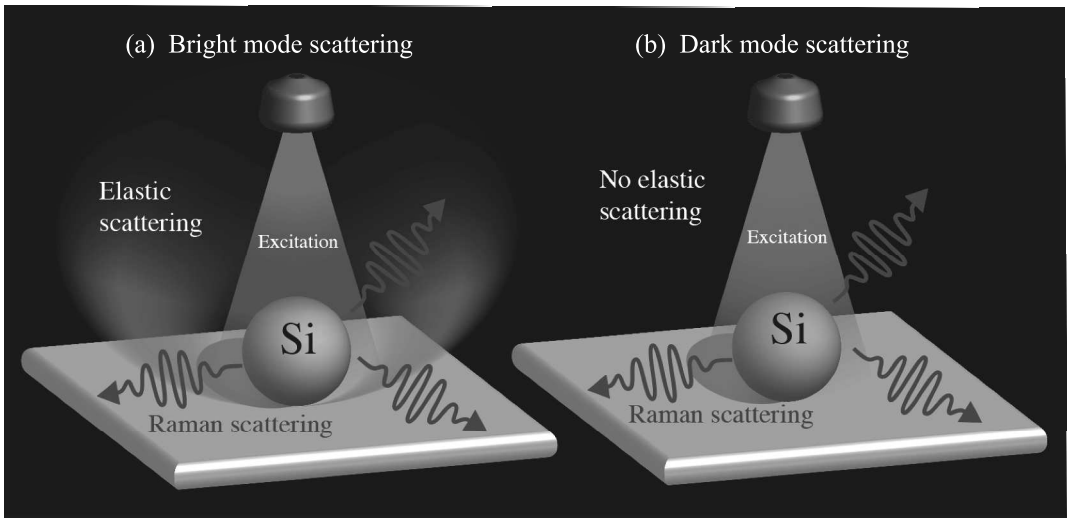

Fig. 1. (Color online) The elastic and inelastic scattering by bright and dark resonant modes in silicon nanoparticle on the top of gold substrate. (a) – The excitation of the bright nanoparticle mode results in both elastic scattering and inelastic (Raman) scattering. (b) – The excitation of the dark mode does not contribute to elastic scattering, but can be detected via Raman scattering signal

approach for broad range of non-plasmonic resonant nanostructures.

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