Particle sizing by mode splitting

It has long been known that the optical resonances of ultrahigh-Q whispering gallery mode resonators can split under the influence of particle scattering. Now scientists have exploited this splitting to accurately determine particle sizes.

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Optical microcavities are dielectric structures that can confine light for an extended amount of time\(^1\). Dielectric microresonators have attracted significant interest since 1989, when it was discovered that they can possess whispering gallery modes (WGMs) with long photon lifetimes\(^2\) (that is, ultrahigh \(Q\)). Such ultrahigh-\(Q\) WGMs travel around the perimeter of a circular microresonator (such as a microsphere or microtoroid) and derive their name from the acoustic waves that propagate in a similar fashion around the dome of St. Paul’s Cathedral in London. Diverse applications of ultrahigh-\(Q\) WGM resonators have emerged over the past two decades, such as for nonlinear optics at exceedingly low light levels (nanowatts), cavity quantum electrodynamics, frequency metrology, biological sensing and optomechanics. One intriguing phenomenon of WGMs is that they can split and appear as doublets. Now in Nature Photonics, Jiangang Zhu et al. explain how they have exploited this splitting to accurately measure the size of particles\(^3\).

The principle can be explained as follows. The WGM resonator has clockwise and counter-clockwise modes, whose frequencies are degenerate (that is, identical). A nanoscale surface defect such as a small imperfection or deposited particle will scatter light from one of the cavity modes (clockwise, for example) into free space, and also scatter a fraction back in the opposite direction (counter-clockwise).

In optical microresonators with ultrahigh \(Q\), this effect has a surprising manifestation: the modes of the resonator can split into a clearly visible doublet, and this induced splitting can significantly exceed the cavity decay rate. Owing to the small resonator mode volume and ultrahigh-\(Q\) microcavity resonance, extremely small defects can induce a significant splitting. The appearance of mode splitting implies that the scattering is highly directional because the splitting frequency directly gives the rate at which photons are scattered from the clockwise to counter-clockwise direction.

The width of the resonance gives the decay rate — the rate at which photons are dissipated and scattered out of the resonator. This phenomenon was first observed in the late 1990s\(^4\). It can be used to create highly efficient narrowband reflectors\(^5\) that can be used for laser stabilization techniques. Most of the scattered power is fed back into the original cavity modes, rather than being lost through dissipation\(^6\). More recent work has shown that this splitting can be artificially produced by, for example, introducing a subwavelength near-field tip into the cavity’s near-field\(^7\) or by embedding small silicon nanocrystals\(^8\) in the cavity. The phenomenon can also be understood as arising from the Purcell effect — the high density of states in the cavity causes the particle to scatter more power compared with the free-space value, leading to preferential emission into the cavity\(^9\) (an effect that had traditionally only been associated with atomic emission).

So far, mode splitting has largely been of interest from a purely fundamental point of view. Now, Zhu et al. have gone one step further and demonstrated that mode splitting can have an interesting practical application — a surprisingly accurate method of measuring particle size\(^1\). In their experiments, Zhu and colleagues start with an on-chip toroidal resonator\(^9\) that does not contain any splitting and has ultranarrow resonances with \(Q\) factors exceeding 50 million. Light is coupled via a tapered optical fibre into the resonator...
so that it propagates in only one direction, either clockwise or counter-clockwise. Small dielectric particles of different sizes are sprayed onto the microresonator using a differential mobility analyser and a nozzle. The particles have varying diameters of 50–170 nm, with their presence and size verified by a scanning-electron microscope.

Deposition of these nanoparticulates leads to the two aforementioned effects. First, the presence of scattering from a particle couples the clockwise mode to the counter-clockwise mode. If the coupling rate is larger than the cavity decay rate, the amplitude of the clockwise and counter-clockwise modes are equal, forming two normal modes with a sine and cosine distribution — easily visible in the spectrum as a doublet (Fig. 1b). The splitting frequency is given by the rate of scattering,

\[
g = \frac{\alpha f(r)^2 \omega}{2V_m}
\]

where \(V_m\) is the resonator mode volume, \(\omega\) is the optical frequency and \(f(r)^2\) describes the overlap of the nanoparticle with the optical mode. The particle size is described by the polarizability, \(\alpha\). Evidently, a small resonator mode volume leads to large splitting.

However, determining the particle size from the splitting frequency is complicated by the fact that the splitting frequency depends on the location of the particle with respect to the transverse optical mode profile, as expressed by \(f(r)^2\). The shift is small where the optical mode profile has low field intensity, and is large where the field has high field intensity.

To overcome this limitation, the authors use an ingenious trick (first proposed in ref. 7): they also monitor the change in the cavity dissipation rate, \(\Gamma\). The presence of the scattering particle not only causes a mode coupling (splitting), but also causes enhanced dissipation because it couples the cavity modes to free space out of the resonator through Rayleigh scattering. This scattering also depends — in the same manner as the splitting frequency — on the overlap of the nanoparticle with the optical mode:

\[
\Gamma = \frac{\alpha^2 f(r)^2 \omega}{6ncV_m}
\]

Thus, by taking the ratio of the splitting frequency to the induced loss rate, the involvement of the overlap \(f(r)^2\) can be eliminated, allowing the dielectric polarizability \(\alpha\) and size of the particle to be determined, if the refractive index is known. Although mode splitting is not a new technique\(^1\), this work elegantly validates the concept for the first time. Indeed, one of the most striking features of the experiment by Zhu and colleagues is the remarkably precise determination of particle size — the scheme claims an error of less than 10% for particle diameters of 50–170 nm. This is surprising given that one would intuitively expect deviations to occur easily due to variations in the optical mode volume, and also given that the theory only applies to particles much smaller than the wavelength of light\(^1\).

The idea of using splitting as a size discriminator is elegant and has several advantages. First, this method is ‘differential’ and therefore does not, for example, depend on the microresonator temperature. This is a significant advantage as the modes of silica WGM resonators are very temperature sensitive; a change of only 1 °C changes the optical resonance by about 1 GHz at room temperature. Furthermore, this method is also largely independent of the exact location where the particles bind.

Among the biggest obstacles for future applications is that a priori information of the refractive index of the particles is needed for this particle sizing technique. This is analogous to techniques that measure the refractive index of coatings, which can never determine coating thickness and refractive index independently using a single wavelength of light. Nevertheless, this proof-of-concept experiment shows that mode splitting, which so far has always been considered a phenomenon of only fundamental interest, can be exploited as a surprisingly accurate method of measuring particle size.

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References

SILICON PHOTONICS

On-chip OPOs

Optical parametric oscillators (OPOs) have now been realized in a CMOS-style process by exploiting nonlinear four-wave mixing. Such multiwavelength sources bring the prospect of ultrafast chip-to-chip optical data communications a step closer.

Jeremy Witzens, Thomas Baehr-Jones and Michael Hochberg

The potential for incorporating photonic functionality in silicon very-large-scale integrated (VLSI) circuits is extremely exciting\(^1\), particularly for applications such as high-speed chip-to-chip data communication, spectroscopy and sensing. As a result, silicon photonics has attracted great attention from both academia and industry in recent years. However, there remain significant scientific and engineering challenges that must be tackled before the full potential of this integration can be achieved. One challenge in particular is the development of appropriate optical sources. Many of the expected advantages of silicon as a platform for integrated photonics emerge from the ability to build large complex optoelectronic systems with high yields and integrated control electronics. Constructing such systems, however, often requires chip-integrated coherent optical sources of multiple wavelengths.

The problem is that making lasers in silicon is not easy. Although both wafer- and die-bonded sources made from InP materials have been demonstrated, they create significant challenges in terms of