Squeezing light for single-molecule spectroscopy

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By using optical nanocavity resonators, the interaction between the light and target molecules is enhanced more than a million times, leading to improved single-molecule detection.

Label-free, single-molecule detection is an exciting avenue of study that will someday impact many areas of research, including drug discovery, biosensing, and early stage cancer detection. In order to develop such a technique, a device must first be fabricated that has a high sensitivity and specificity as well as the ability to assay small-volume samples. Optical spectroscopy is a very powerful technique with many properties that make it attractive for application in single-molecule detection, including its high selectivity, remote sensing capabilities, and fast detection time. Furthermore, many molecules of interest, including most proteins, have unique absorption features in the mid- to long-wave IR range. Unfortunately, they also have an extremely weak interaction with the light due to their negligible size compared with the broad IR wavelength. The traditional solution to this problem has been to place the molecules in an optical cavity where light will be reflected multiple times, allowing frequent interactions between the light and the target. In fact, recent progress in ultra-high quality (Q)-factor cavities has led to accurate label-free, single-molecule detection.

Unfortunately, an ultra-high Q factor leads to an extremely narrow linewidth. As a result, these sensors are quite sensitive to environmental variables such as temperature fluctuations. Also, it is not possible to have sufficient molecular specificity without the use of functionalized surfaces. This is because the narrow linewidth does not include enough of the unique molecular spectral fingerprint to accurately distinguish one molecule from another: see Figure 1 (top).

We were interested in finding a method that would provide a strong interaction between the light and target molecule while maintaining a broad linewidth. The figure of merit that shows the interaction strength between light and the electronic states of materials inside a cavity is called Purcell’s constant: \[ p = \frac{3}{4\pi^2} \frac{Q}{V} \lambda^3, \]
where \( V \) is the volume of the optical mode and \( \lambda \) is the wavelength of light. Traditionally, the mode volume is not much smaller than the wavelength cubed, but our team decided the only way to have a large Purcell factor and a wide bandwidth was to radically reduce the mode volume. By taking advantage of the light-squeezing properties of a photonic crystal (PC) microcavity resonator and a metal-dielectric-metal sandwich, we were able reach this goal. The result is a plasmon-polariton crystal (PPC) nanocavity resonator: see Figure 1 (bottom left).

Detailed finite-domain time-difference simulations (see Figure 1, bottom right) show that a PPC crystal with a cavity...
defect of 40nm (radius) has a modal volume of ~20 zeptoliters at \( \lambda = 3.45\mu m \) and a Q-factor of 23.3, which is many orders of magnitude smaller than the best PC Q-factors\(^3\) of \(10^6\)–\(10^7\). This small Q-factor gives an extremely broad linewidth of about 7.4THz (as opposed to the narrow linewidth of tens of MHz for the PCs). However, because of the ultra-small volume, the PPC has a Purcell factor (\(p\)) of \(\sim 10^6\) compared to \(\sim 10^5\) for PCs.

Moving toward application of this technology, our team has also fabricated our first PPC devices using plasma enhanced chemical vapor deposition and a focused ion beam (see Figure 2). We plan to couple light to the cavity using the extremely weak coupling of normal incident light and the in-plane cavity mode. The broadband sensing properties of these PPC devices should provide enough of the spectral fingerprint to discriminate between unique molecules within a given sample.

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Ryan Gelfand is a second-year PhD student, and his research interest involves novel plasmonic devices.

References