Plasmonically enhanced optical sensors

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Exploiting optical modes generated at the surface of some metals or nanoparticles can enhance absorption, fluorescence, and Raman spectroscopy for sensing applications.

Optical sensors are ubiquitous in every walk of life, and find increasing applications in medicine, atmospheric monitoring, process control, and defense. Most organic and inorganic substances have distinct spectral features in their absorption, luminescence, or Raman characteristics, and using optical methods is the most direct way to detect and identify these. However, the detection process becomes challenging when the target substance is present in minute quantities, such as parts per billion, and the optical signal becomes too weak to detect in the presence of noise. There are two factors limiting the strength of these optical signals. First, only a small fraction of the optical pump energy gets captured by the molecules that we want to detect. Second, only a small proportion of the captured signal is emitted, due to intrinsic inefficiencies in the molecules’ optical processes.

We can remedy the first problem of insufficient absorption by concentrating the incoming radiation into a small volume around the molecules, using (for example) micro-resonators. Also, it is possible to enhance the efficiency of emission—as realized by Purcell in 1946—by combining an optical mode (or any other electromagnetic mode) within a small volume (the ’Purcell effect’). Regrettably, the degree to which we can squeeze the light into a small volume is restricted by one of the more fundamental laws of optics: diffraction limit. This states that the minimum volume of light confinement in a dielectric medium with refractive index $n$ is on the scale of $(\lambda/n)^3$, where $\lambda$ is the wavelength.

However, we can circumvent diffraction limit by introducing a metal with a negative dielectric constant (and hence an imaginary refractive index) in addition to a dielectric with a positive constant. Consequently, a new type of optical mode—the surface plasmon polariton (SPP)—appears at the metal-dielectric interface, or around metal nanoparticles. In a conventional optical mode, the energy oscillates between the electric and magnetic fields; however, in SPP it oscillates between the electric field and the current of free electrons in the metal, while the magnetic field is very weak. As a result, SPP can be confined to sub-wavelength volume. If this volume happens to contain some molecules of interest, their efficiency of absorption and emission increases substantially. This is the principle of plasmonic enhancement of optical processes, illustrated in Figure 1 for fluorescence sensing, one of the most widely used optical sensing techniques.

The pump radiation at wavelength $\lambda_{pump}$ first couples with the sub-wavelength SPP mode, which in Figure 1 is in the gap of the metal nanoantenna. There, the molecule efficiently absorbs and re-emits the radiation into the SPP mode at a longer (or Stokes-shifted) wavelength $\lambda_{stokes}$, and the Purcell effect further enhances the efficiency of emission. From the SPP mode, the Stokes-shifted light couples into the propagating wave, enabling its detection. There have been numerous experiments performed with fluorescence sensing, but the most spectacular enhancement is achieved with Raman scattering. Here, the processes of absorption and re-emission by the molecule occur simultaneously, and the amount of Stokes Raman shift serves to identify and distinguish different molecules by their vibrational

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spectra. This phenomenon—surface enhanced Raman scattering (SERS)—originated in the 1970s, and thus predates the whole field of plasmonics. Currently, it is the only widespread commercial application of SPPs.

Nonetheless, there remain limitations to plasmonic enhancement. In SPPs, the energy is constantly transferred to and from the free electrons in the metal. The energy is lost as the scattering processes take place in the metal at a rate as high as $10^{14}$ s$^{-1}$, even in metals with the lowest loss, such as silver. Furthermore, the efficiency of radiation coupling out of SPP mode can be low. Hence, most of the energy emitted by the molecule into SPP mode may end up dissipated in the metal: a process referred to as quenching of fluorescence. Obviously, if the original efficiency of fluorescence by the given molecule is reasonably high, we may end up worse off using SPPs than not. However, for the very low efficiency emitter, obtaining even the few photons that escape SPP mode is better than none. We can think of Raman scattering as fluorescence with efficiency approaching zero, hence the Raman process can always be enhanced, and we observe no quenching in SERS experiments.

Careful design of plasmonic structures—using dimers, for example—can alleviate some of the quenching. However, overall, plasmonic enhancement works well only for low-efficiency optical processes. Where efficiency is paramount, such as in solar energy cells, radiation detectors, light emitting diodes, and lasers, plasmonically enhanced devices remain elusive, despite significant effort to develop them. At present, the most most viable applications of plasmonics\textsuperscript{4} are where absolute efficiency is only a secondary concern, such as in plasmonically enhanced sensors. Our future work will focus on optimizing plasmonic structures to further improve the performance of these sensors.

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References