Near-Field Optical Spectroscopy Based on the Field Enhancement at Laser Illuminated Metal Tips

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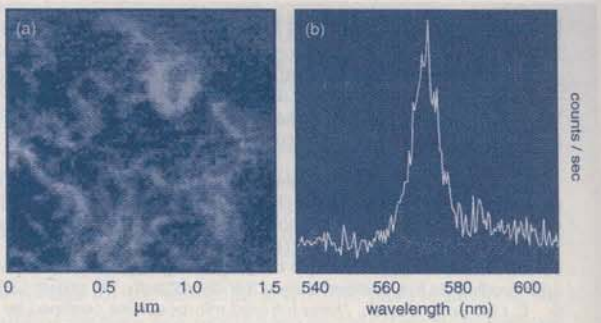
Modern science of condensed matter relies strongly on spectroscopy and microscopy. Optical spectroscopy is of particular importance because the light-matter interaction provides a wealth of information on structure and dynamics. Combining optical spectroscopy with microcopy is especially desirable because the spectral features can be spatially resolved. Unfortunately, the diffraction limit has been preventing researchers from resolving features smaller than half a wavelength of the applied radiation.

In recent years a novel microscopy, near-field optical microscopy,[1,2] has extended the range of optical measurements beyond the diffraction limit and stimulated interest in many disciplines, especially material sciences and biological sciences. In the most widely adapted aperture approach,[3] light is sent down an aluminum-coated fiber tip of which the foremost end is left uncoated to form a small aperture. Unfortunately, only a tiny fraction of the light coupled into the fiber is emitted through the aperture because of the cut-off of propagation of the waveguide modes. The low light throughput and the finite skin depth of the metal are the limiting factors for resolution. Many applications require higher spatial resolution than that obtainable with the aperture technique.

To overcome this limitation, a new apertureless scheme has been introduced.[4] It makes use of the strongly enhanced electric field close to a sharply pointed metal tip under laser illumination. The energy density close to the metal tip is shown to be three to four orders of magnitude larger than the energy density of the illuminating laser light. The tip is held a few nanometers above the sample surface so that a highly localized interaction between the enhanced field and the sample is achieved. To obtain a high image contrast, we take advantage of nonlinear optical interactions based on multi-photon processes. Recent experiments performed at the Pacific Northwest National Laboratory in Richland, WA demonstrated resolutions on the order of 20nm by making use of the field enhancement effect combined with two-photon excitation of the sample.[5,6] In these experiments, the sample was locally excited by the simultaneous absorption of two photons. The subsequent fluorescent emission is detected and used to construct an optical image of high resolution by raster scanning the sample.

Figure (a) shows the near-field fluorescence image of J-aggregates of PIC dye molecules in polyvinyl sulfate (PVS). The optical resolution in this image is limited by the actual width of the J-aggregate strands (30nm). Figure (b) is the spectrum of the emitted fluorescence when the tip was positioned over a particular J-aggregate strand at a fixed location. The spectrum disappeared when the tip was retracted from the sample surface, proving that the spectrum indeed originates from the near-field excitation of the tip.

The new method holds promise for gaining deeper insight into the photophysics of nanometric structures such as quantum dots or single molecules embedded in complex environments. For example, we are optimistic that the new method will make it possible to optically resolve individual protein molecules in biological membranes.



Novotny Figure 1. Figure **(a)** shows the near-field fluorescence image of J-aggregates of PIC dye molecules in polyvinyl sulfate (PVS). The optical resolution in this image is limited by the actual width of the J-aggregate strands (30nm). Figure **(b)** is the spectrum of the emitted fluorescence when the tip was positioned over a particular J-aggregate strand at a fixed location. The spectrum disappeared when the tip was retracted from the sample surface, proving that the spectrum indeed originates from the near-field excitation of the tip.

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Nonlinear Optics

Transient Coupling of Electromagnetic Radiation To Surface Plasmons in Solid-State Structures With Time-Varying Plasma Density

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Surface electromagnetic waves propagate along the interface between two media and exponentially decay with distance from the interface. The simplest interface that can guide surface waves is the boundary of a plasma or plasma-like material such as metal or a semiconductor. In this case, the surface waves are called surface plasmon polaritons or, simply, surface plasmons. The strong localization of surface plasmons makes them useful for waveguiding since in the absence of losses in the guiding medium, the surface plasmons can propagate without attenuation. But this also poses a problem: bulk electromagnetic radiation cannot launch surface plasmons, and neither can surface plasmons decay into radiation from the boundary. This follows from the fact that it is impossible to simultaneously satisfy two conservation laws: those of in-plane photon momentum (or wavevector) and of photon energy (or frequency).

To couple bulk and surface waves, the techniques most commonly used are to break the translational symmetry of the boundary using rough (or grated) surfaces[1] or to synchronize the waves spatially using the method of attenuated total reflection. In both cases the coupled bulk and surface waves have the same frequency. Clearly, breaking the time symmetry by using a medium with time-varying properties should also provide the coupling between surface and bulk waves. Recently we proposed using a rapidly created plasma in semiconductors to excite surface plasmons by incident bulk radiation.[2]