HIGH-RESOLUTION SPECTROSCOPY OF SINGLE MOLECULES IN SOLIDS

W. E. MOERNER, *Department of Chemistry and Biochemistry, University of California at San Diego, La Jolla, CA 92093-0340.*

A single impurity molecule in a solid is an exquisitely sensitive probe of the immediate local environment of the molecule (the "nanoenvironment"). Over the past few years, precision optical spectroscopy of the electronic transitions of individual, single impurity molecules in crystals and polymers has become an expanding field of study. Experimenters have observed a variety of fascinating physical effects, for example, the shifts in resonance frequency of a single molecule arising from nearby two-level-system transitions in the solid (spectral "diffusion"), the resonance Raman spectrum of a single molecule, quantum optical effects such as photon antibunching, and magnetic resonance of a single molecular spin (see *Science* 265, 46 (1994)).
The time-dependent resonance frequency of a single molecule (the spectral trajectory) provides information on dynamics of the nearby matrix which is not obscured by the usual need for ensemble averaging. Having such detailed dynamical trajectories of the spectral diffusion effect has stimulated theorists to produce a microscopic stochastic model for the local configurational degrees of freedom. In recent work, study of the spectral shifting behavior of single molecules has been extended to Shpol'skii matrices (frozen alkanes) in which a two-state or few-state behavior has been observed. This behavior can be followed with a fluorescence microscope in real time at video frame rates. Motivated by the need for higher signal-to-noise ratios and increased spatial resolution, we have used a low-temperature near-field optical source of subwavelength dimensions to record excitation spectra of single molecules located below the surface of a solid sample [a].

[a].W. E. Moerner, T. Plakhotnik, T. Irngartinger, U. P. Wild, D. W. Pohl, and B. Hecht, *Phys. Rev. Lett.* 73, 2764 (1994)