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Variance Spectroscopy: A New Bridge between Ensemble and Single-Particle Studies

R. BRUCE WEISMAN, Rice University

We have developed a new experimental technique that probes variations in the spectra from small regions of heterogeneous bulk samples resulting from statistical variations in composition. The method is demonstrated using suspensions of single-walled carbon nanotubes (SWCNTs), which contain mixtures of distinct structural species emitting photoluminescence at characteristic short-wave infrared wavelengths. Using dilute SWCNT suspensions, focused excitation beams, multichannel detection, and quick data collection, we capture several thousand emission spectra representing different spatial regions of the sample. The data sets are analyzed to find emission mean and variance values as a function of wavelength. The combined mean and variance spectra contain information unavailable from conventional methods, including the abundances of different emissive species and their relative emission efficiencies. The variance data are also analyzed for correlations between intensity fluctuations at different wavelengths to give novel two-dimensional maps that reveal the spectra of homogeneous sub-populations within heterogeneously broadened bulk spectra. The off-diagonal features in these maps expose spatially correlated concentration variations for nanotubes of different types, which arise from earliest stages of aggregation. Variance spectroscopy should prove a powerful new experimental tool for characterizing nanoparticle samples.