A Single Molecule Approach to Defect Studies in ZnO


— Single molecule investigations are a powerful tool for understanding molecular systems with inhomogeneous behavior that is either broadened or completely washed out of ensemble measurements. Here we apply single molecule microscopy methods to defects in ZnO. In addition to its status as an emerging optoelectronic material, ZnO hosts point defects which may have useful quantum properties akin to those of nitrogen-vacancy centers in diamond, which are promising as single photon sources and solid-state qubits. We present confocal fluorescence measurements of single defects in ZnO nanoparticles and sputtered films that are selectively excited by sub-bandgap light. The resulting 560-720 nm emission often exhibits two broad spectral peaks separated by approximately 100 meV. Photon correlation measurements yield both antibunching and bunching, indicative of single photon emission from isolated defects with a metastable shelving state. Excited state lifetimes span 1-13 ns and are uncorrelated with doping concentration. We report discrete jumps in the fluorescence intensity between a bright and dark state. The dwell times are exponentially distributed in each state and the average dwell time in the bright (dark) state does (may) depend on the power of the excitation laser.

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