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Highly Efficient Defect Emission from ZnO:Zn and ZnO:S Powders[1]

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Bulk Zinc Oxide (ZnO) is a wide band gap semiconductor with an ultraviolet direct band gap energy of 3.4 eV and a broad, defect-related visible wavelength emission band centered near 2 eV. We have shown that the external quantum efficiency can exceed 50% for this nearly white emission band that closely matches the human dark-adapted visual response. To explore the potential of ZnO as a rare earth-free white light phosphor, we investigated the mechanism of efficient defect emission in three types of ZnO powders: unannealed, annealed, and sulfur-doped. Annealing and sulfur-doping of ZnO greatly increase the strength of defect emission while suppressing the UV band edge emission. Continuous wave and ultrafast one- and two-photon excitation spectroscopy are used to examine the defect emission mechanism. Low temperature photoluminescence (PL) and PL excitation (PLE) spectra were measured for all three compounds, and it was found that bound excitons mediate the defect emission. Temperature-dependent PLE spectra for the defect and band edge emission were measured to estimate trapping and activation energies of the bound excitons and clarify the role they play in the defect emission. Time-resolved techniques were used to ascertain the role of exciton diffusion, the effects of reabsorption, and the spatial distributions of radiative and non-radiative traps. In unannealed ZnO we find that defect emission is suppressed and UV band edge emission is inefficient ($< 2\%$) because of reabsorption and non-radiative recombination due to a high density of non-radiative bulk traps. By annealing ZnO, bulk trap densities are reduced, and a high density of defects responsible for the broad visible emission are created near the surface. Interestingly, nearly identical PLE spectra are found for both the band edge and the defect emission, one of many indications that the defect emission is deeply connected to bound excitons. Quantum efficiency, also measured as a function of excitation wavelength, closely mirrors the PLE spectra for both emission bands. Sulfur-doped ZnO exhibits additional PLE and X-ray features indicative of a ZnS-rich surface shell that correlates with even more efficient defect emission. The results presented here offer hope that engineering defects in ZnO materials may significantly improve the quantum efficiency for white light phosphor applications.

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