Breakdown of Kasha's Rule in a Ubiquitous, Naturally Occurring, Wide Bandgap Aluminosilicate (Feldspar)

Excitation-energy-dependent emission (EDE) is well known from photoluminescence (PL) studies of polar solvents and carbon-based nanostructures. In polar solvents, this effect known as the 'red edge effect' (REE) is understood to arise from solute-solvent interactions, whereas, in case of carbon-based nanostructures, the origin is highly debated. Understanding this effect has important bearings on the potential applications of these materials. EDE has never been reported from large crystalline materials, except very recently by our group. Here, we make detailed investigations to understand the universality and the mechanism behind the EDE in a wide band gap aluminosilicate (feldspar), which comprises more than half of the Earth's crust, and is widely used in geophotonics (e.g., optical dating). We observe EDE up to 150 nm at room temperature in our samples, which is unprecedented in rigid macroscopic structures. Based on PL investigations at 295 K and 7 K, we present a novel model that is based on photoionisation of a deep lying defect and subsequent transport/relaxation of free electrons in the sub-conduction band tail states. Our model has important implications for potential photonic applications using feldspar, measurement of band tail width in wide bandgap materials, and understanding the EDE effect in other materials.

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