

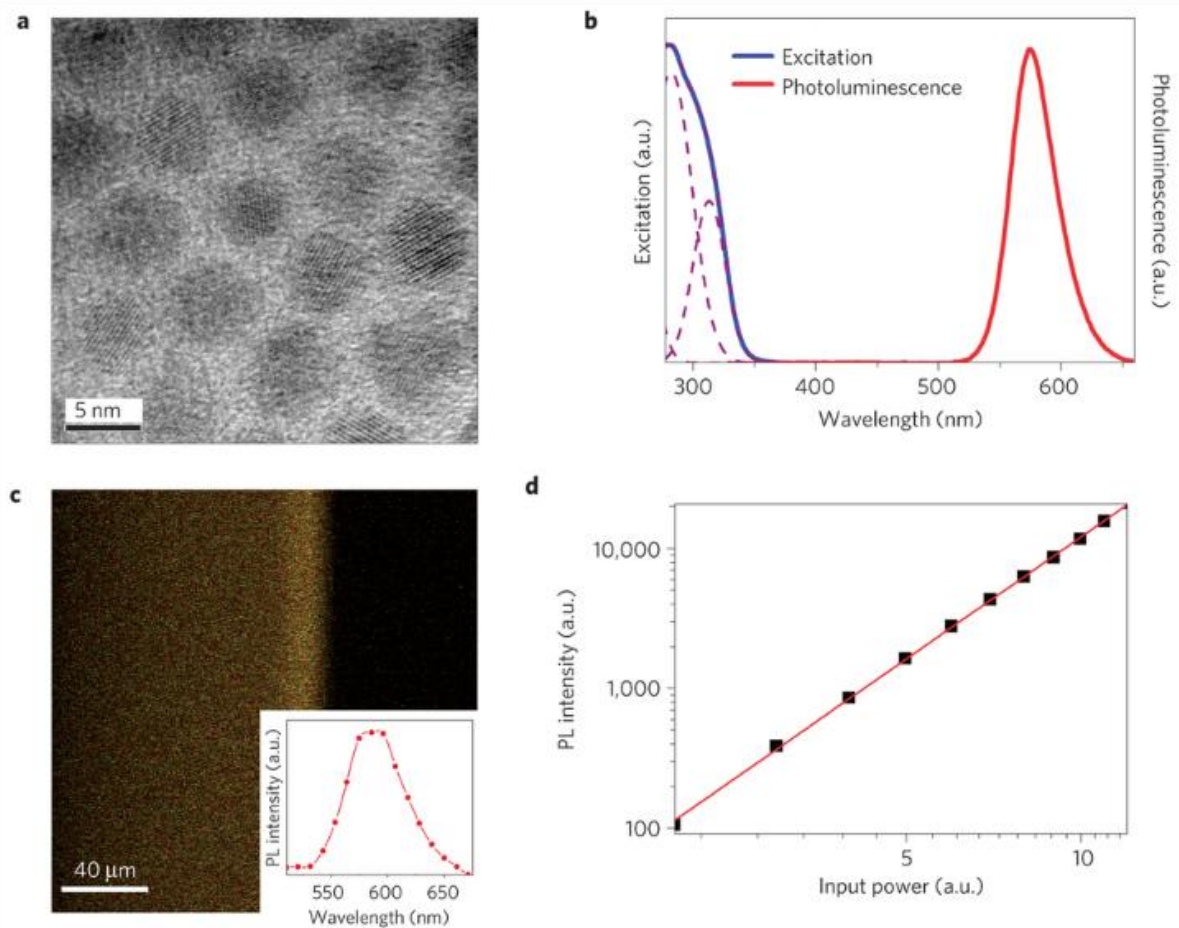
Figure 1: Synthesis and characterization of ZnS:Mn NCs.

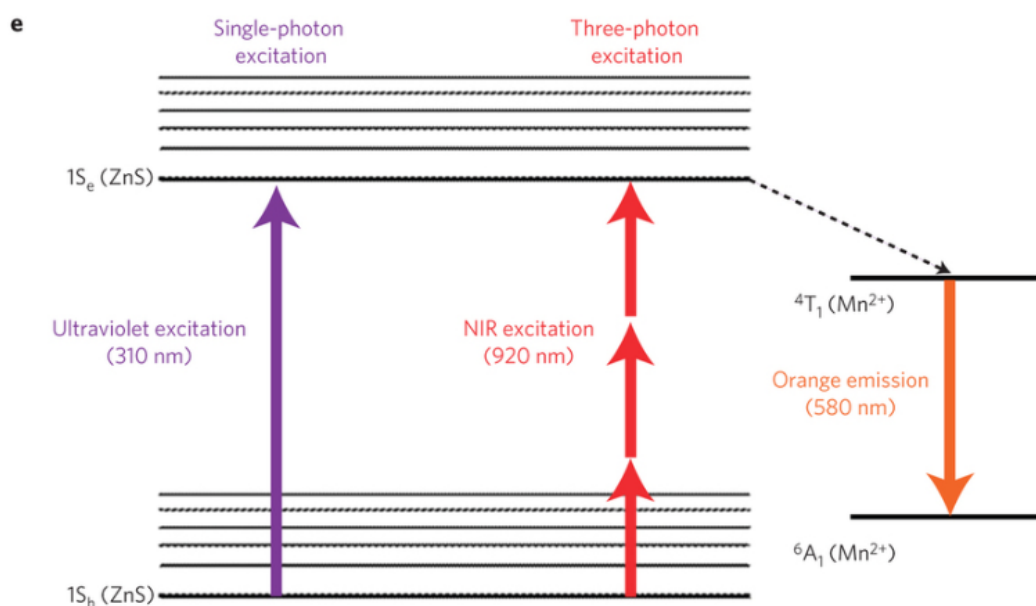
From

High-resolution three-photon biomedical imaging using doped ZnS nanocrystals

Jung Ho Yu, Seung-Hae Kwon, Zdeněk Petrášek, Ok Kyu Park, Samuel Woojoo Jun, Kwangsoo Shin, Moonkee Choi, Yong Il Park, Kyeongsoon Park, Hyon Bin Na, Nohyun Lee, Dong Won Lee, Jeong Hyun Kim, Petra Schwille & Taeghwan Hyeon

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a, Transmission electron micrograph of ZnS:Mn NCs (average size, 5.5 nm; s.d., 15%). b, Normalized one-photon PLE (blue) and photoluminescence (red) spectra of ZnS:Mn NCs. The PLE spectrum, fitted by summing multiple Gaussian functions (purple dashed line), shows the 1S<sub>e</sub>–1S<sub>h</sub> transition peaked at 314 nm. c, Multiphoton-excited luminescence spectral image of the ZnS:Mn NC thin film. The NCs were excited with a 920 nm Ti:sapphire laser. Inset: luminescence spectrum extracted from the image. d, Log–log plot of the power dependence obtained from c. The slope of the power dependence (red line) is 2.9. e, Jablonski diagram of one-photon and three-photon excited luminescence in a NC.

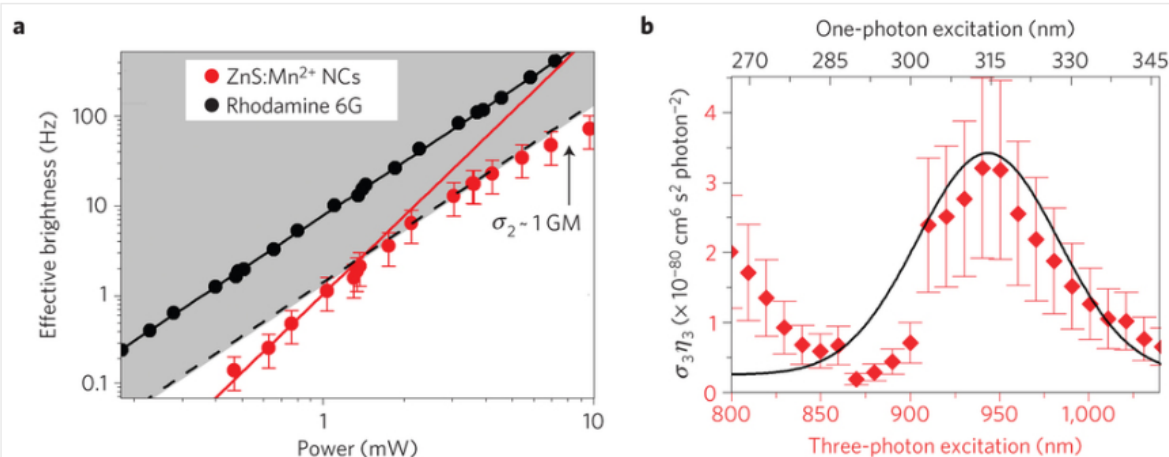
Figure 2: Three-photon luminescence characteristics of ZnS:Mn NCs.

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a, The power dependence of effective single-particle brightness for 3PL of a NC (red circles) and for 2PL of Rhodamine 6G (black circles), obtained from fluorescence correlation spectroscopy. The NCs and Rhodamine 6G in water were excited with a 920 nm Ti:sapphire laser, and the detected photon counts were normalized to the numbers of the NCs and Rhodamine 6G fluorophores within the focal volume (see Supplementary Information). The slope of the power-dependence function is 2.9 (red line) for the NC and 2.0 (black line) for Rhodamine 6G, respectively. Dashed line, power-dependence curve corresponding to the two-photon action cross-section of 1 GM unit, calibrated from the two-photon cross-section of Rhodamine 6G to be 5.7 GM. The grey region corresponds to brightness over 1 GM. b, The wavelength-dependent three-photon action cross-section (red diamonds) and the 1S<sub>e</sub>–1S<sub>h</sub> transition curve (black line), which were derived from the wavelength dependence of the 3PL and the multiple Gaussian fitting of the one-photon PLE spectrum (Fig. 1b), respectively. The error bars represent s.d., calibrated from the standard deviation of the NC concentration based on their size distribution.