Uncovering the structure of InP-based quantum dots

Mary Burkitt-Gray,[a] Simon Fairclough[a], Jack Chun-Ren Ke[b], Wijittra Wichiansee[a], Philippa Clark[b], Edward Lewis[c], Wendy Flavell[b], Sarah Haigh[c], Mark Green[a]*

[a] *Department of Physics, King's College London, UK*
[b] *School of Physics and Astronomy, University of Manchester, UK*
[c] *School of Materials, University of Manchester, UK*

InP-based quantum dots (QDs) are an attractive alternative to II-VI materials, particularly for applications where toxicity or waste disposal is of concern [1, 2, 3]. The growth of a ZnS shell, formation of an InZnP alloy, and use of suitable surface species can result in quantum yields of up to 70% [4], making InP-based systems promising for fluorescence applications.

InP/ZnS QDs are among the most commonly researched III-V systems due to their biocompatibility [3], visible fluorescence, and colour-tunable emission [4]. Heterostructured InP/CdS has also been reported [5], but there has been little in-depth research into the bonding of the system. It is typically assumed that both InP/ZnS and InP/CdS have traditional core/shell structures [5, 6], despite the difficulties faced in achieving a crystalline InP core [4, 7]. Only a few previous reports, most notably by Reiss et al. [4, 8], have discussed the bonding and alloying within InP and InP/ZnS nanoparticles. There have been no detailed studies on alloying between InP QDs and CdS, and alterative InP-based heterostructures have largely not been considered.

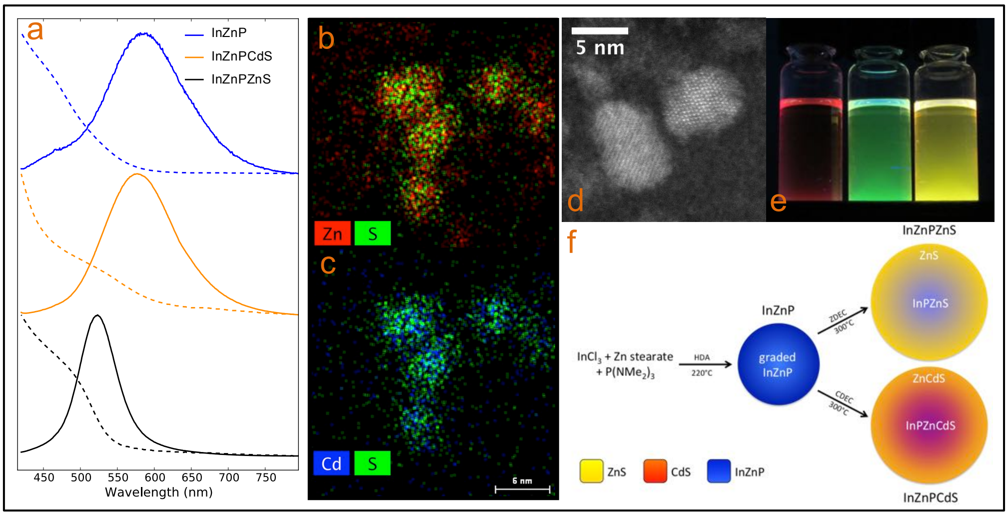

Fig. 1 a. Emission and absorption spectra InZnP, InZnPCdS, and InZnPZnS; b. EDS mapping of Zn and S in InZnPCdS; c. EDS mapping of Cd and S in InZnPCdS; d. HAADF-STEM of InZnPCdS; e. fluorescent InZnP, InZnPZnS and InZnPCdS QDs; f. schematic of the reaction process and alloyed products.

We have performed a typical "core/shell" InZnP/ZnS synthesis using a single-source ZnS precursor [9], as shown in figure 1. However, both the InZnP core and the "core/shell" particles were found to have an unusual graded structure with visible-range fluorescence. The CdS analogue of the procedure [9] identified that both Zn^{2+} and Cd^{2+} ions were highly mobile within InZnP, forming bright InZnPCdS QDs -- the first known report of a CdS-alloyed InP-type nanoparticle. In-depth structural analysis revealed unusual bonding with no clear phase boundaries, in contrast to the crystalline core/shell systems that are typically reported. These results question whether many of the so-called InP/ZnS and InP/CdS nanoparticles may instead have more complex alloyed structures, highlighting the need for further research on this important family of quantum dots.

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