Electronic supplementary information

Impact of Coherent Core/Shell Architecture on Fast Response in InP-based Quantum Dot Photodiodes

Kazuhiro Nemoto,[a,b] Junpei Watanabe,[b,c] Hiroyuki Yamada,[a,b] Hong-Tao Sun,[b] and Naoto Shirahata*[a,b,c]

[a]Graduate School of Chemical Sciences and Engineering, Hokkaido University, Sapporo 060-0814, Japan.

[b]International Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, 305-0044, Japan.

[c]Department of Physics, Chuo University, 1-13-27 Kasuga, Bunkyo, Tokyo 112-8551, Japan.

Fig. S1. A typical digital photograph of the dried powder of the coherent InP/ZnS core/shell QDs capped with MCH monolayers.

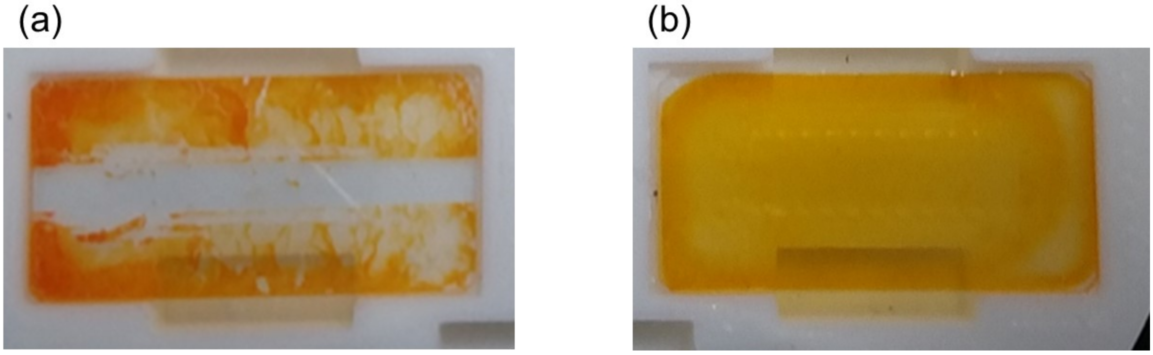

Fig. S2. The representative digital photographs of the QD films prepared by dropping (a) poor MCH-capped QD and (b) MCH-capped QD dispersed in DMF on ITO-covered glass substrates.

NOTE: Unlike the poor MCH-capped QD which was obtained by inadequate ligand exchange, the MCH-capped QDs could formed a uniform and flat film on the ITO surface.

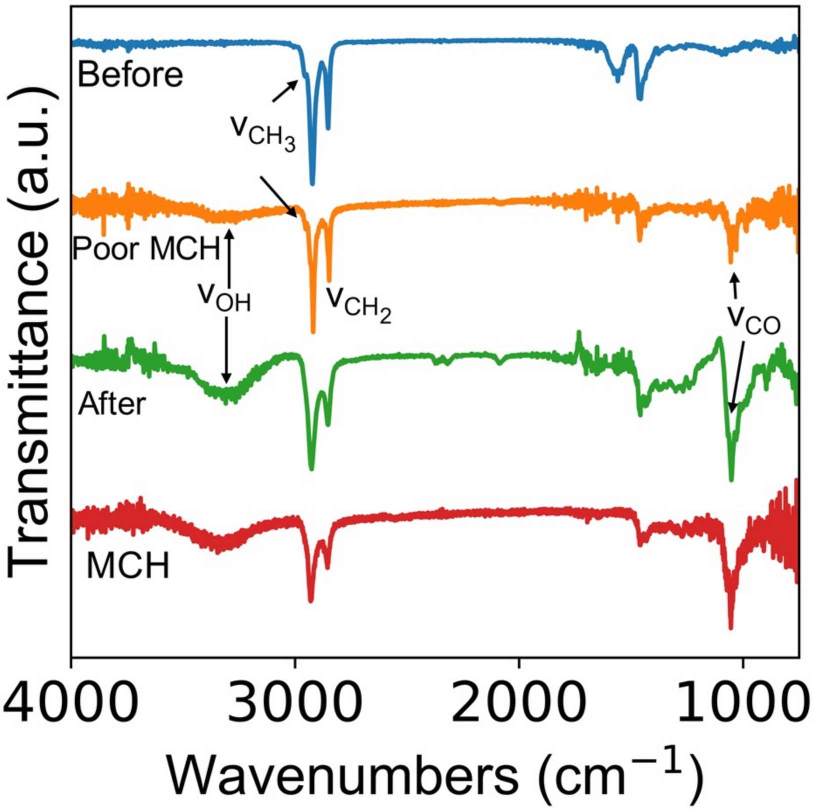

Fig. S3. Comparison of FTIR between Poor MCH QD before and after ligand exchange QD.

NOTE: Poor MCH QD has low intensity ratio from MCH. In addition, the peak of the stretching motion of CH_3 can be seen. Thus, the QD of Poor MCH QD represents having two ligands, OA or PA and MCH.

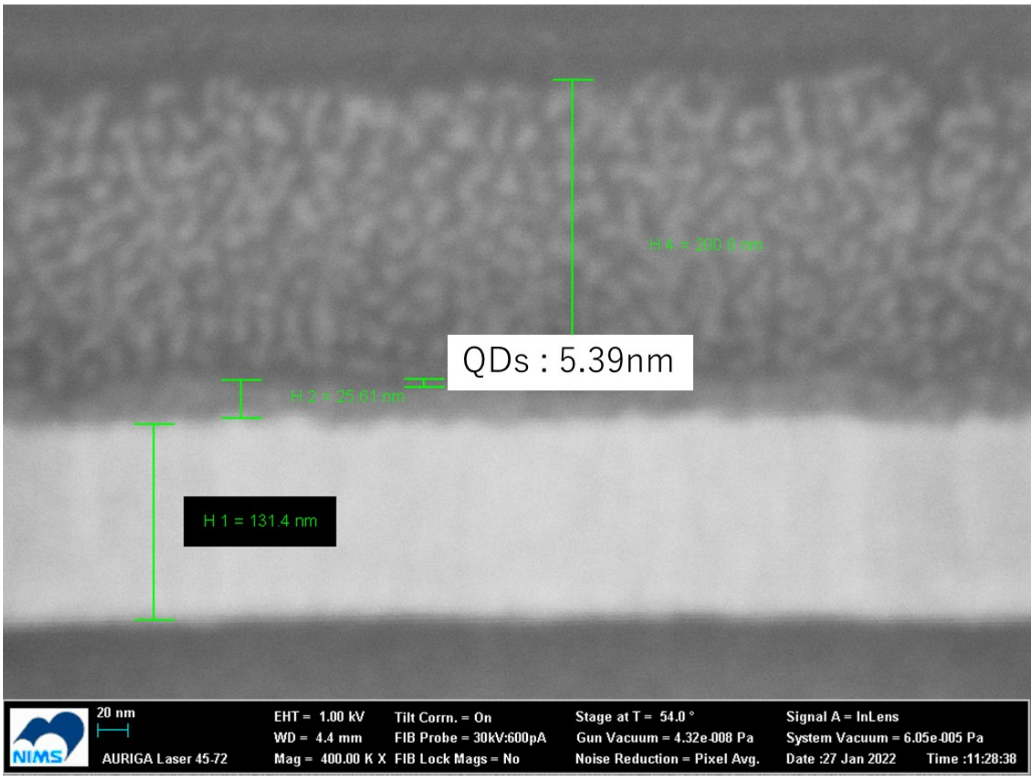

Fig. S4. Cross-sectional SEM photographs of photodiode device with a multilayer structure of ITO/ZnO(Al)/poor MCH-InP/ZnS QD/Al.

NOTE: For the QD ink, MCH-capped InP/ZnS QD with inadequate ligand exchange was used. As is clearly seen in the image, we could not obtain the uniform film of QDs. Furthermore, it was difficult to obtain the QD film thicker than 5.39nm even by repeating the spin-coating process with the QD ink.

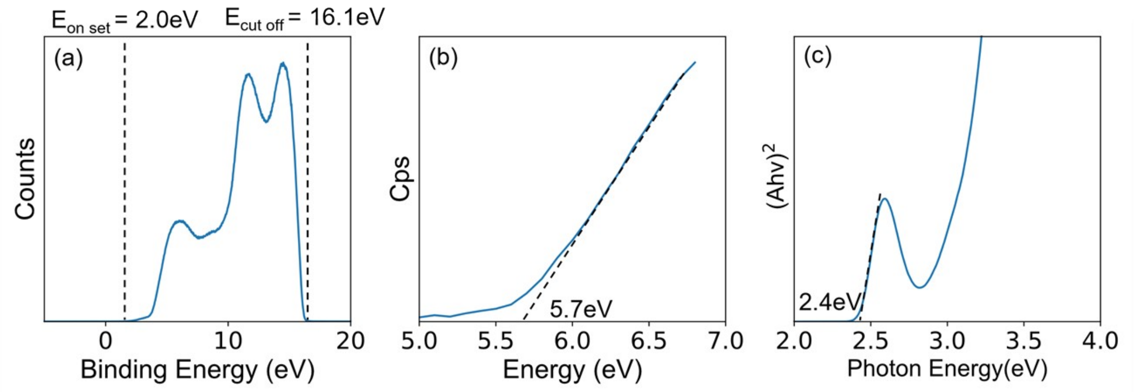

Fig. S5. (a) Ultraviolet photoelectron spectroscopic (UPS) spectrum of the Al-doped ZnO spin-coated thin film plotted as a function of the energy and (b) Photoelectron yield spectroscopic (PYS) spectrum of a film form of the coherent InP/ZnS core/shell QDs capped with MCH ligands.

NOTE

The value of ionization energy (E) was calculated using the values derived from E_{cutoff} and E_{onset} according to the following equation [1].

$$-E = \hbar v - (E_{cut\,off} - E_{on\,set}) \qquad (1)$$

where ħv is the irradiation energy (i.e., 21.21 eV). The high binding energy E_{cutoff} and E_{onset} are 16.1eV and 2.0eV, respectively. As a result, the value of ionization energy was calculated to be 7.12 eV.

REFERENCES

1. S.-H. Wu, M.-Y. Lin, S.-H. Chang, W.-C. Tu, C.-W. Chu and Y.-C. Chang, J. Phys. Chem. C, 2018, 122, 236–244.

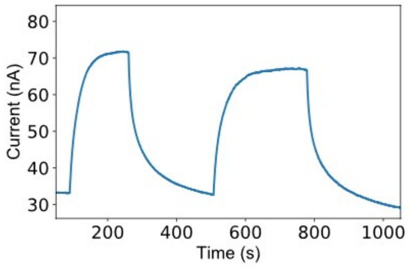

Fig. S6. Photo-response characteristics of the photodiode where PA-QD was used as a light-absorbing layer at a voltage bias of 0 V under irradiation of 470-nm light with power density of 10 mW/cm^2.