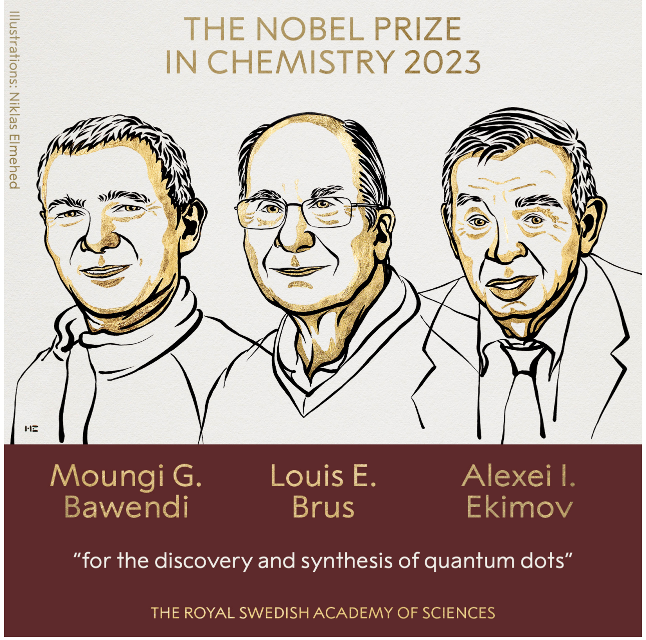
Water-based Quantum Dots Liquid Scintillator for Radiation Detection

Noble prize in Chemistry 2023 is about quantum dots!



Outline

1. Introduction
2. Optical measurements
3. Cosmic ray test
4. Conclusion

Collaboration with Miao Zhao and Sasha Rakovich (photonics and nanotechnology group at King's),
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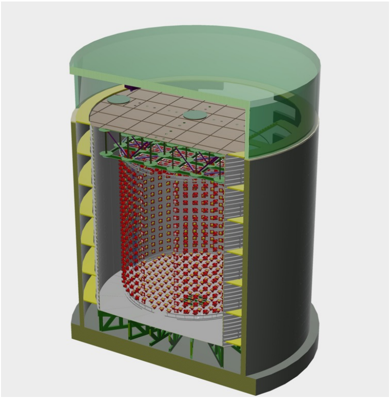
1. Nuclear reactor monitoring neutrino detector

Anti-neutrino detector offers remote monitoring of fission reactor activities
- Illegal core extraction
- Discover unknown nuclear reactor

Study found gadolinium-doped water-based liquid scintillator detector has the best sensitivity to detecting anti-neutrinos from nuclear reactors

Here, we study the feasibility of a neutrino detector based on the water-based quantum dots liquid scintillator



Large detector (~kton) = far field
Small detector (~1ton) = near field

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1. Quantum dots (QDs)

Semiconductor nano-crystal

- Emission spectrum is tunable (core type, size)
- Surface layers to protect, change properties

Water-based QDs

- Hydrophilic layer to make water solution

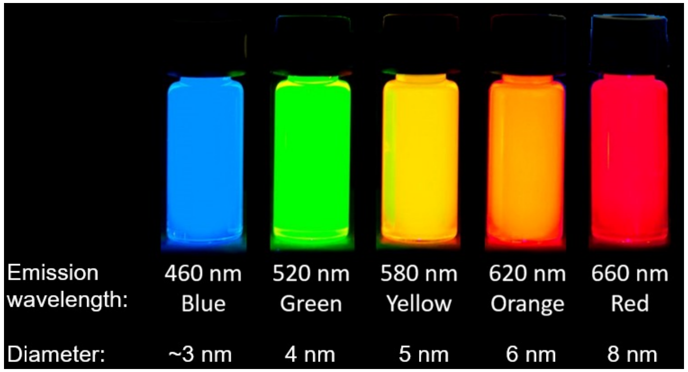
QDs are superior to traditional organic dyes and many pros

Cons:

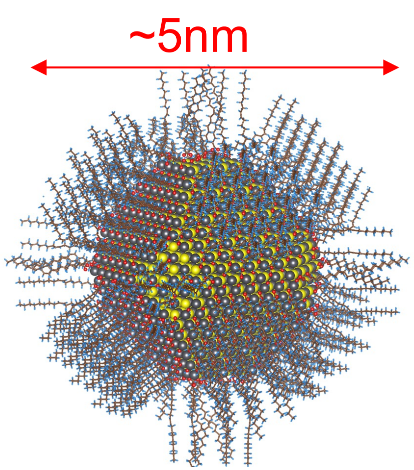
- expensive
- core material is often toxic heavy metal

We choose CdS core ZnS shell QDs

- Relatively cheaper
- Shorter emission (~450nm)
- Application of cadmium neutron capture (neutrino detector)



CdSe core QDs with CdS/ZnS shell
https://www.lateralflows.com/quantum-dots/



Colloidal quantum dot
https://en.wikipedia.org/wiki/Quantum_dot

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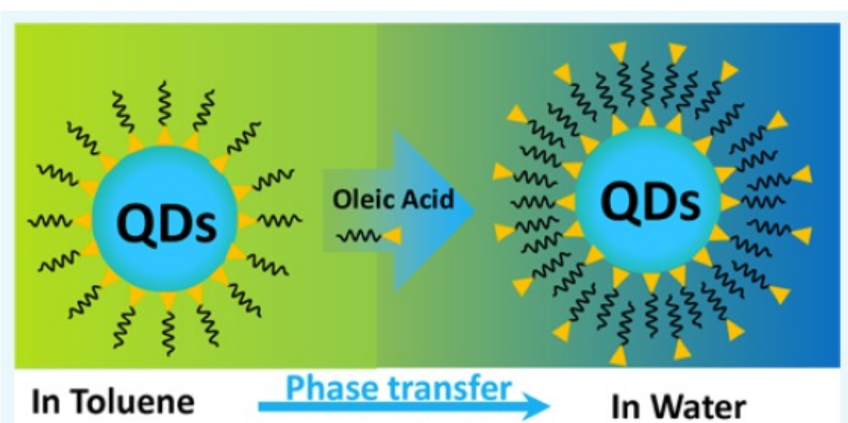
https://dx.doi.org/10.1021/acsomega.7b00316

1. Phase transfer

How to make water-based quantum dot solution

1. Dissolve CdS-ZnS QDs (10mg) in toluene
2. Evaporate toluene
3. Add hexane, sonic bath 1min
4. Add oleic acid
5. Add water
6. Sonic bath
7. Shake
8. Sonic bath, shake ... (1hr)
9. Leave in dark 24 hours



~~Witchcraft~~ Chemistry is complicated

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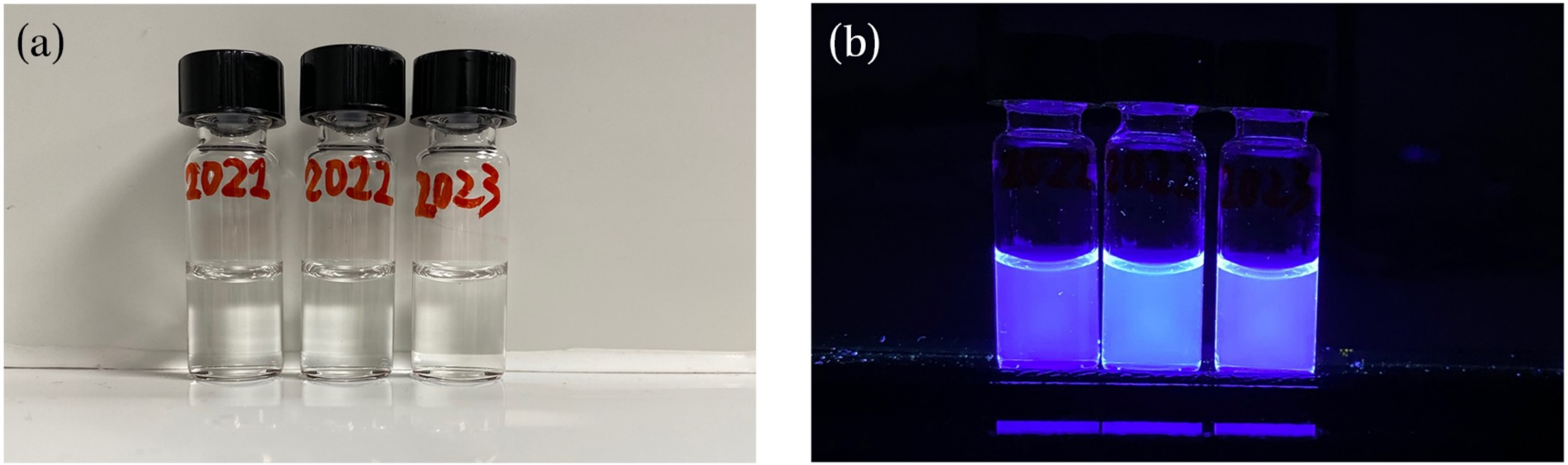
1. Water-based quantum dots

We made 3 water-based QDs sample
- 2021, 2022, 2023
- Optical measurements are based on 2023 sample in 2023
- Cosmic ray test is based on 2022 sample in 2022
- 2022 and 2021 samples are also used to study ageing



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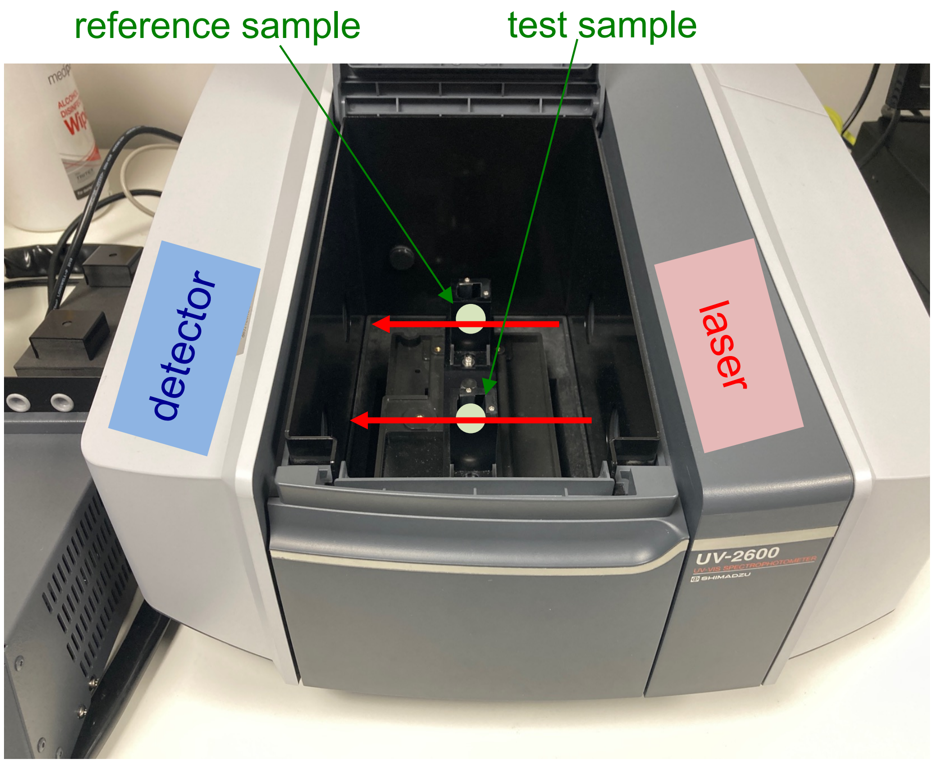
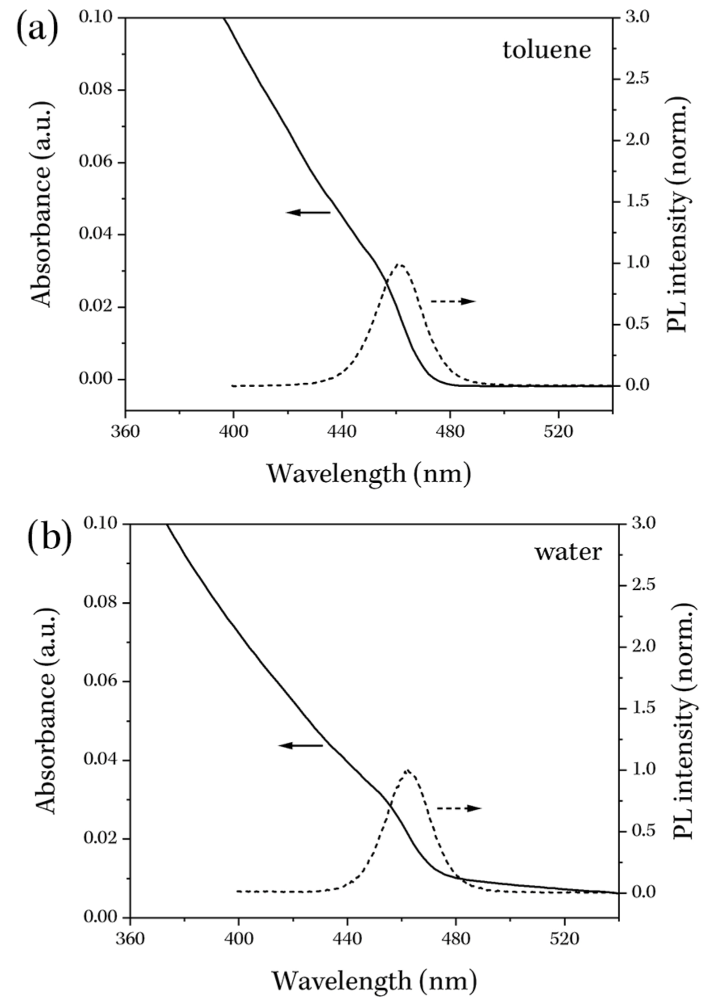
2. Absorbance measurement

Shimadzu UV-2600i spectrometer

- toluene QDs and water QDs show the same absorption peak (~460nm)
- No significance change of QD core by phase transfer





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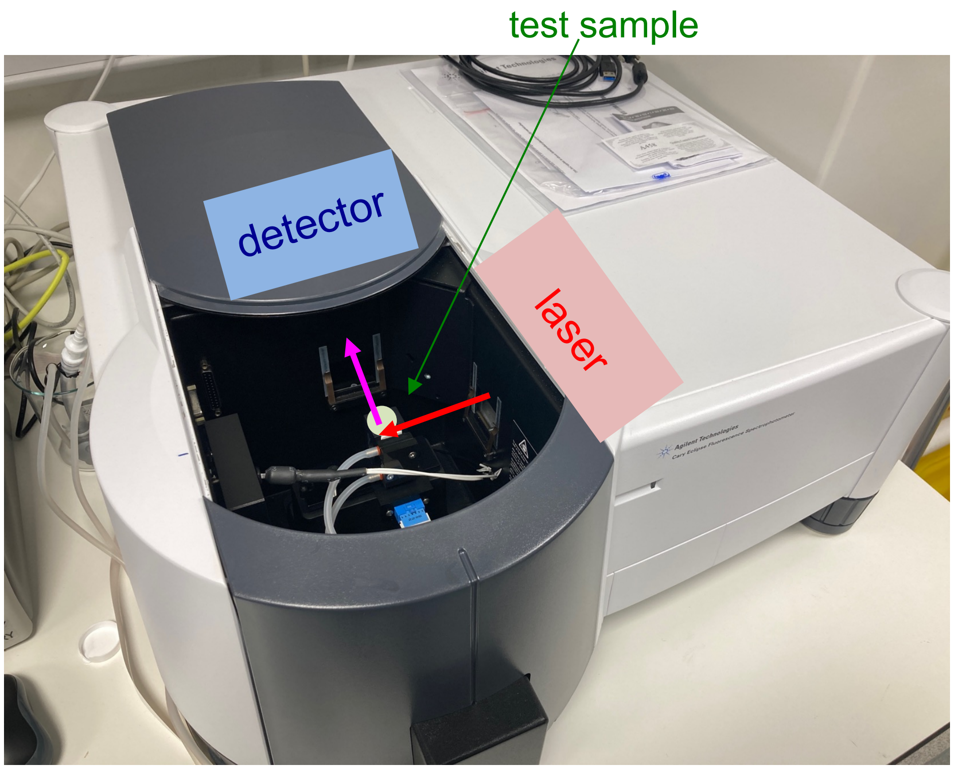
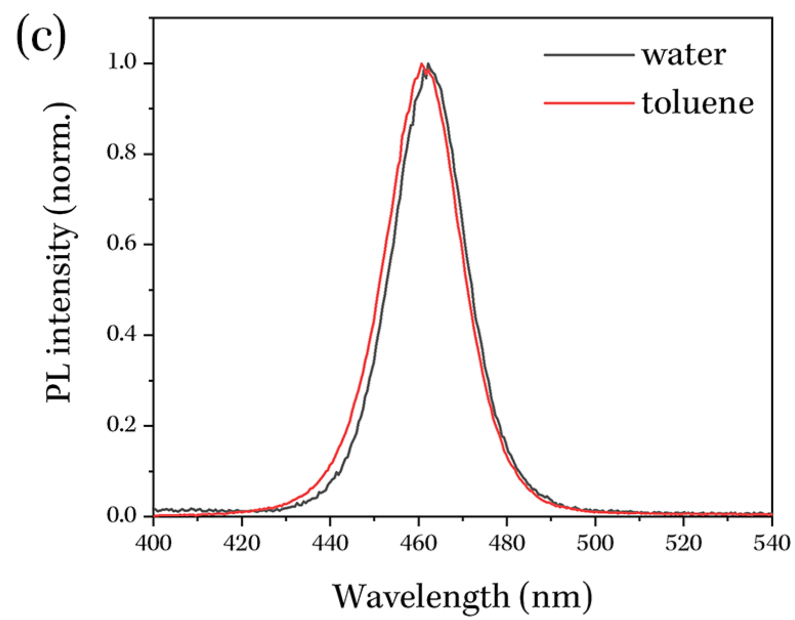
2. Fluorescence measurement

Agilent Technologies Cary Eclipse Fluorescence Spectrometer

- toluene QDs and water QDs show the same emission peak
- No significance change of QD core by phase transfer





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2. Fluorescence measurement

Agilent Technologies Cary Eclipse Fluorescence Spectrometer

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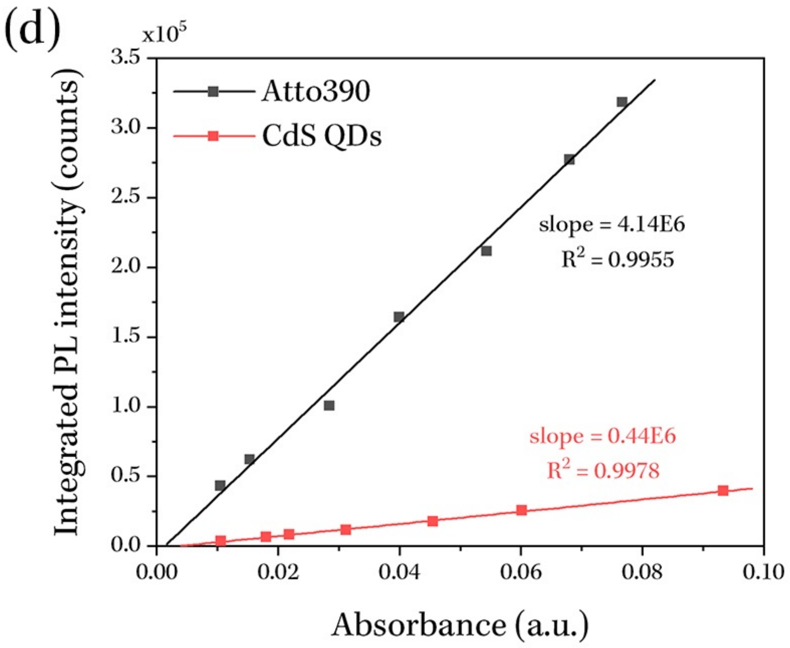
Photoluminescence quantum yield (PLQY)

$$I(A) = I_o + K \cdot A$$

- Integrated PL intensity is a linear function of absorbance. Both integrated PL intensity and absorbance are measured with different concentration of the sample
- Obtained slope “K” is compared with the reference dye Atto390
- Measured PLQY is lower than the company value (~50%) suggesting the phase transfer lose some PLQY



PLQY = 9.5±0.5%

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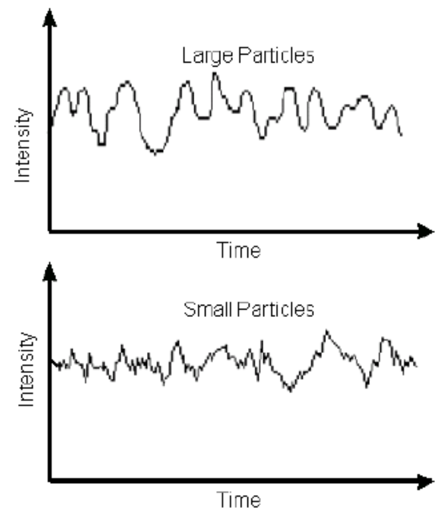
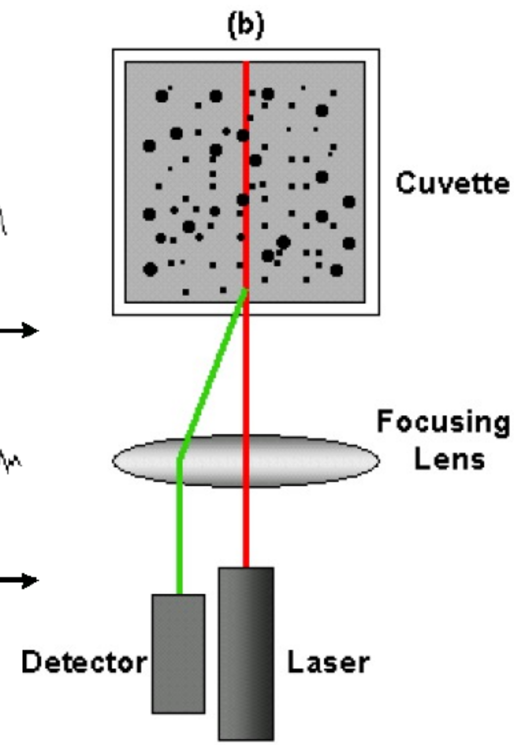
2. Dynamic light scattering (DLS) measurement

Malvern instruments Zetasizer Nano

- Measure the size of QDs from the back-scatted light
- Brownian motion gives time-dependent data





https://www.malvernpanalytical.com/en/products/technology/light-scattering/dynamic-light-scattering

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2. Dynamic light scattering (DLS) measurement

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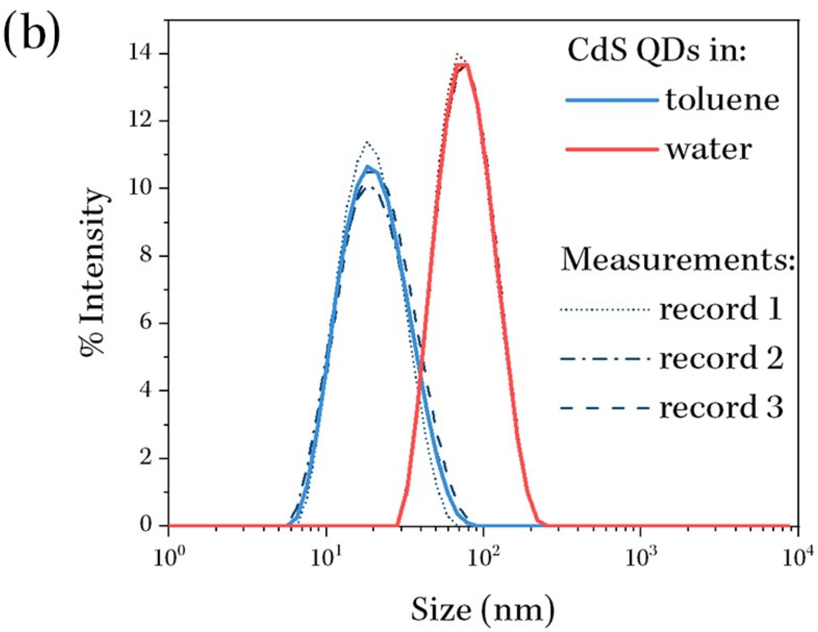
Hydrodynamic diameter d_H

$$d_H = \frac{k_B T}{3\pi\eta D}$$

- Correlation function to extract average hydrodynamic diameter of QDs
- Phase transfer adds oleic acid hydrophilic layer to the QDs
- Interesting to check the result by other method to measure the QD size



Toluene QDs ~ 20nm
Water QDs ~ 70nm

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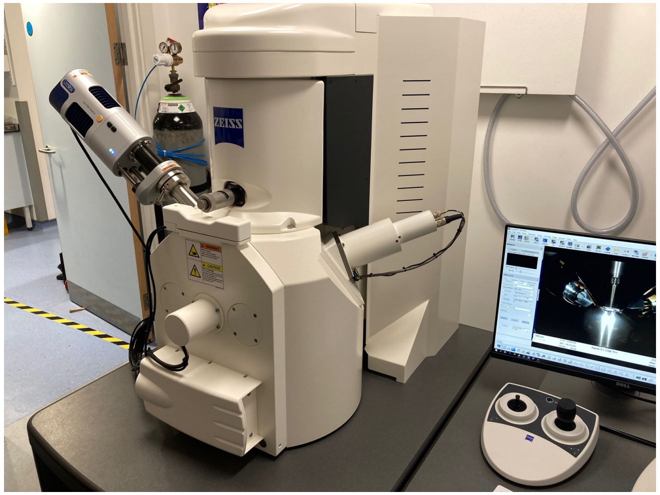
2. SEM, AFM, TEM

Scanning electron microscope (SEM)
- Sample is dried
- Gold is vacuum evaporated to make it conductive
- Not sensitive to QD size (~20nm)

Atomic force microscope (AFM)
- Sample is dried
- Not sensitive to X-Y because of large tip
- Z measurement may be good?

Transmission electron microscope (TEM)
- Our next step



Scanning electron microscope (SEM)

Atomic force microscope (AFM)

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2. Ageing

Absorbance and fluorescence
- No change over the period of 3 years

Size
- Slight decrease of the size (a few nm)
- Likely not losing layers, but re-organization of surface structure
- Zeta potential (surface charge) is changing over time, supporting this interpretation
- No sign of agglomeration

Photoluminescence quantum yield (PLQY)
- Large sample variation makes difficult to conclude

It seems water QDs are stable, but without ageing information of PLQY it's hard to say if they are suitable for particle detectors (~several years operation)

Quantum dots lifetime is
3 years (company value)

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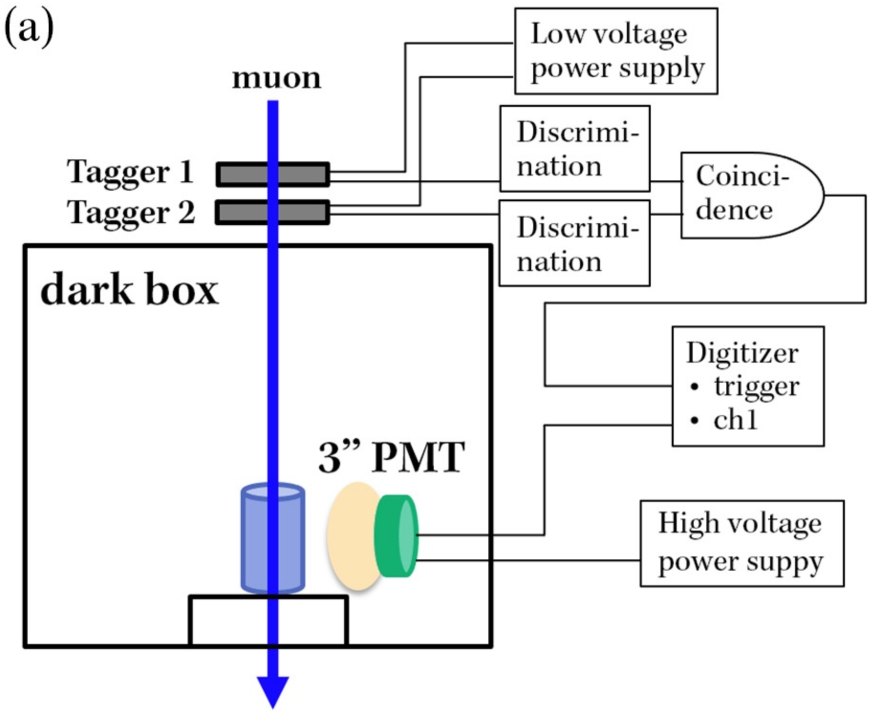
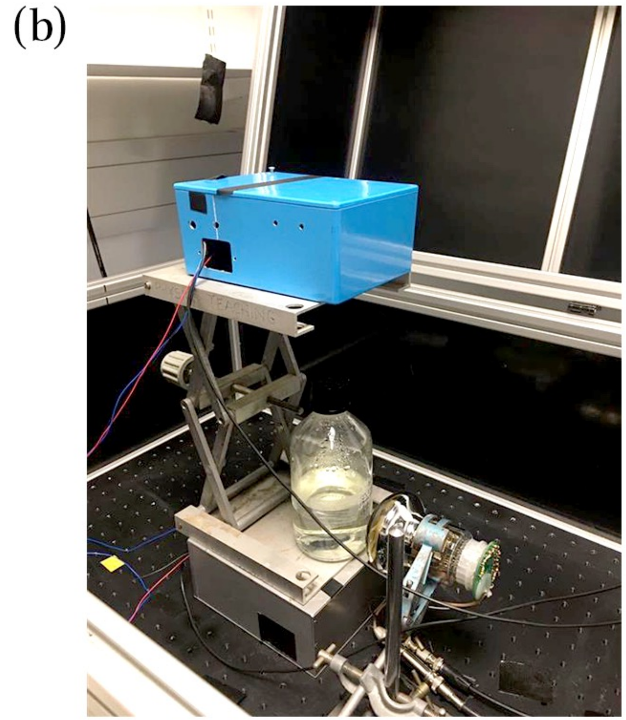
3. Cosmic ray test

Set up

- Cosmic ray triggers (plastic scintillator + SiPM)
- 3-inch NNVT PMT
- CAEN 250MS/s digitizer





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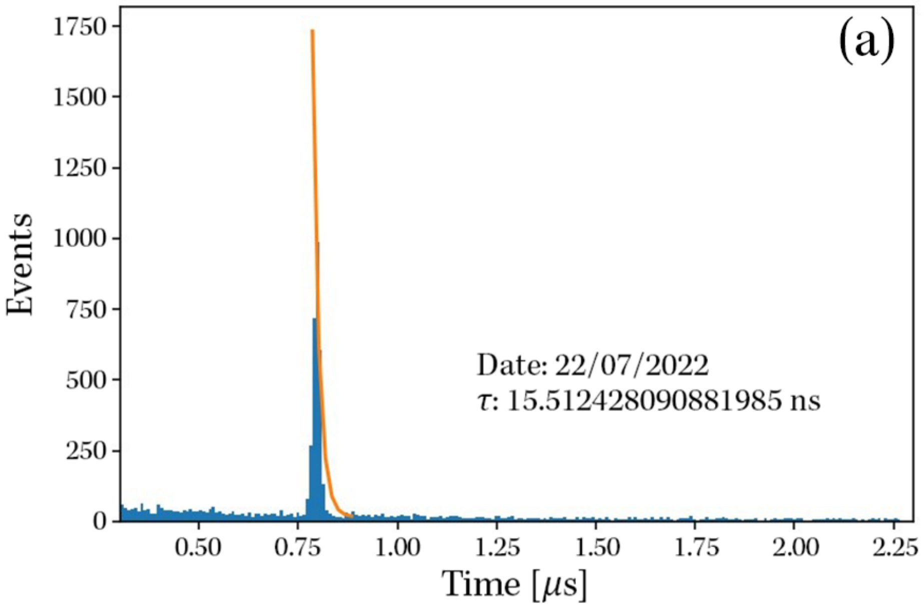
3. Cosmic ray test

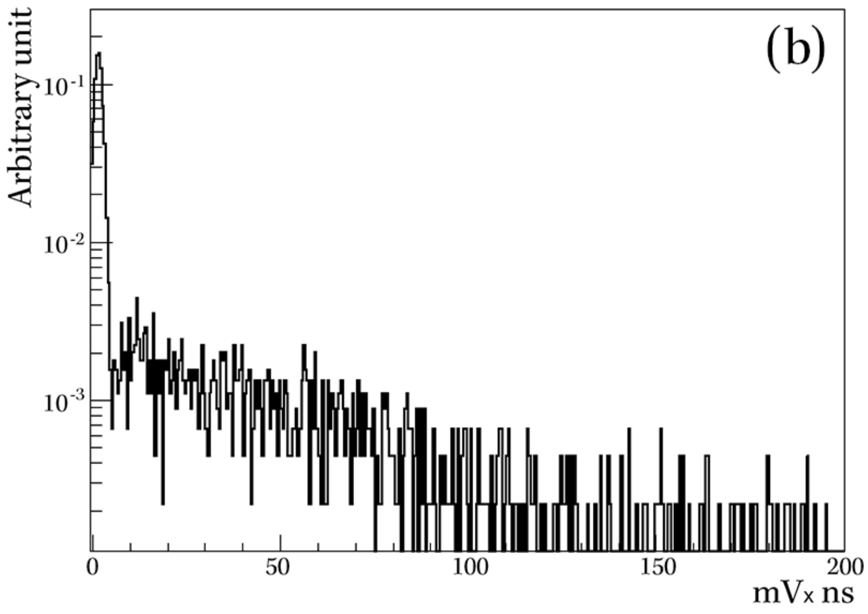
Timing data

- Quantum dots decay time is around 5ns
- Too fast to measure by 250MS/s (4ns bin) digitizer

Charge data

- Need simulation to extract information

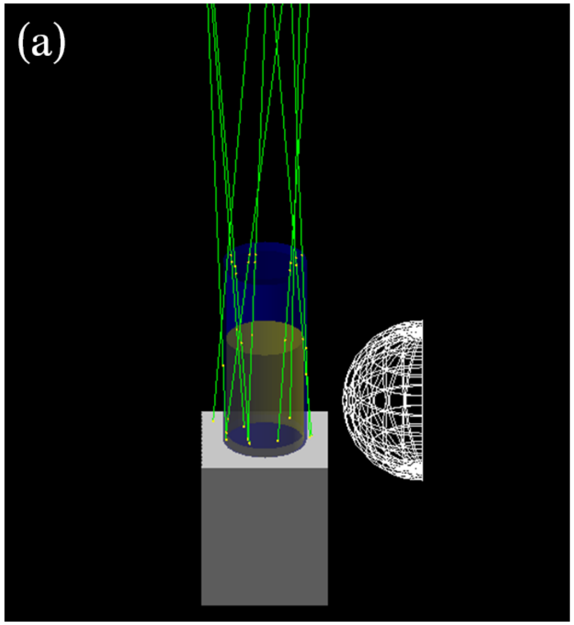


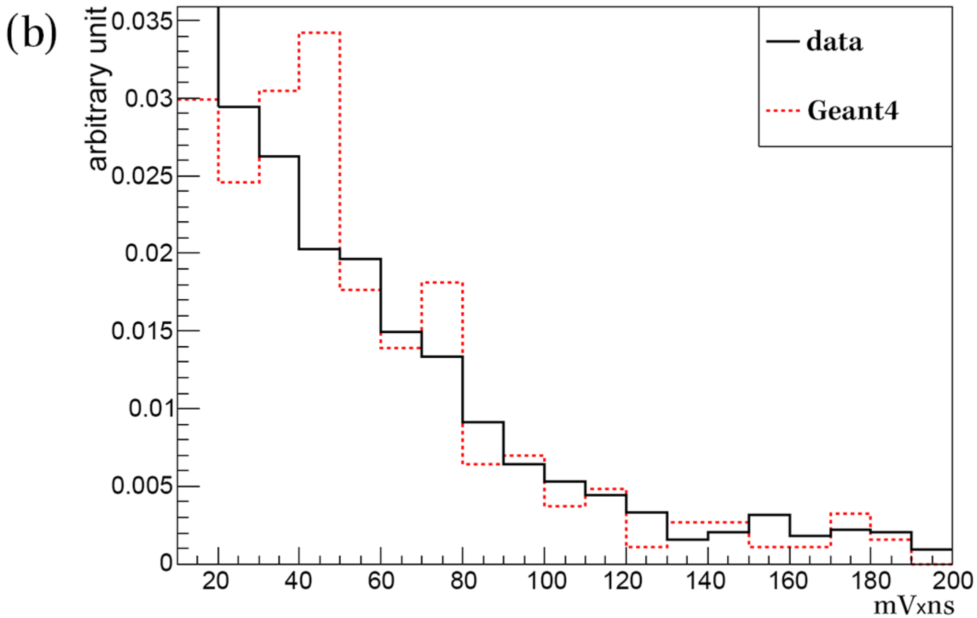


3. Cosmic ray test

Data-Simulation comparison

- Geant4-based simulation
- Simulation is adjusted to match the tail of the charge distribution
- Data seems to have several 1000s photons / MeV
- Comparable emission strength with modern organic scintillator





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4. Conclusions

Water-based liquid scintillator is successful made using CdS-ZnS quantum dots.

Water QDs and toluene QDs have the same absorbance and fluorescence peaks.

Water QDs photoluminescence quantum yield is 9.5±0.5%, lower than the company value (~50%).

Hydrodynamic diameter is measured by DLS, and water QDs (~70nm) is larger than toluene QDs (~20nm) as expected.

QD sizes change slightly aver the time, and no sign of agglomeration. Absorbance and emission spectrum don’t change over 3 years. PLQY ageing result is not conclusive.

Scintillation response is measured from cosmic muons. Time constant seems very fast as expected (<15ns), and charge response suggests photon yield is comparable as typical organic liquid scintillators.

Thank you for your attention!

Backup