Heralded spectroscopy with a linear SPAD array spectrometer

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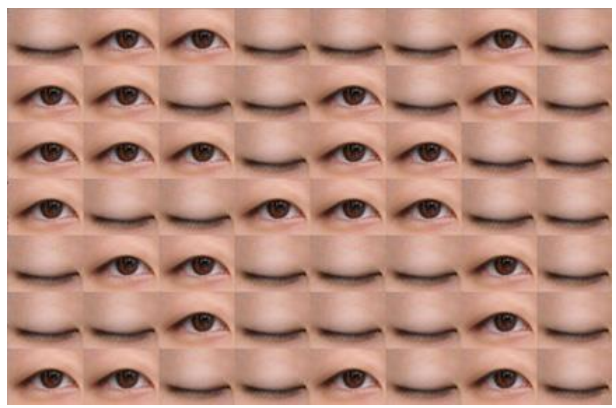


ISSW 2022, June 2022

Outline

- Reminder on photon correlations
- What are colloidal quantum dots?
- Some applications of photon correlations in quantum dot spectroscopy

  - Multiexciton spectroscopy by photon statistics

  - Heralded spectroscopy of quantum sources

- Conclusion

The Hanbury Brown and Twiss stellar interferometer

HB&T proposed a new kind of telescope to measure the angle subtended by an object in the sky – which does not require a large mirror to resolve the size

1046 NATURE November 10, 1956 Vol. 178

A TEST OF A NEW TYPE OF STELLAR INTERFEROMETER ON SIRIUS

By R. HANBURY BROWN
Jodrell Bank Experimental Station, University of Manchester
AND
Dr. R. Q. TWISS
Services Electronics Research Laboratory, Baldock

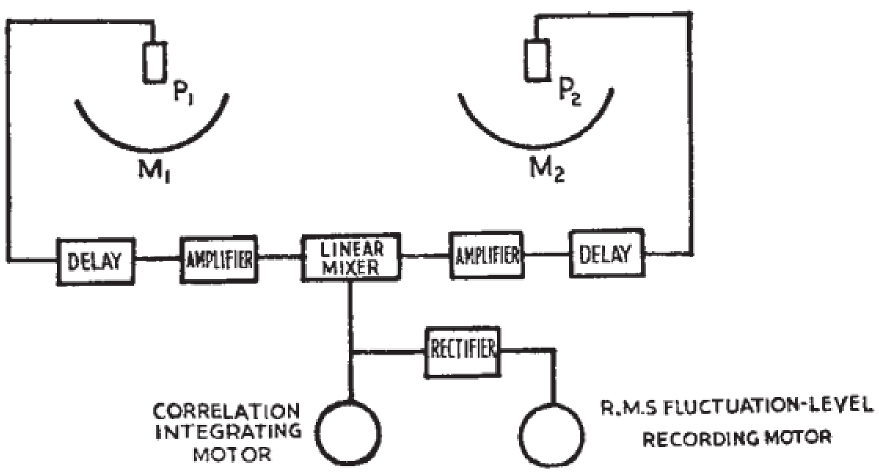


Fig. 1. Simplified diagram of the apparatus

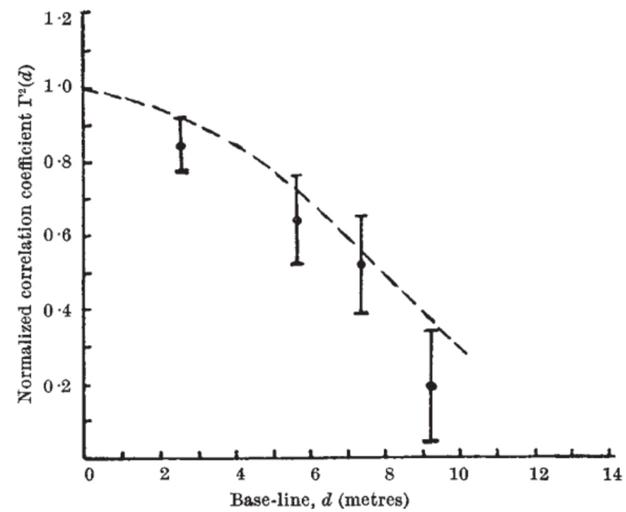


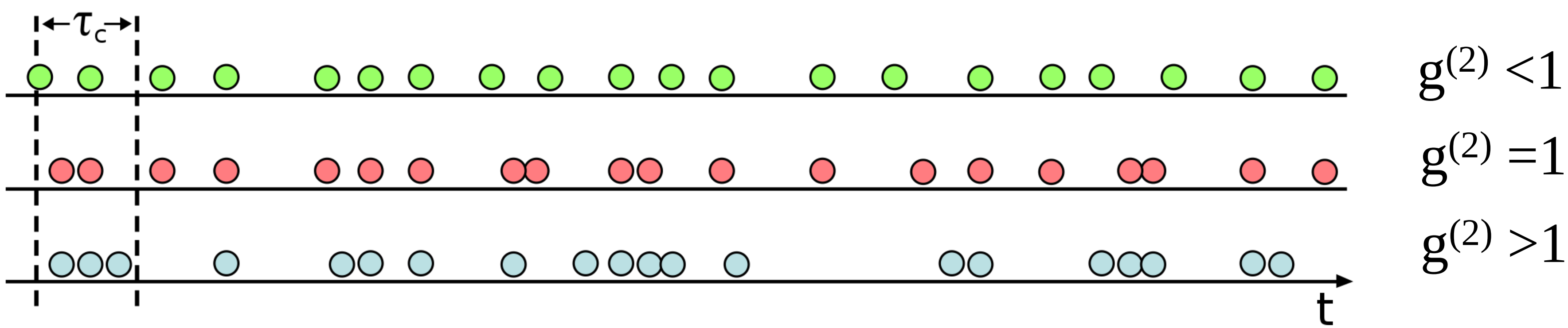
Fig. 2. Comparison between the values of the normalized correlation coefficient $\Gamma^2(d)$ observed from Sirius and the theoretical values for a star of angular diameter 0·0063″. The errors shown are the probable errors of the observations

Photon statistics and photon time distribution

A convenient characteristic of photon statistics is the second order correlation coefficient $g^{(2)}$

$$g^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2) = \frac{\langle E^*(\mathbf{r}_1,t_1)E^*(\mathbf{r}_2,t_2)E(\mathbf{r}_1,t_1)E(\mathbf{r}_2,t_2)\rangle}{\left\langle |E(\mathbf{r}_1,t_1)|^2\right\rangle \left\langle |E(\mathbf{r}_2,t_2)|^2\right\rangle}$$

For which we only consider the temporal degree of freedom

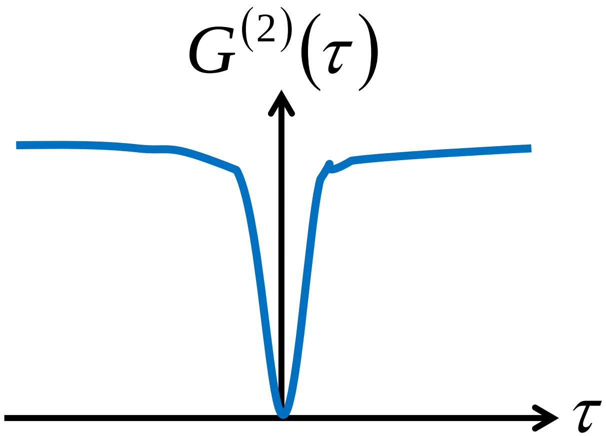


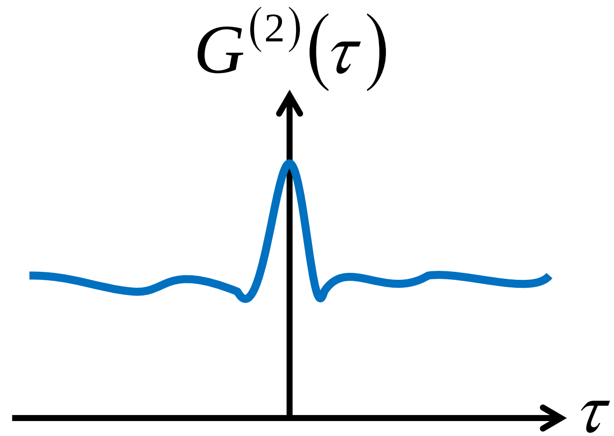
Photon detections as function of time for a) antibunched, b) random, and c) bunched light

$g^{(2)} < 1$ is a sufficient condition for nonclassicality!

What can we learn from photon statistics?

Intensity Correlation $G^{(2)}(\tau) = \langle I(t) I(t+\tau) \rangle$





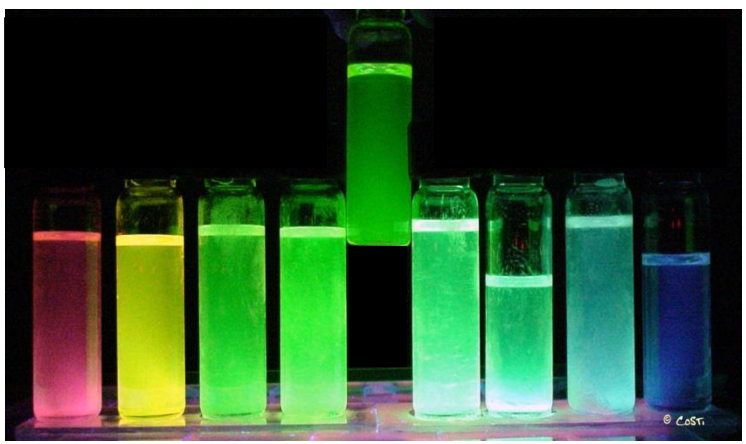
Our ‘targets’:
Colloidal semiconductor quantum dots

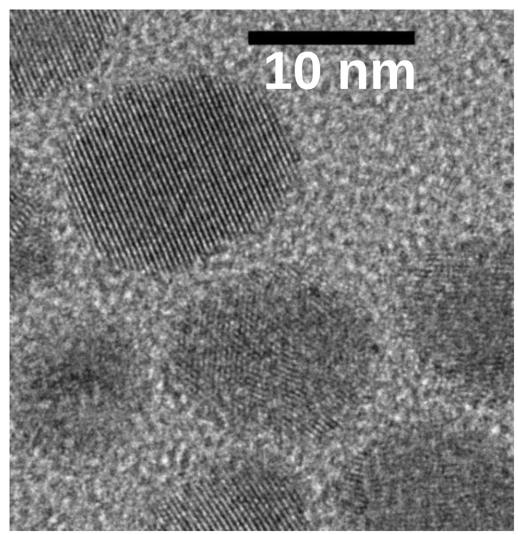
Nanocrystals prepared in solution from ionic precursors

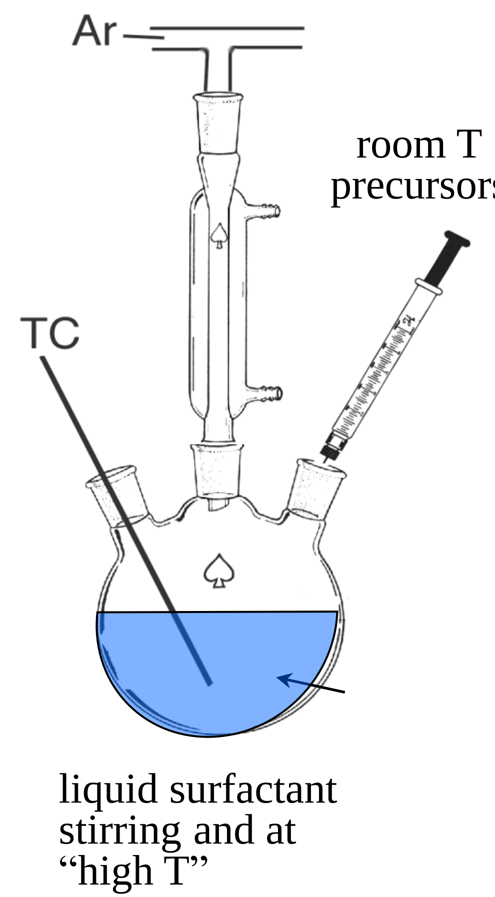
Well controlled size distribution via “nucleation and growth” mechanism

CdSe nanocrystal solutions

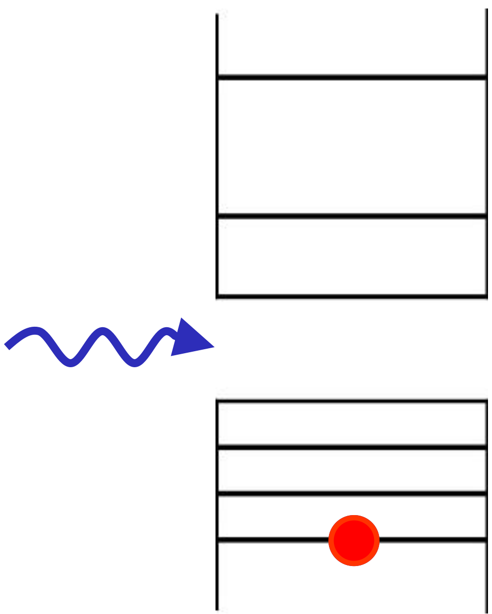
d=6nm ← d=2nm







What happens when you excite a quantum dot?



Phonon
bottleneck?

picoseconds

nanoseconds

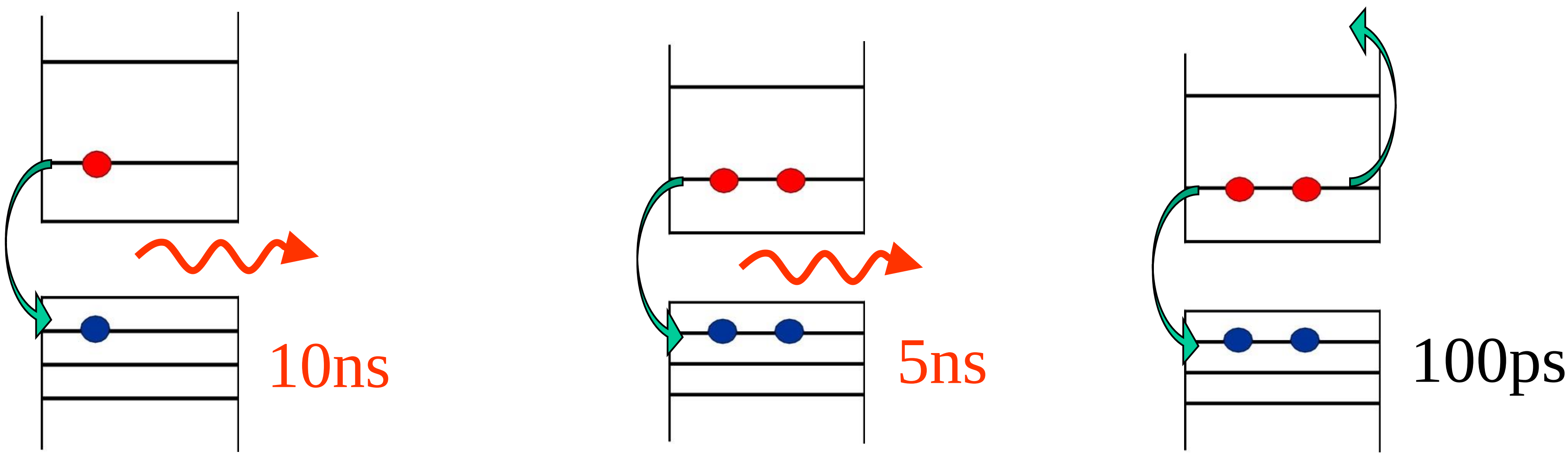
Multiexcitons and Auger recombination

Single exciton
undergoes radiative recombination

Biexciton
Can decay to an exciton via two recombination channels

Radiative recombination

Auger recombination



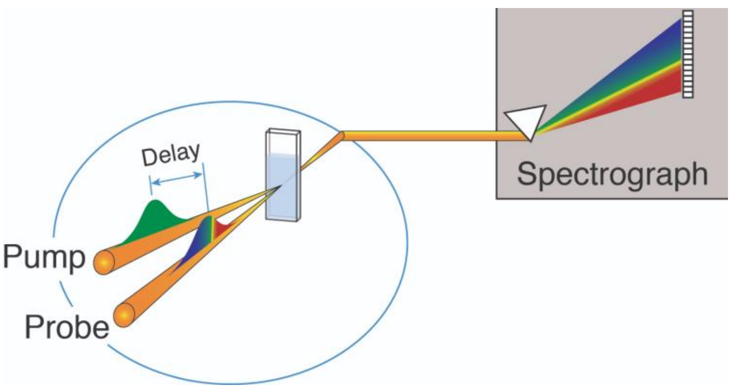
Concomitantly, there are spectral shifts due to exciton-exciton interactions

Chepic et al., J. Luminescence 47, 113 (1990) ; Woggon et al., J. Luminescence 59, 135 (1994)

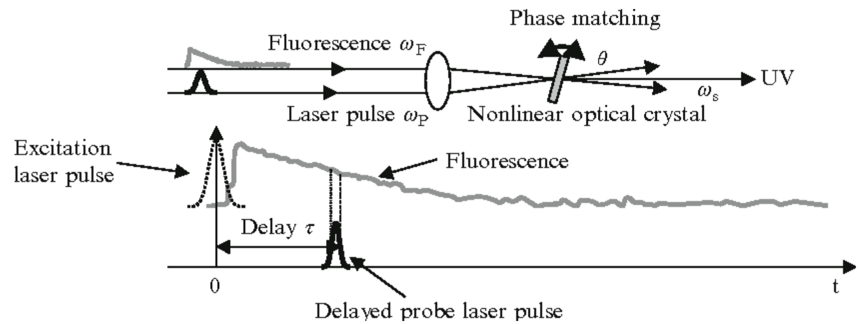
‘Common’ multiexciton spectroscopy methods

Need to resolve both temporal and spectral data

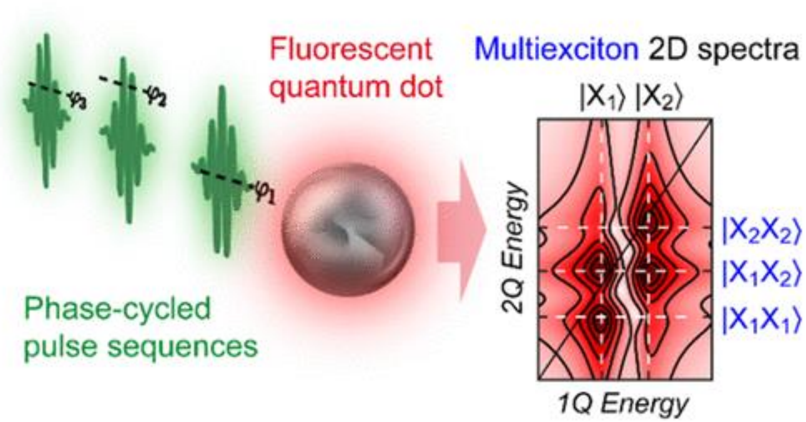
Transient absorption



Transient PL



Multidimensional spectroscopy



But all these are ensemble measurements, averaging over temporal and inter-QD heterogeneity!

Why single nanoparticles

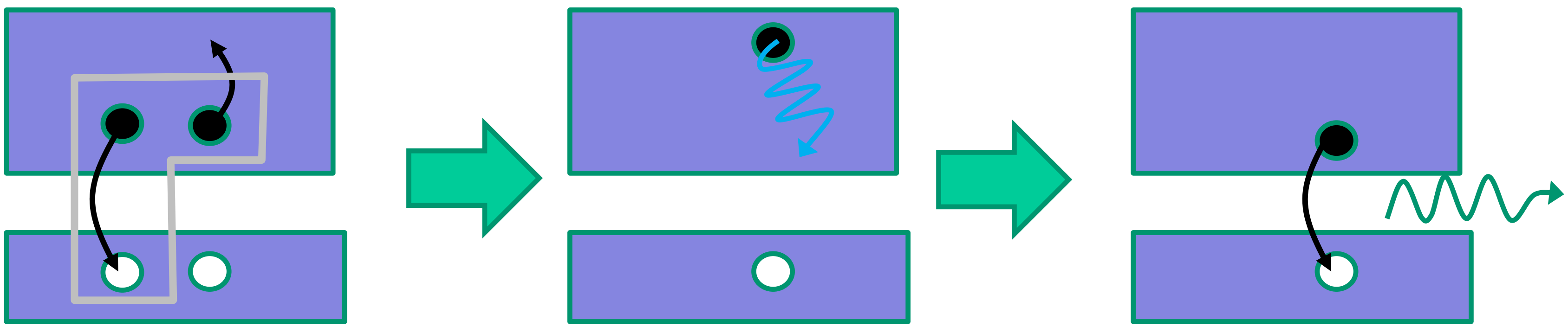
- Local measurements
- Overcoming inhomogeneous broadenings
- Quantum nature of light emission

How?

- Scattering (very hard, scales as V^2)
- Absorption (hard, scales as V but small)
- Photoluminescence (easy, background free)

From Auger recombination to photon statistics

Auger Recombination

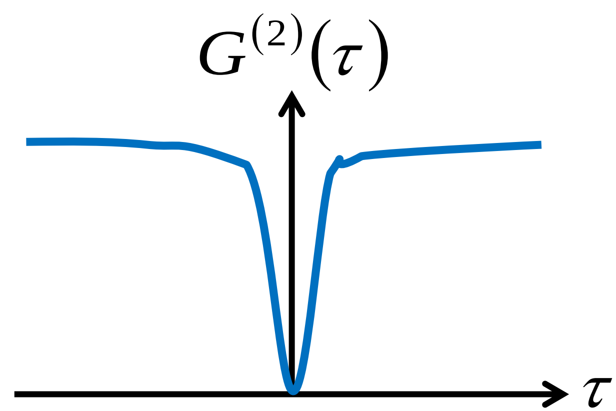


Photon Anti-Bunching

Intensity Correlation

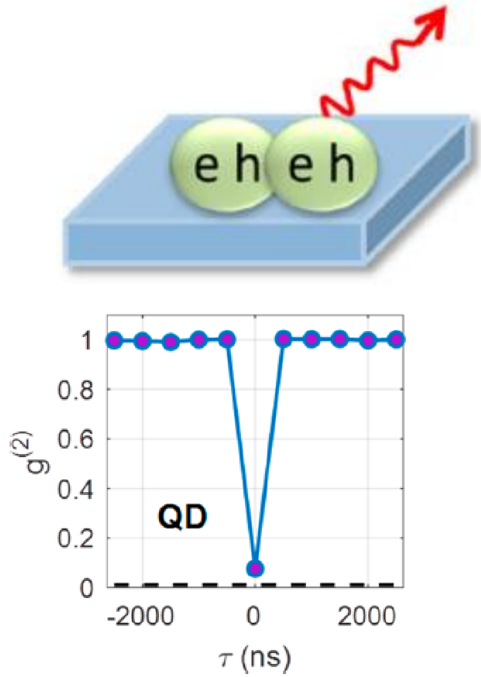
$$G^{(2)}(\tau) = \langle I(t)I(t+\tau)\rangle$$

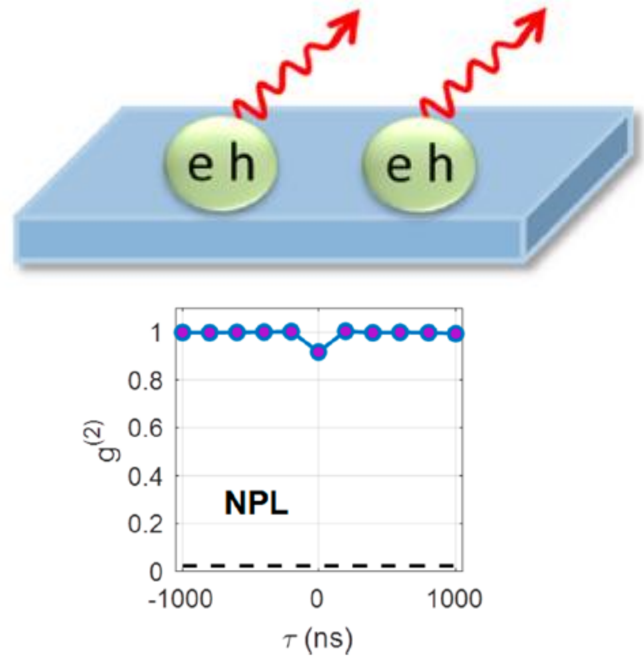
Reduced quantum fluctuations



Quantum spectroscopy

Can we replace 'traditional' spectroscopy with photon statistics?



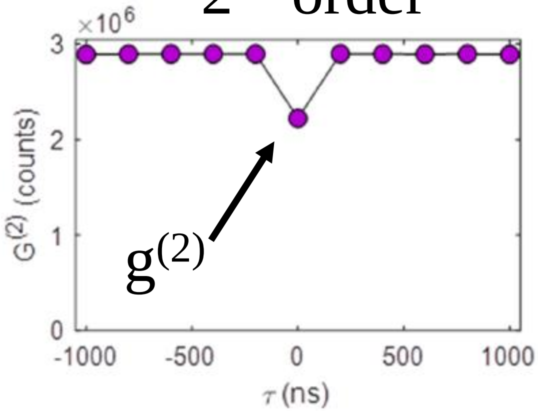


In larger nanocrystals (e.g. nanoplatelets) antibunching is not complete.
Is there information in the higher order photon correlations?

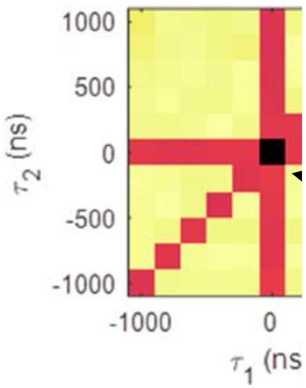
The short answer is "Yes" …

Higher order antibunching spectroscopy

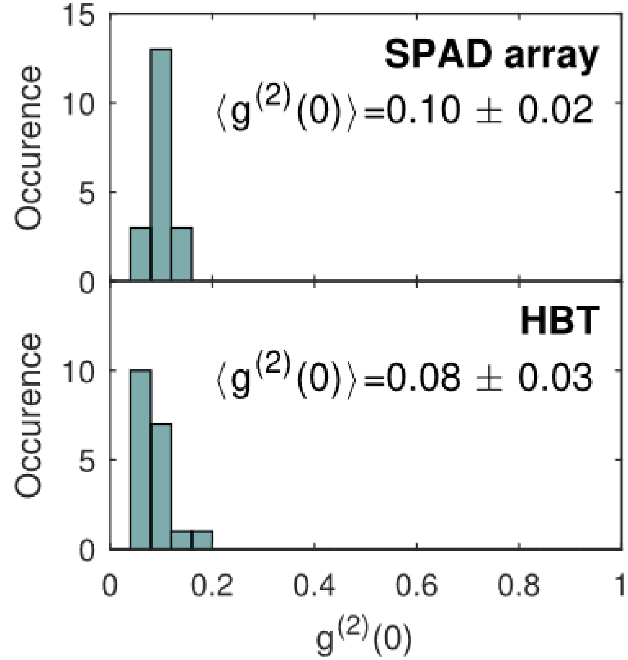
2^{nd} order

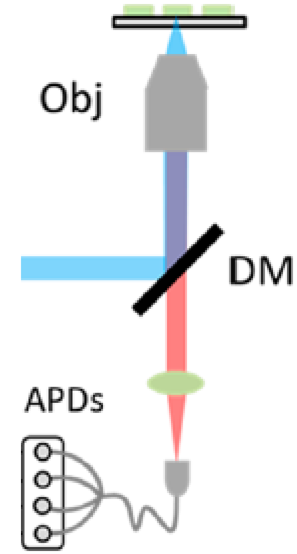


3^{rd} or

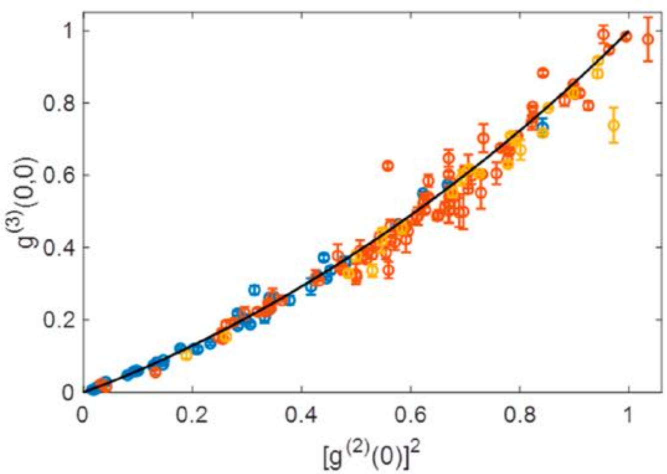


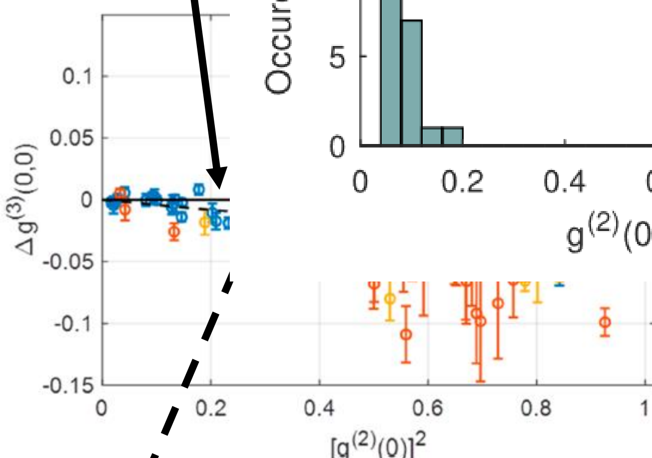
Can also be done with SPAD arrays

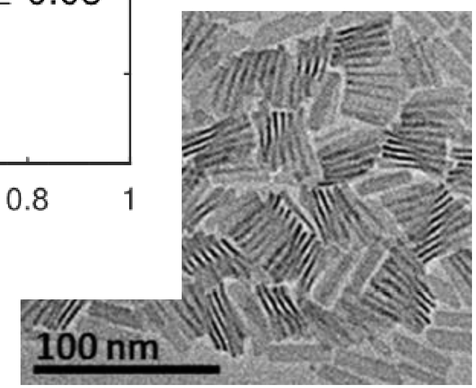




Two body inte







With three body correction

D. Amgar, DO, et al., Nano Lett. 19, 8741 (2019) ; G. Lubin, DO et al., Optics Express 27, 32863 (2019)

Heralded multiexciton spectroscopy

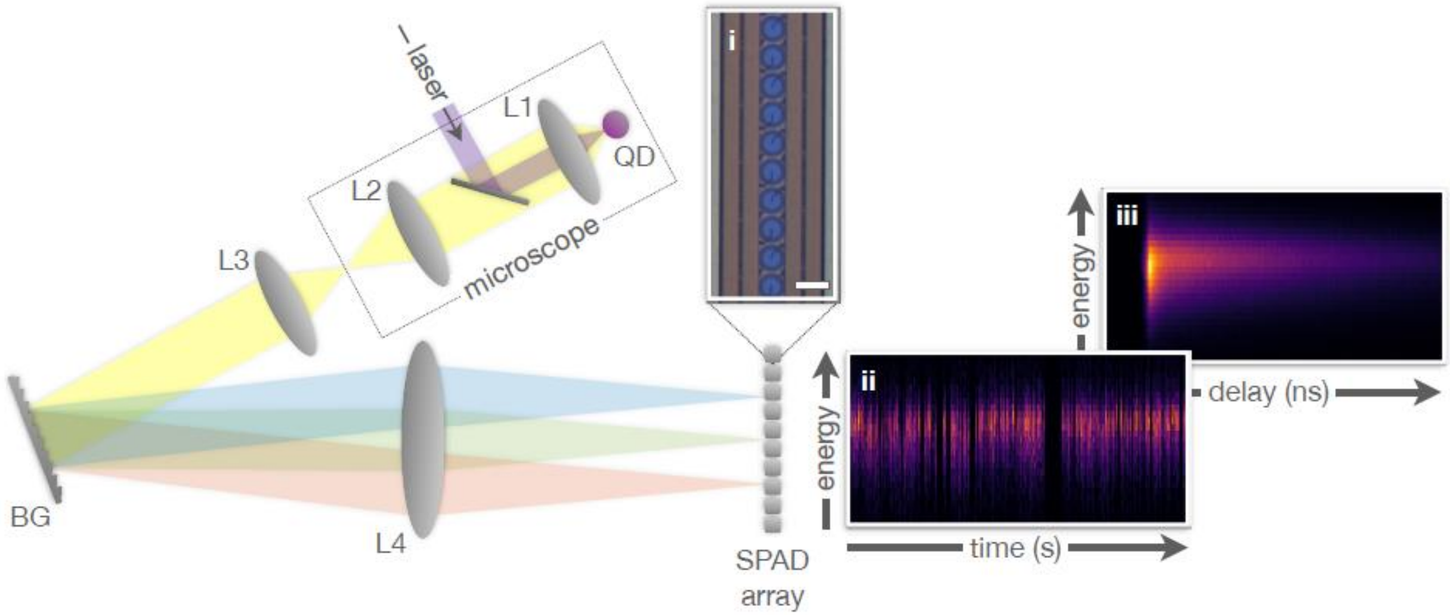
But in performing photon statistics experiments we had to give something up …

we have lost all spectral information!

Is there a way around this?

Heralded multiexciton spectroscopy

New technologies such as monolithic arrays of single photon spectrometers can provide access to previously unexplored properties at the single particle level



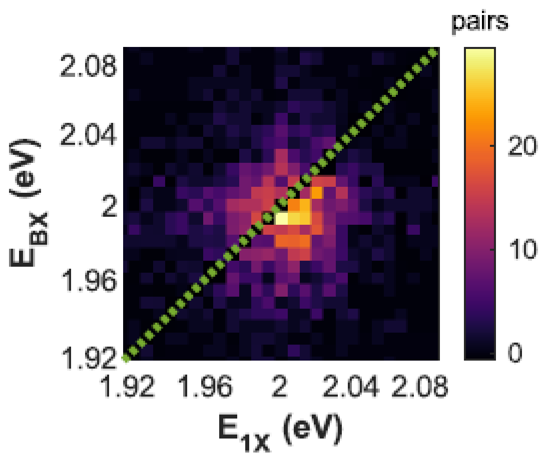
Single-photon time resolved spectrometer based on a 1D SPAD array:
~1ns time resolution, 2nm spectral resolution - simultenaously

G. Lubin, DO, et al., Nano Lett. 21, 6756 (2021)

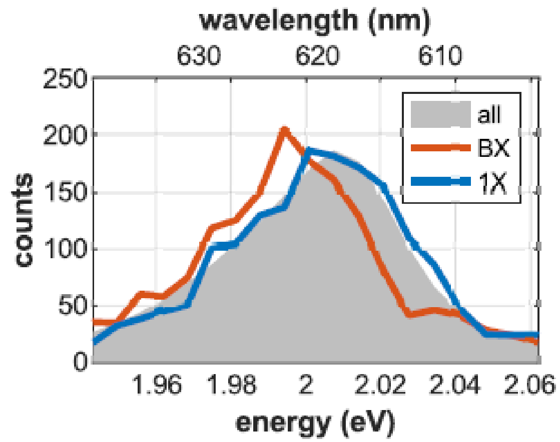
Heralded multiexciton spectroscopy

This enables to identify photon pairs emitted following a single excitation cycle and post-select only events involving a pair of photons (BX-X cascaded emission)

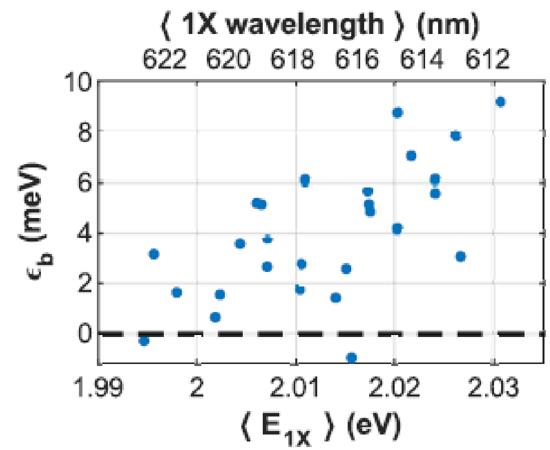
First photon



Second photon



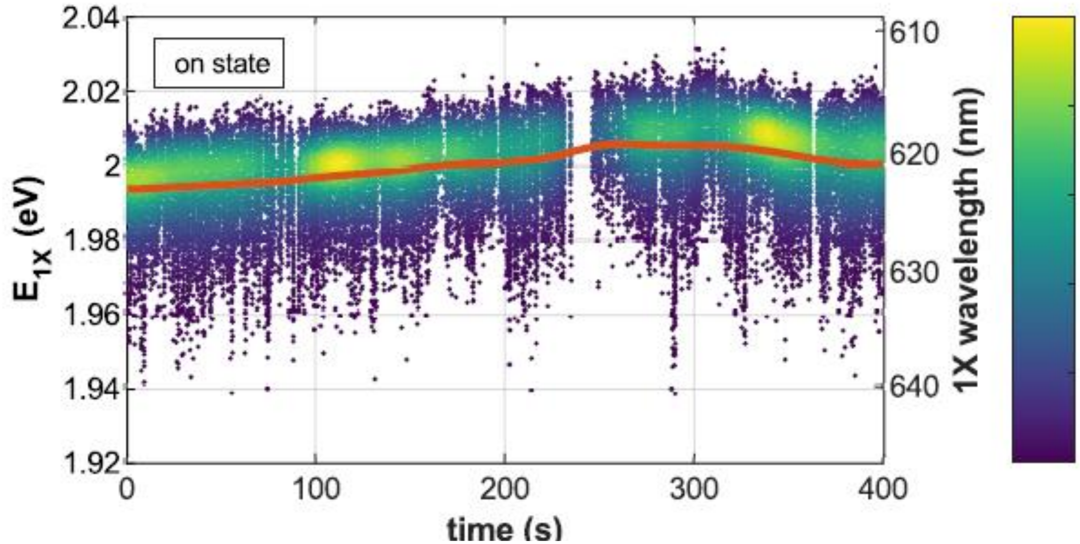
This reveals “hidden” inhomogeneous broadening



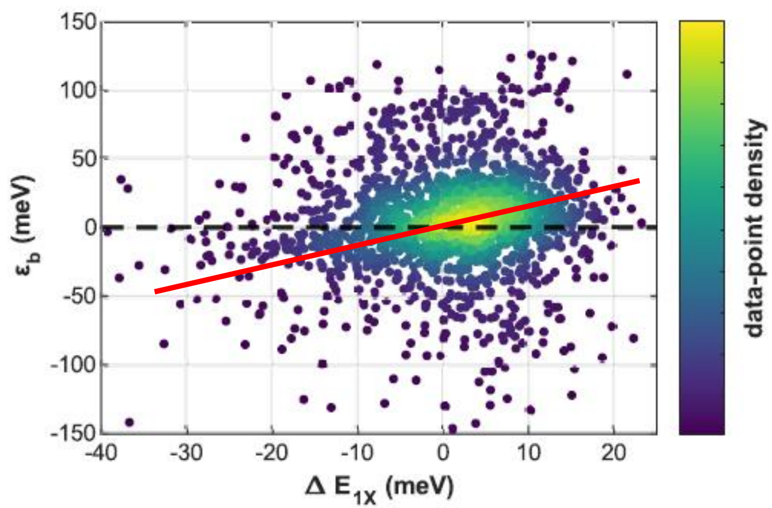
G. Lubin, DO, et al., Nano Lett. 21, 6756 (2021)

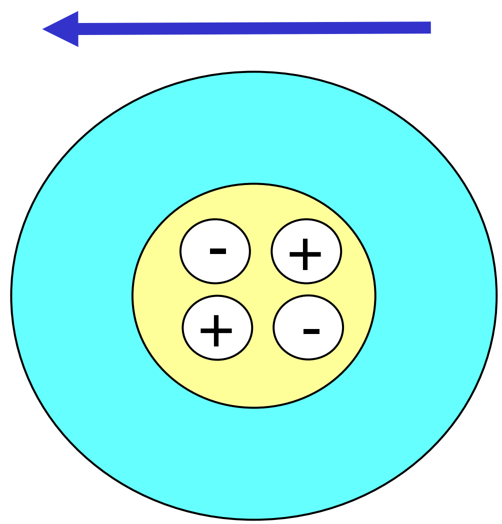
Heralded multiexciton spectroscopy

And study correlations at the single particle level, for example between spectral diffusion of BX and X transitions



BX less bound when X emission is redder (stronger stray field)





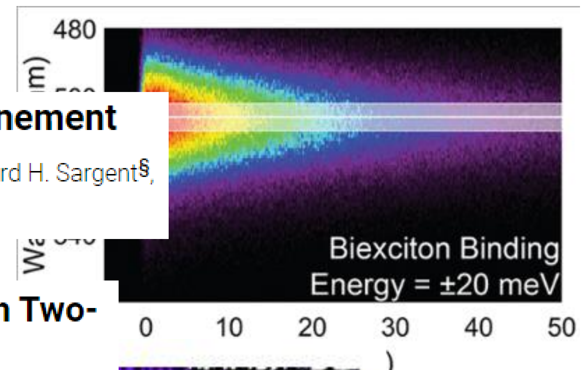
G. Lubin, DO, et al., Nano Lett. 21, 6756 (2021)

An ‘easy’ solution to problems which are hard To solve on an ensemble level

What is the biexciton binding energy in a $CsPbBr_3$ perovskite nanocrystal?

Setting an Upper Bound to the Biexciton Binding Energy in $CsPbBr_3$ Perovskite Nanocrystals

Katherine E. Shulenberger, Matthew N. Ashner, Seung Kyun Ha, Franziska Krieg, Maksym V. Kovalenko, William A. Tisdale*, and Moungi G. Bawendi*

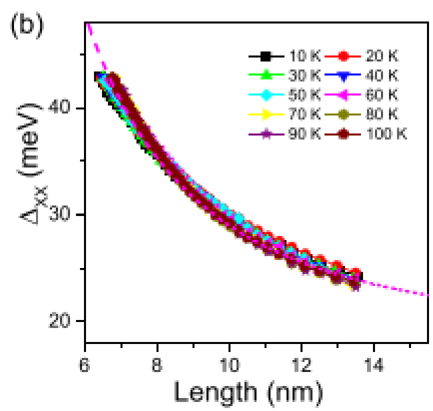


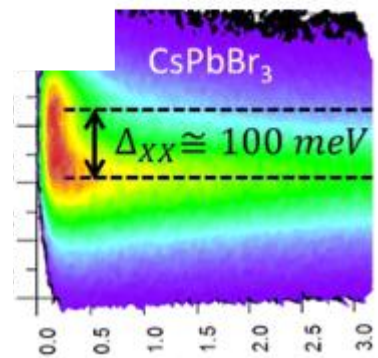
Efficient Biexciton Interaction in Perovskite Quantum Dots Under Weak and Strong Confinement

Juan A. Castañeda[†], Gabriel Nagamine[†], Emre Yassitepe[‡], Luiz G. Bonato[‡], Oleksandr Voznyy[§], Sjoerd Hoogland[§], Ana F. Nogueira[‡], Edward H. Sargent[§], Carlos H. Brito Cruz[†], and Lazaro A. Padilha[*†]

Inhomogeneous Biexciton Binding in Perovskite Semiconductor Nanocrystals Measured with Two-Dimensional Spectroscopy

Xinyu Huang, Lan Chen, Chunfeng Zhang*, Zhengyuan Qin, Buyang Yu, Xiaoyong Wang, and Min Xiao*

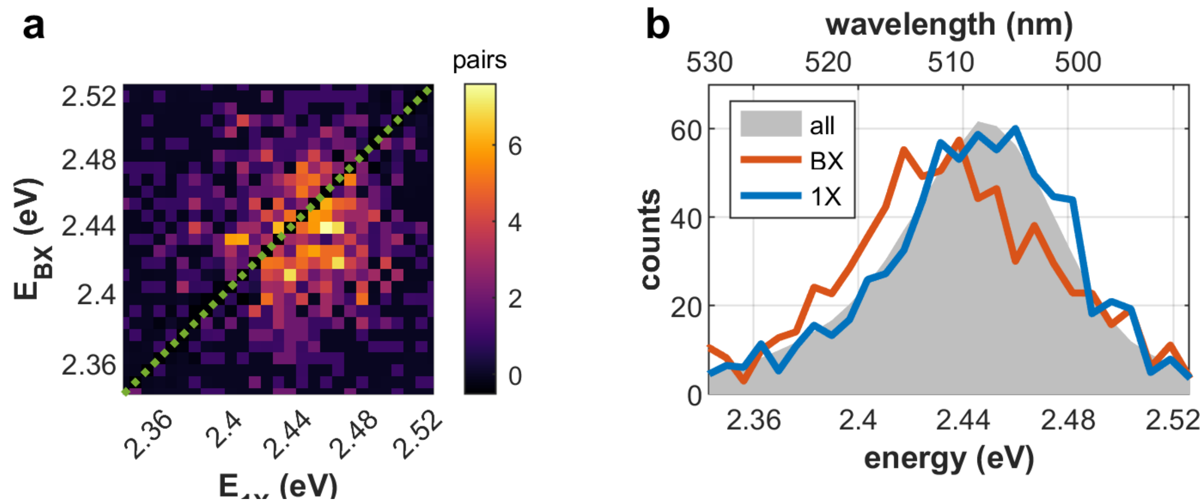




# of answers = # of methods used in the measurement …

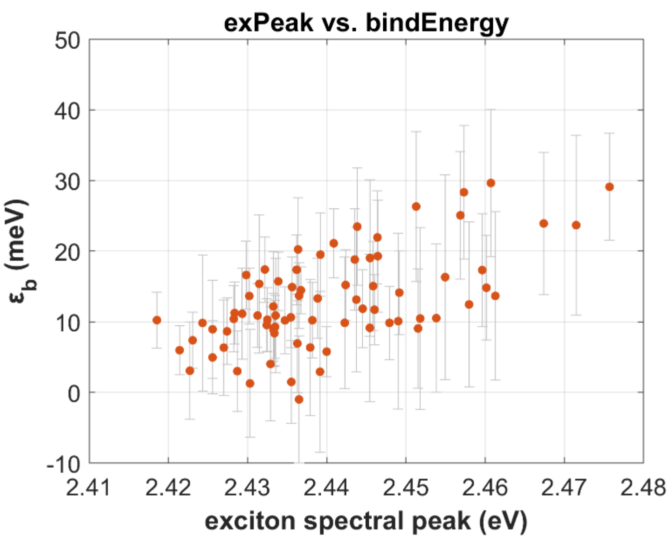
An ‘easy’ solution to problems which are hard To solve on an ensemble level

Heralded spectroscopy provides an unambiguous answer for every particle



$BX_{peak} \approx 2.4335\ eV$ $1X_{peak} \approx 2.4491\ eV$

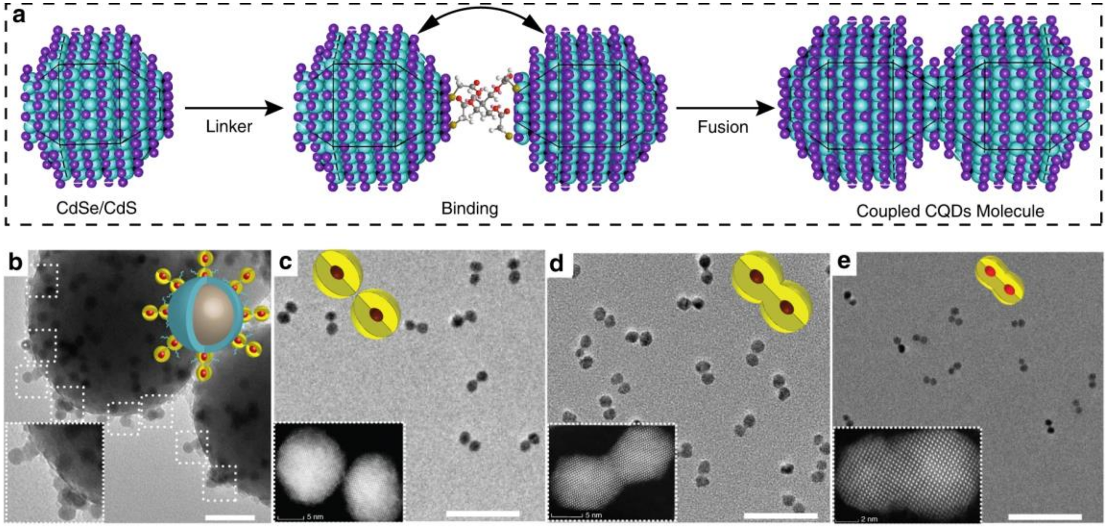
$$\varepsilon_b = 15.6 \pm 3.6\ meV$$



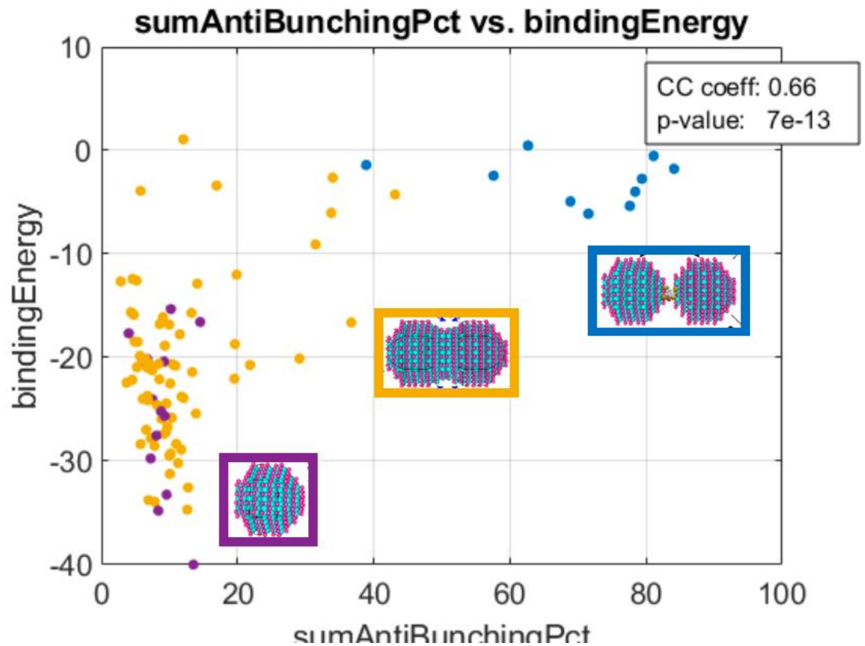
As well as correlations with other parameters (lifetime, $g^{(2)}$), providing a simple and comprehensive understanding

G. Lubin, DO, et al., submitted (2021)

Teaser: Quantum dot molecules



Towards being able to classify and identify 'complex' emitters via spatiotemporal photon statistics



Cui et al., Nature Commun. 10, 5401 (2019) ; Work in progress … (2022)

Conclusions

Photon correlations are ubiquitous and are becoming not so hard to measure

They often contain information which is hard or impossible to obtain by other means

Advances in detector technology (especially CMOS-compatible SPAD arrays) will make this a simple and cheap tool to use, even in “standard” tools such as spectrometers (or cameras).

Acknowledgements

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