Resonance Fluorescence as a Highly Sensitive Probe of Spectral Diffusion in Nanowire Quantum Dots

Toby E. Rawlings [1]*, Philip J. Poole [2], Dan Dalacu [2], Alistair J. Brash [1]

[1]School of Mathematical and Physical Sciences, University of Sheffield, Sheffield S37RH, United Kingdom
[2]National Research Council Canada, Ottawa K1A OR6 Ontario, Canada

Semiconductor quantum dots (QDs) have emerged as a leading platform for the development of quantum light sources due to their exemplary brightness, purity and indistinguishability. However, due to their solid-state nature, they are subject to environmental interactions which can cause dephasing. One such interaction is charge noise, which results from the probabilistic occupation of nearby charge traps. This creates a fluctuating local electric field which shifts the QD resonance and causes spectral diffusion. Efficient and accurate characterisation of these noise processes is necessary to optimise device performance for the demands of optical quantum technologies. Also essential to the performance of QD light sources is resonance fluorescence (RF) - an operating scheme in which the excitation laser energy is in resonance with an s-shell QD exciton state. RF has been extensively performed in epitaxial QDs but remains challenging in nanowire QDs due to scattering from the sub-diffraction limit nanowire tip.

Here, we present RF of nanowire QDs [1] with an excellent signal to background ratio, enabling clear observation of coherent dynamics such as Rabi oscillations in the second-order correlation function (Fig. 1(a)). We then apply this method to perform a detailed study of spectral diffusion resulting from charge noise. By performing second-order correlation measurements on the fluorescence, we resolve noise dynamics with far greater time-resolution than comparable first-order methods [2]. Furthermore, the nature of resonant excitation provides a spectral sensitivity superior to that of second-order methods using spectral filtering [3]. The second-order correlation function is fitted with a model that accounts for the coherent dynamics of RF – antibunching and Rabi oscillations – with the product of N exponential terms to account for bunching due to spectral diffusion (solid lines in Fig. 1(c)). This allows us to study the timescales involved down to nanosecond resolution.

Finally, the influence of weak above-bandgap excitation has been demonstrated to reduce spectral diffusion, as shown by the narrowing linewidths in Fig. 1(b). This can be attributed to a reduction in charge noise as the above-bandgap excitation populates charge traps. The second-order correlation function is also correspondingly modified, as shown in Fig. 1(c), with the bunching amplitude decreasing, and the faster noise processes being eliminated.

We believe that our results present an important new insight into the noise dynamics of nanowire QDs, with resonant excitation and charge noise suppression marking important steps to unlocking their potential. Our methods could also readily be applied to study noise processes in many other types of solid-state quantum emitter with high sensitivity and time-resolution.

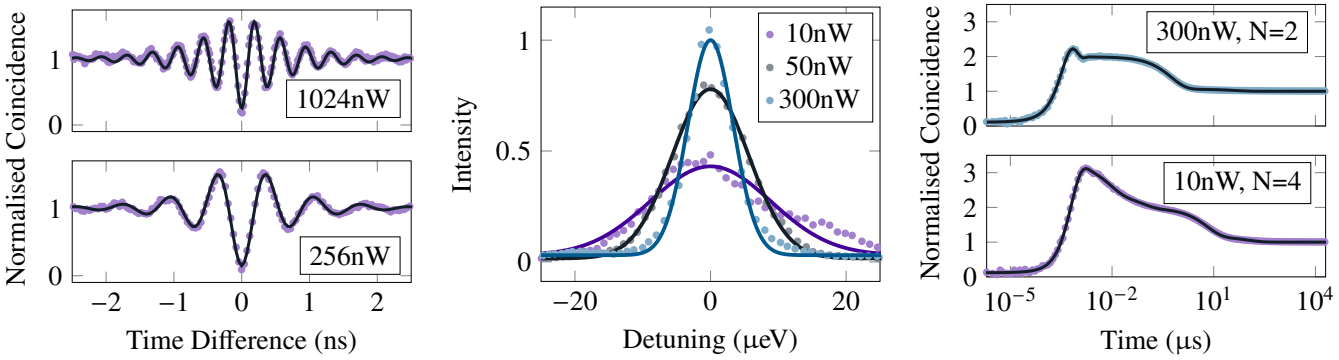

Figure 1: a) Second-order correlation function under different resonant powers, showing Rabi oscillations with different frequencies. b) Resonance fluorescence linewidths with varying above-bandgap excitation powers. c) Second-order correlation function measured up to 10ms with different above-bandgap excitation powers

References

[1] S. Haffouz et al., Nano Letters **18**, 3047–3052 (2018).

[2] C. Matthiesen et al., Scientific Reports **4**, 4911 (2014).

[3] G. Sallen et al., Nature Photonics **4**, 696–699 (2010).

*E-mail: terawlings1@sheffield.ac.uk