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RESEARCH ARTICLE

Single-dot spectroscopy uncovers the effects of surface trapping and Auger recombination on quantum dot emission

Bin Li[1], Xiaopeng Chen[1], Yuke Gao[1], Guofa Qu[1], Ruixiang Wu[1], Shengzhi Wang[1], Guofeng Zhang[2,†], Xiangyang Miao[1,‡]

[1] College of Physics and Electronic Engineering, Shanxi Normal University, Taiyuan 030031, China

[2] State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan 030006, China

Corresponding authors. E-mail: [†]guofeng.zhang@sxu.edu.cn, [‡]xymiao@sxnu.edu.cn

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Supporting Information

Supplementary Note 1

The explanation for the discrepancy between the experimental radiative lifetime scaling factor (1.6) and the theoretical value (2).

In an idealized scenario, the negative trion (X^-) consists of two electrons and one hole. Theoretically, both electrons can recombine with the single hole. This would result in a radiative decay rate twice that of the neutral exciton (X^0), which has only one electron-hole pair. Therefore, the expected scaling factor is 2. The deviation arises from the reduced spatial overlap of the electron and hole wave functions in the negative trion state (X^-) due to altered Coulomb interactions [1]. In the neutral exciton (X^0) state, the single electron and hole are strongly attracted to each other. This attraction confines both carriers to a small volume near the center of the QD, maximizing their wavefunction overlap, as illustrated in Fig. 3(a). In the negative trion (X^-) state, an additional electron is introduced. The two electrons in the conduction band experience strong repulsion due to Coulomb's law. This forces them to occupy a larger spatial volume to minimize their energy. Consequently, the electron wavefunction becomes significantly delocalized across the entire potential well, as depicted in Fig. 3(c). This delocalization reduces the spatial overlap of the electron wavefunction with the more localized hole wavefunction. Since the radiative recombination rate is proportional to the square of the electron-hole wavefunction overlap integral, the reduced overlap results in a slower recombination rate than in the ideal case.

Thus, while the presence of a second electron creates an additional recombination channel, increasing the factor above 1, the concurrent reduction in wavefunction overlap due to Coulomb repulsion offsets this effect. The net result is an experimental scaling factor of 1.6, which is less than the theoretical maximum of 2.

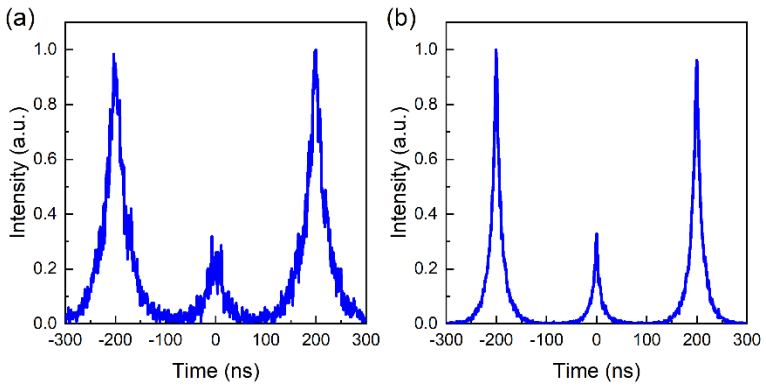


Fig. S1 (a) Second-order correlation measurements for the same single QD presented in Fig. 1. **(b)** Second-order correlation measurements for the same single QD presented in Fig. 2. The pronounced antibunching dip at zero-time delay ($g^{(2)}(0) < 0.5$) confirms that these are single QDs.

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References

1. Y. S. Park, W. K. Bae, J. M. Pietryga, and V. I. Klimov, Auger recombination of biexcitons and negative and positive trions in individual quantum dots, *ACS Nano* 8(7), 7288 (2014) https://doi.org/10.1021/nn5023473

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