Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary information for

Memories in the Photoluminescence Intermittency of Single Cesium Lead Bromide ($CsPbBr_3$) Nanocrystals (NCs)

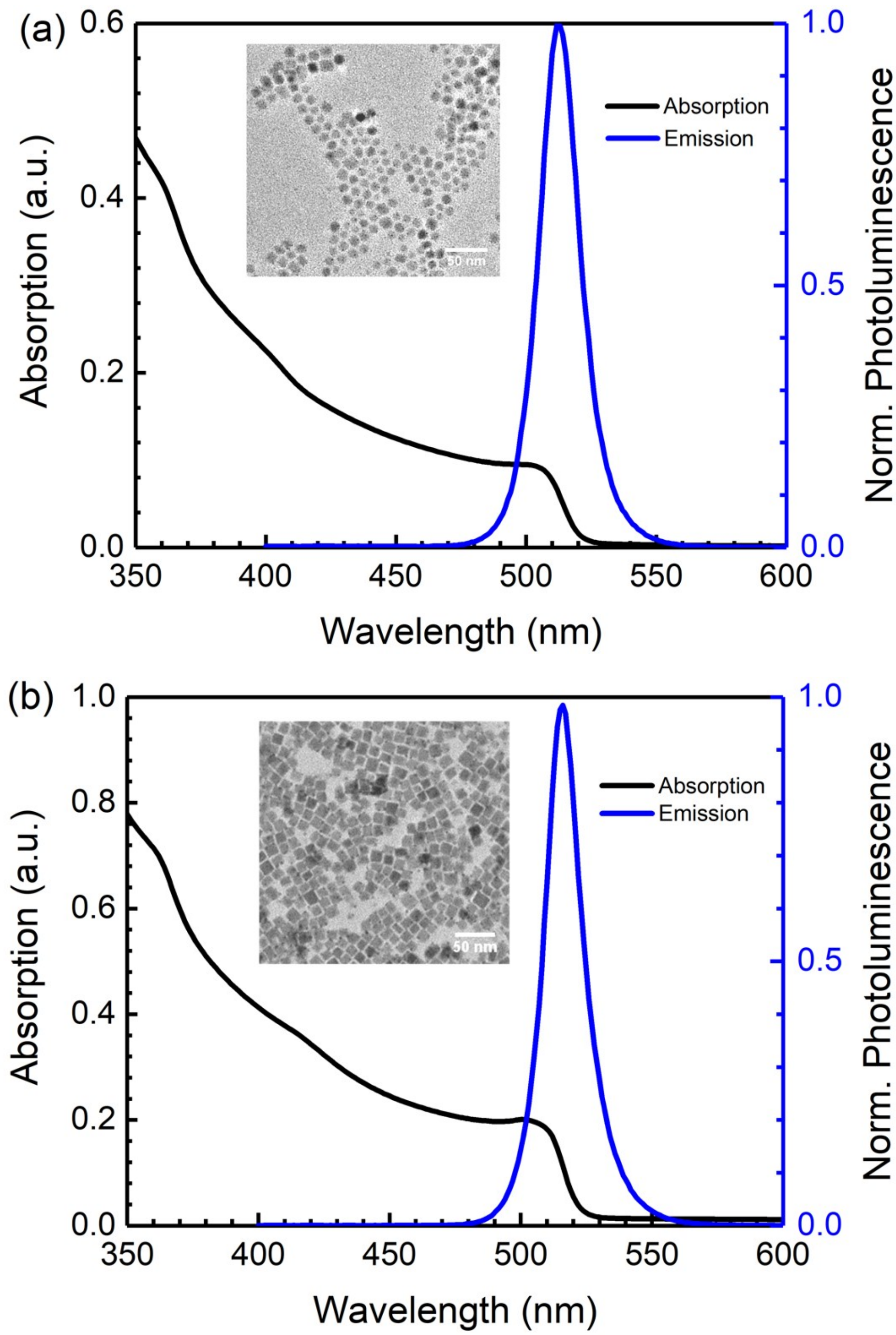


Figure S1: Absorption and PL spectra of $CsPbBr_3$ NCs colloids and TEM images. (a) Spectra of colloidal $CsPbBr_3$ NCs with OAm/OA capping ligands in toluene. The scale bar in the insert TEM image is 50 nm. The average size is 10.2 nm. (b) Spectra of colloidal $CsPbBr_3$ NCs with zwitterionic capping ligands in toluene. The average size measured over 76 NCs is 10 nm. The measured PL quantum yield of colloid (b) is about 0.75.

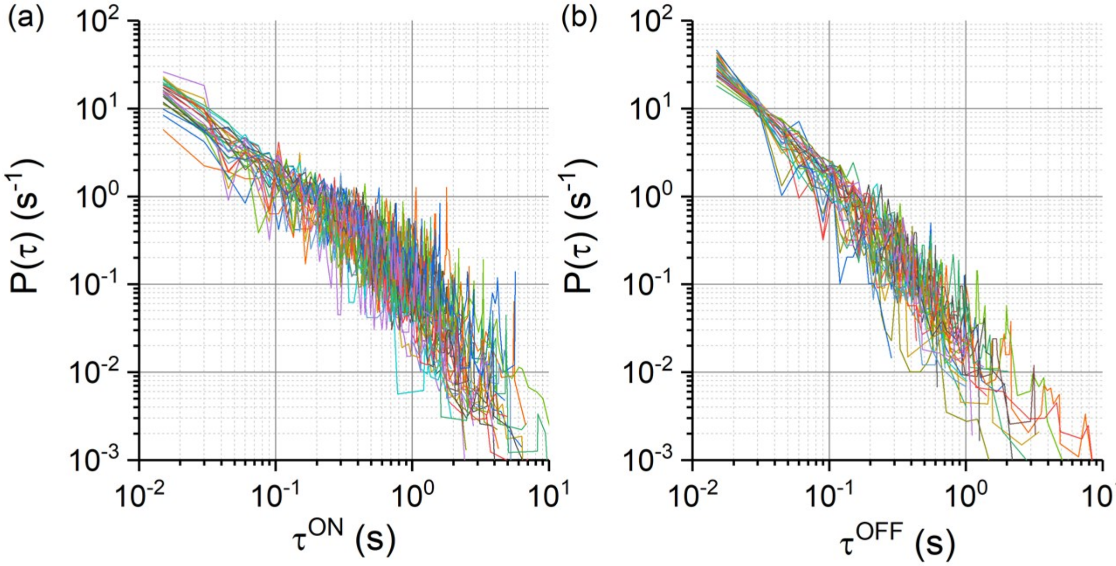


Figure S2: Dwelling time distributions of single CsPbBr3 NCs with OAm capping ligands in PMMA polymer. (a) ON and (b) OFF time distributions. Data of different NCs are indicated with different colors. The time bin is 15 ms.

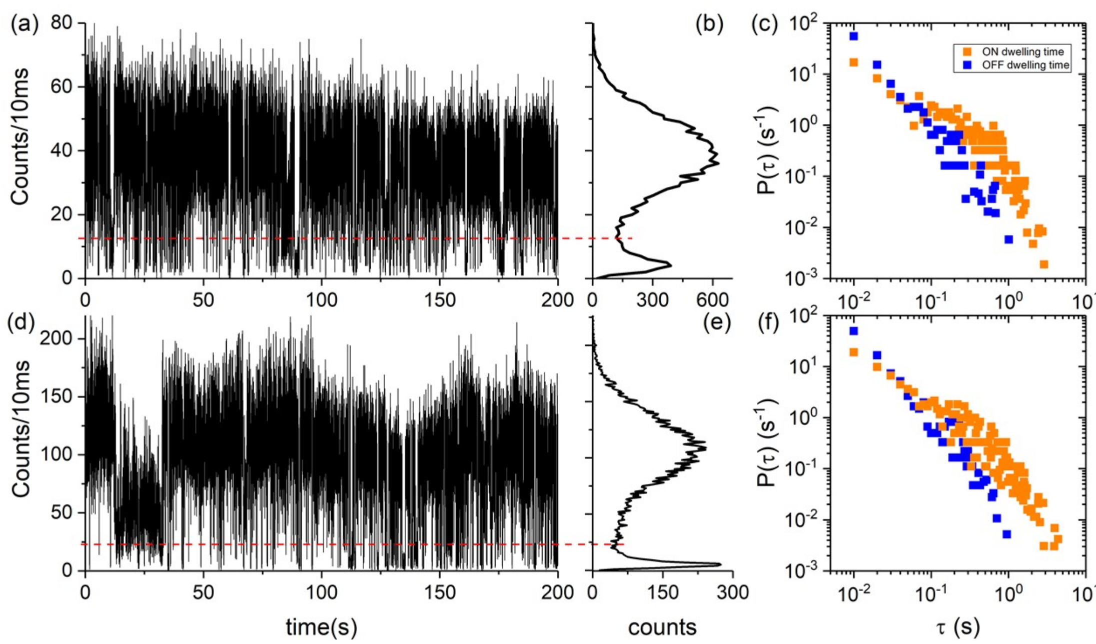


Figure S3: Comparison of PL intermittency under cw and pulsed excitations. (a-c) Results under pulsed excitation at 488 nm; (d-f) Results under cw excitation. The experiments were done on the same NC. The distributions of ON (in orange) and OFF (in blue) dwelling times in (c) and (f) are similar in both regimes of excitation.

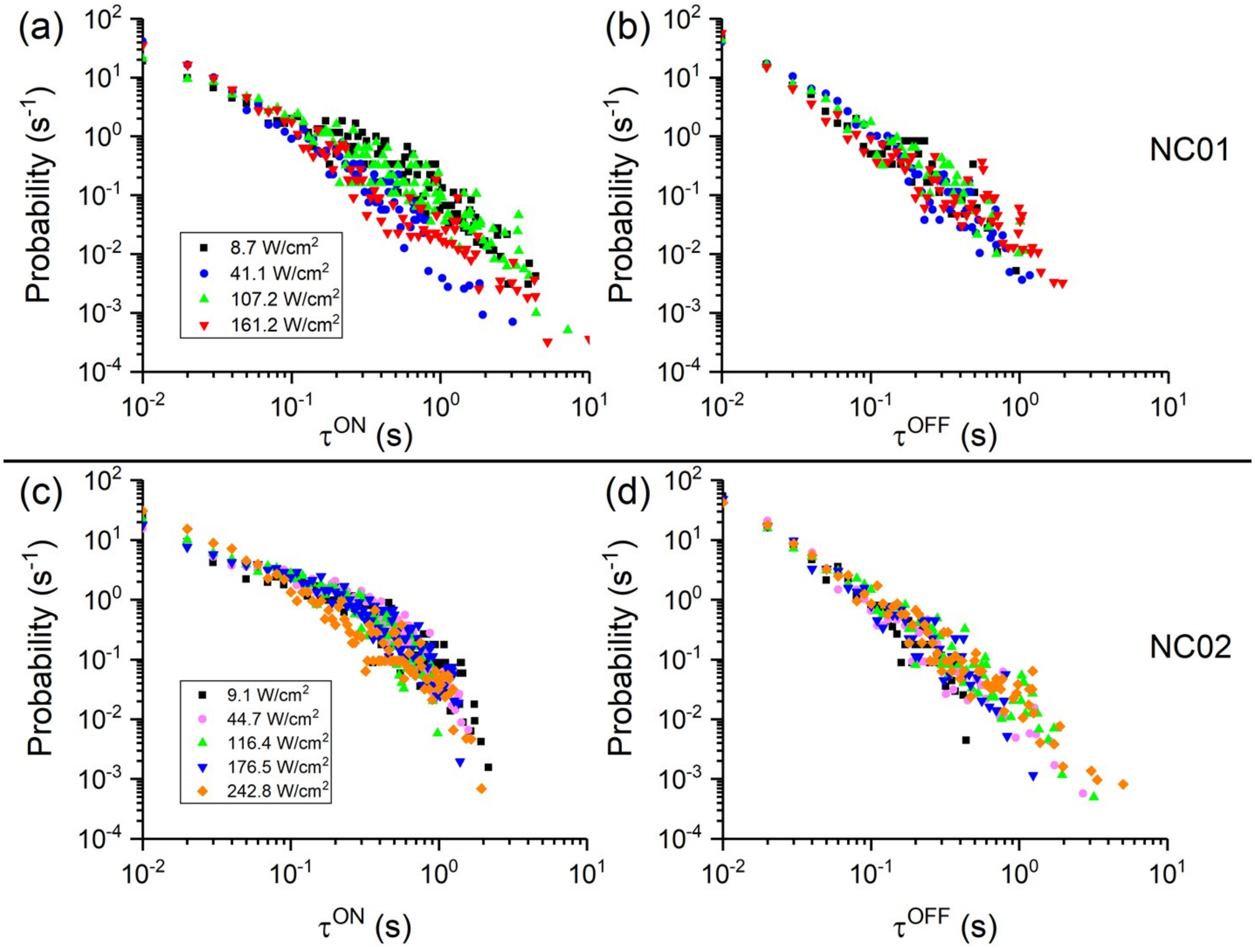


Figure S4: Excitation power dependence of ON and OFF dwelling time distributions. Data of two $CsPbBr_3$ NCs (NC01 and NC02) are shown. (a, c) ON time probability densities; (b, d) OFF time probability densities. The time bin is 10 ms for both NCs.

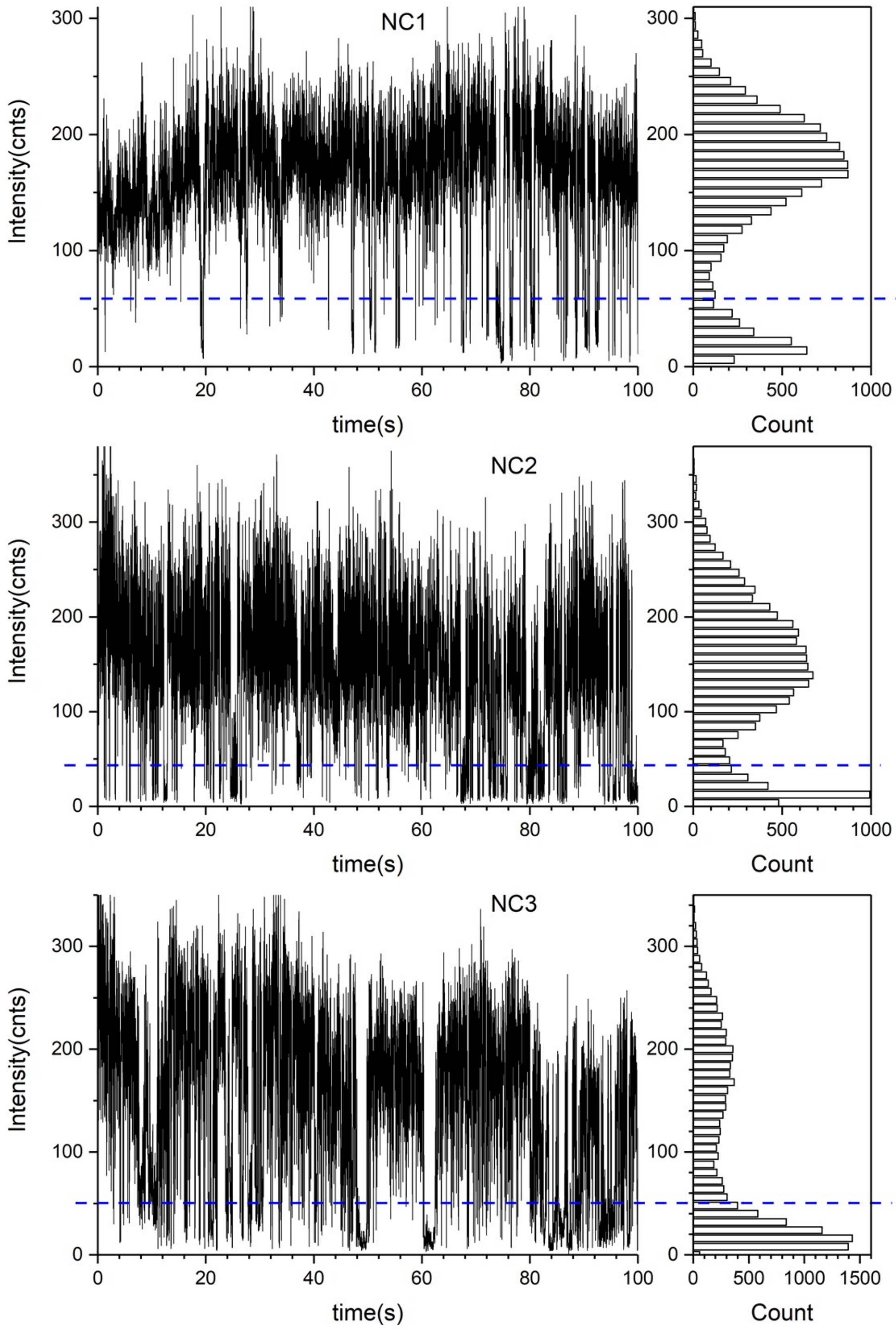


Figure S5: PL blinking traces and intensity histograms of three $CsPbBr_3$ NCs mentioned in the main text. The time bin is 15 ms for all the traces. The excitation power is about 8 W cm^{-2} at 488 nm.

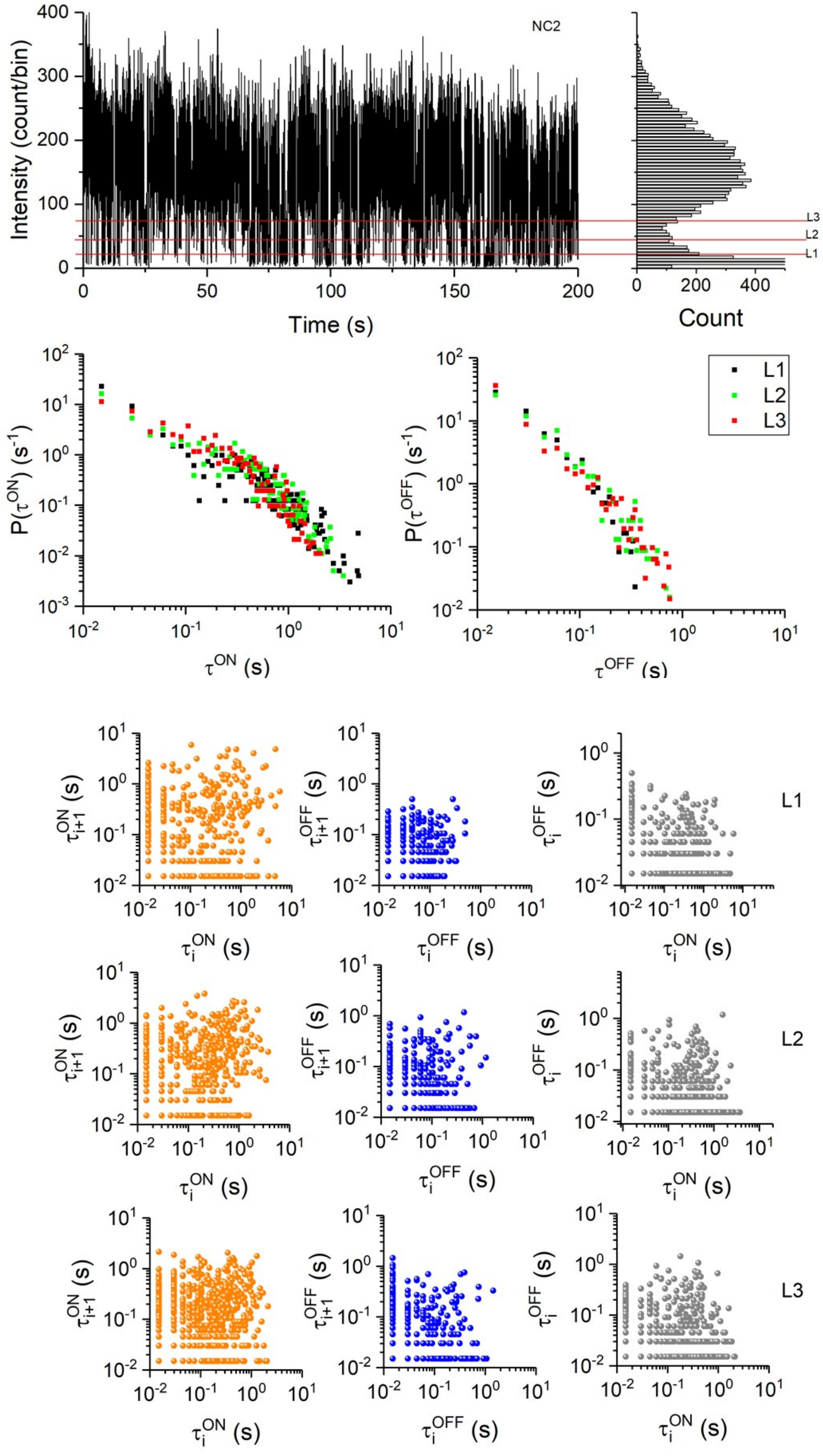


Figure S6: Influence of the intensity threshold on the dwelling time statistics and the correlation coefficient.

The choice of the intensity threshold in the blinking analysis impacts the values of the extracted correlation coefficients [1], but does not influence the distributions of the ON- and OFF-times (Fig.S6 (b)) as well as the overall trends of the observed correlations (Fig.S6 (c)). We checked the influence of the threshold on the dwelling time distribution and the correlation coefficients by performing the analysis for three threshold values named L1, L2 and L3 (see red lines in Fig.S6 (a)). L1 and L3 are the extreme acceptable levels for the threshold given the noise of the time trace. The correlation coefficients and the scatter plots between successive dwelling times are shown in Table S1 and Fig.S6 (c).

Threshold level	L1	L2	L3
$R_{log}^{ON\text{-}next\text{-}ON}$	0.302	0.37	0.197
$R_{log}^{OFF\text{-}next\text{-}OFF}$	0.282	0.14	0.112
$R_{log}^{ON\text{-}next\text{-}OFF}$	-0.11	-0.1	-0.069
$R^{ON\text{-}next\text{-}ON}$	0.22	0.19	0.11
$R^{OFF\text{-}next\text{-}OFF}$	0.285	0.19	0.089
$R^{ON\text{-}next\text{-}OFF}$	-0.078	-0.02	-0.018

Table S1: Correlation coefficients when different intensity thresholds are chosen.

With the increase of threshold intensity, more ON periods are inappropriately attributed to the OFF periods. This results in the decrease of correlation coefficients, as ON and OFF periods are not positively correlated. Considering the low percentage of events in the overlapping region and the small value of correlation coefficients, we conclude that the influence of threshold intensity on the correlation is not significant, as long as it is chosen close to the minimum between the intensity distribution peaks.

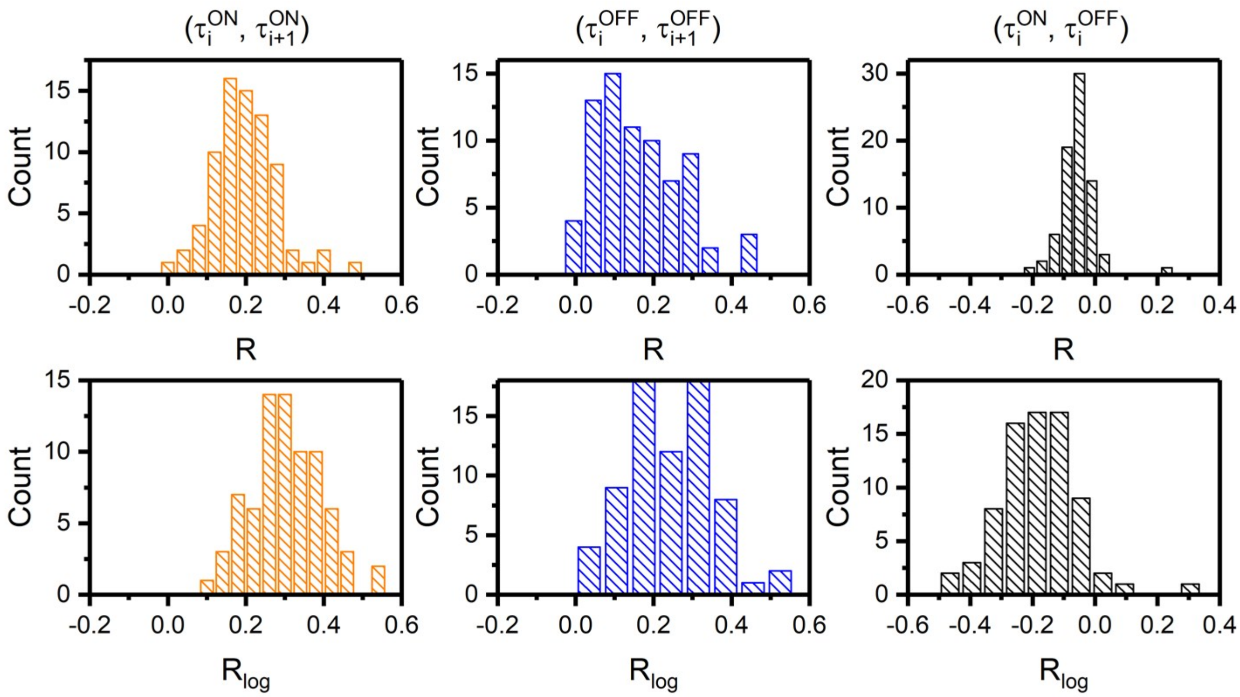


Figure S7: Histograms of correlation coefficients R and R_{log} for successive dwelling times. Successive ON times (orange), successive OFF times (blue) and subsequent ON-OFF times (black). The histograms are built with 76 NCs' data. The distribution in R and Rlog values reflects the inhomogeneity in NCs and/or in the local environments.

In the histograms shown here and in the main text, there are rare cases where positive τ_n^{ON} versus τ_n^{OFF} correlation or negative τ_n^{OFF} versus τ_{n+1}^{OFF} correlation were observed, which is most likely caused by the limitation of the threshold method and the overlapping region in the PL intensity histogram. The accuracy of such correlation analysis indeed depends on the threshold level discussed above to discriminate the ON and OFF states as well as the binning time of the blinking trace (15 ms in our analysis) [1,2]. In order to minimize such influences, we choose blinking traces presenting two well-defined peaks in the intensity histogram. Stefani et.al have examined these influences and concluded that as long as the maximal Poisson-distributed OFF intensity can be identified, the influence of threshold on the correlation coefficients is negligible [3].

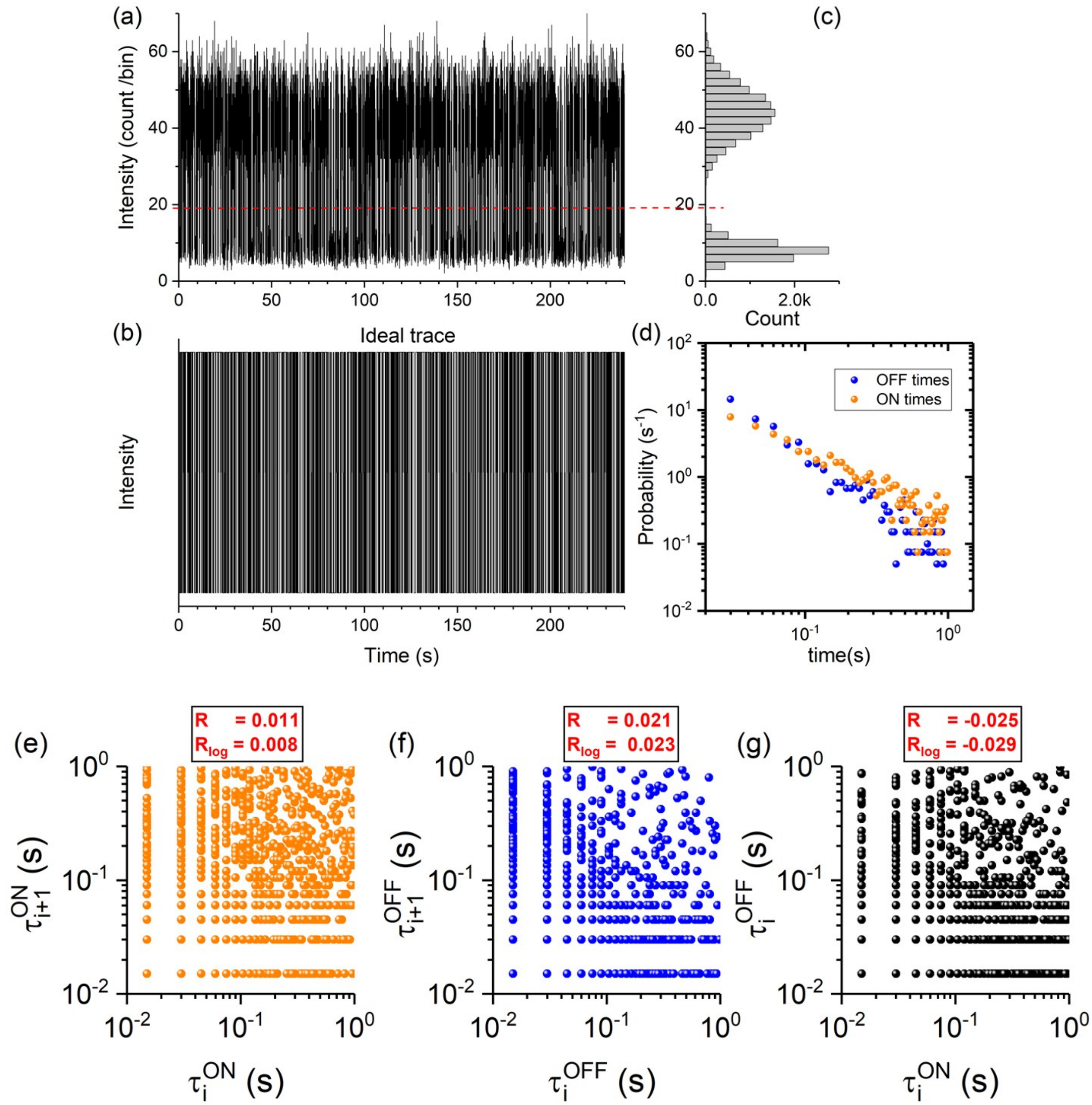
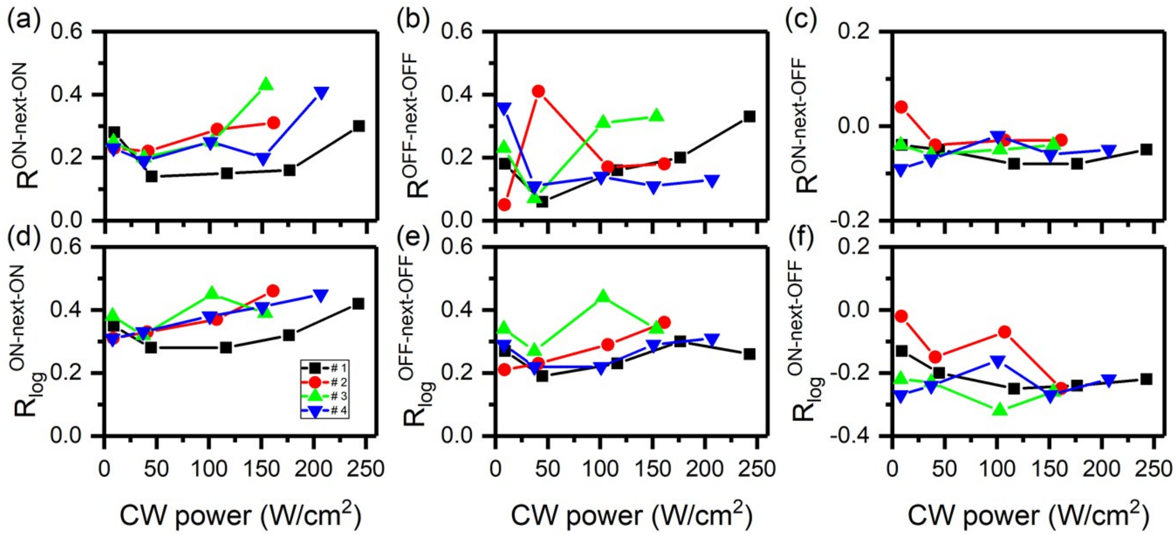


Figure S8: Blinking simulations with a Monte Carlo method. (a) Simulated time trace, together with the intensity histogram (c). (b) Ideal time trace composed of randomly generated time intervals which follow inverse power-law statistics with power components of -1.09 (ON) and -1.47 (OFF) respectively. The reason of choosing these values is based on the average power-law components of 29 NCs. (d) The ON (in orange) and OFF (in blue) time probability densities when the threshold to distinguish ON and OFF states is chosen at the red dashed line. (e, f, g) scatter plots of successive ON times (in orange), successive OFF times (in blue) and subsequent ON-OFF times (in black). The calculated correlation coefficient R and R_{log} values are shown on the top of each plot and have vanishing values compared with those of NCs.



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gure S9: Excitation power dependence of the correlation coefficients R and R_{log} for four $CsPbBr_3$ NCs embedded in PMMA polymer. Each color represents a single NC. (a, b, c) R values; (d, e, f) R_{log} values.

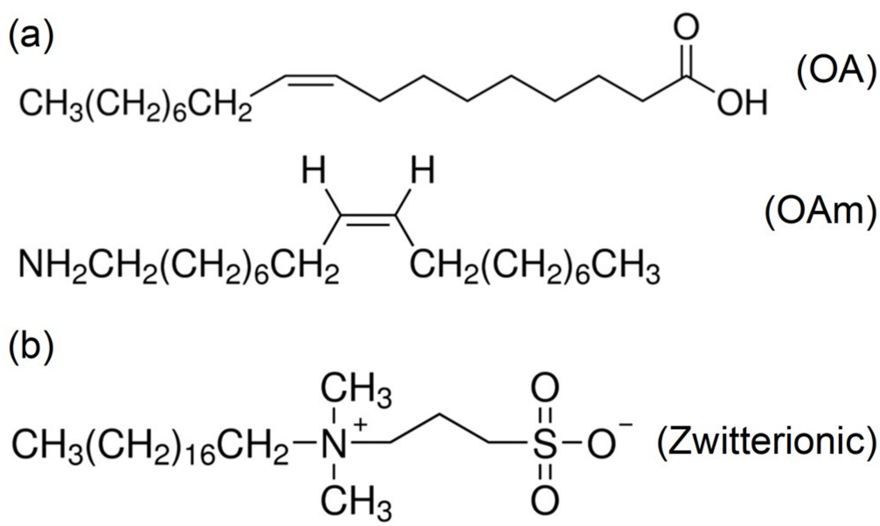

Figure S10: Chemical structures of the two surface capping ligands used in our studies. (a) Oleic acid (OA) and oleylamine (OAm) ligands. In the precursor for the synthesis of $CsPbBr_3$ colloid, OA is the anion and OAm is the cation. The OAm will bond to the bromide-rich surface and serve as passivation layers for the NCs. (b) Zwitterionic molecule.

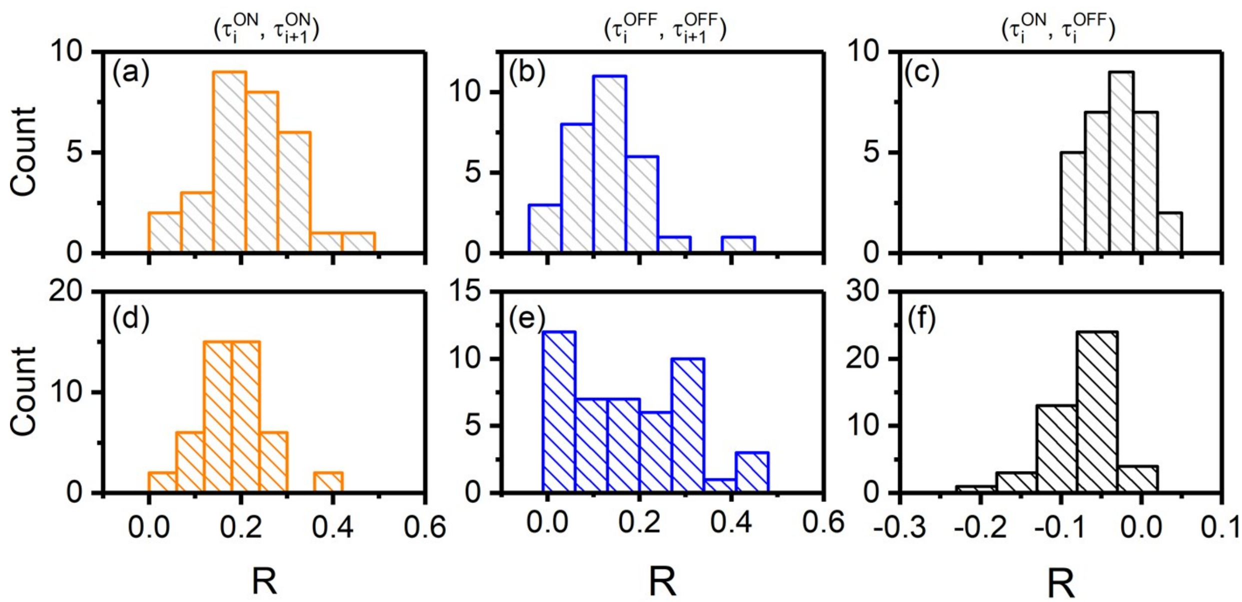

Figure S11: Histograms of R-values of subsequent periods for different ligands capping the NCs. (a, b, c) R values for NCs with OAm ligands; (d, e, f) R values for NCs with zwitterionic capping ligands. (a, d), (b, e), (c, f) were calculated with successive ON times, successive OFF times and subsequent ON-OFF times, respectively.

	SEBS	PS	PMMA	PVA
ε_r	2.2	2.53	3.4	14

S12: Table of the relative dielectric permittivity of different polymers from ref. [4]. These are the values at low frequency about 1 kHz.

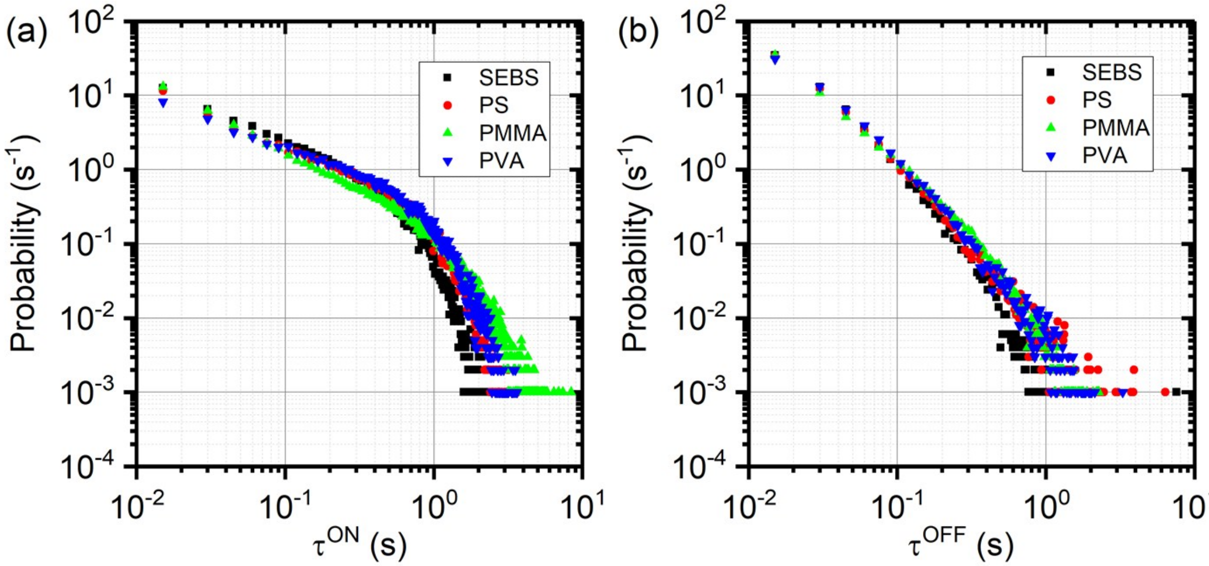

Figure S13: Dwelling time distribution for NCs embedded in different polymers. (a) ON time distribution; (b) OFF time distribution. The points are built from average results over a hundred single $CsPbBr_3$ NCs with a size of ~ 6 nm.

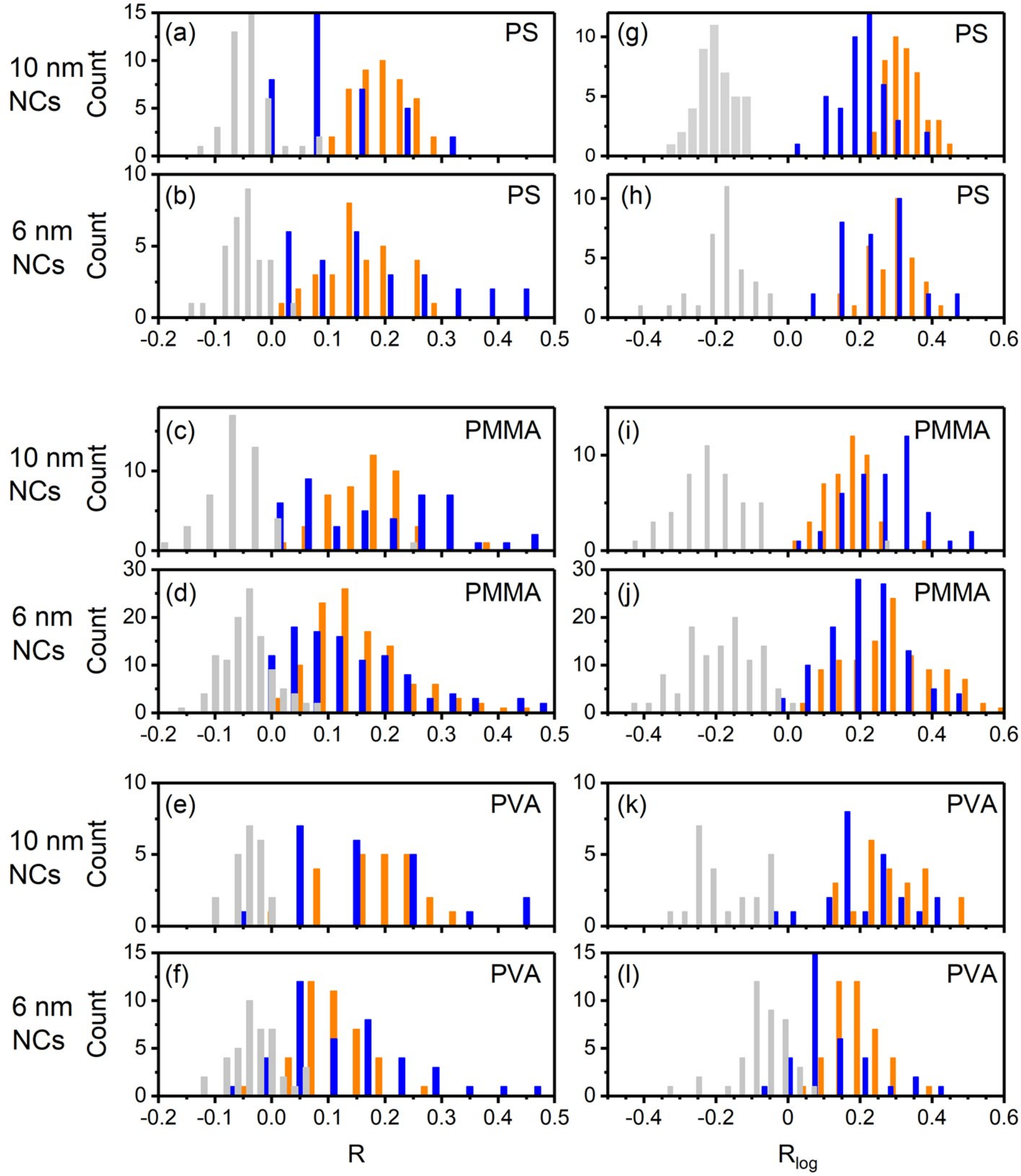

Figure S14: Correlation coefficient histograms for $CsPbBr_3$ NCs of various sizes in various polymers. The average NC sizes are indicated on the left of the figures. (a-f) R values in three polymer matrixes; (g-l) R_{log} values.

References:

1 C. H. Crouch, O. Sauter, X. Wu, R. Purcell, C. Querner, M. Drndic, M. Pelton, Facts and Artifacts in the Blinking Statistics of Semiconductor Nanocrystals. *Nano Lett.* 2010, **10**, 1692.

2 P. A. Frantsuzov, S. Volkán-Kacsó, B. Jankó, Model of Fluorescence Intermittency of Single Colloidal Semiconductor Quantum Dots Using Multiple Recombination Centers. *Phys. Rev. Lett.* 2009, **103**, 207402.

3 F. D. Stefani, X. Zhong, W. Knoll, M. Han, M. Kreiter, Memory in quantum-dot photoluminescence blinking. *New J. Phys.* 2005, **7**, 197.

4 A. Issac, C. von Borczyskowski, F. Cichos, Correlation between photoluminescence intermittency of CdSe quantum dots and self-trapped states in dielectric media. *Phys. Rev. B* 2005, **71**, 161302.