Suppression of Blinking and Enhanced Exciton Emission from Individual Carbon Nanotubes

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ABSTRACT Blinking and spectral diffusion are hallmarks of nanoscale light emitters and a challenge for creating stable fluorescent biomarkers or efficient nonclassical light sources. Here, we demonstrate suppression of blinking and spectral diffusion of individual single-wall carbon nanotubes by manipulation of their dielectric environment, resulting in 5-fold enhanced light emission. In addition, it was found that the characteristic slopes of the blinking power laws are largely independent of the dielectric environment in the limit of a large number of switching events. In contrast, the on/off ratio determined from statistical occurrence analysis is found to be improved by 3 orders of magnitude toward the on state, making the on/off ratio an important measure for charge transfer from/into the local dielectric environment of a quantum emitter. Furthermore, our approach is compatible with integration into cavities, in contrast to previous demonstrations of spectral diffusion suppression achieved in free-standing single-wall carbon nanotubes. This opens up possibilities to couple the exciton emission of nonblinking carbon nanotubes to cavity modes to further benefit by the Purcell effect and to enhance the light extraction efficiency, in order to ultimately demonstrate efficient photonic devices.

KEYWORDS: single-wall carbon nanotubes · excitons · fluorescence intermittency · spectral diffusion · power law

Single-walled carbon nanotubes (SWCNTs) have recently gained particular interest as quantum emitters for photonic and optoelectronic devices because the optical emission in these quasi one-dimensional systems arises from excitons with a strongly enhanced Coulomb interaction, leading to very large exciton binding energies of 0.2−0.4 eV stable up to room temperature.[1] Electroluminescence of excitons from individual SWCNTs was demonstrated at room temperature,[2] and applications as photodetectors or light-emitting diodes thus seem feasible.[3] Recently, Högele *et al*. demonstrated nonclassical light emission (photon antibunching) from a (6,5) chirality SWCNT at cryogenic temperature,[4] demonstrating the potential to create single photon sources,[5] in particular if SWCNTs are chosen with an emission wavelength matching the optical communication window and if they are embedded into cavities to efficiently extract the exciton emission.[6]

While efficient photonic devices require stable long-term emission, almost all quantum emitters such as single molecules,[7] semiconductor quantum dots,[8] or SWCNTs[9−11] exhibit the phenomenon of fluorescence intermittency (blinking).[12−14] As a result of intermittency, the sample emission randomly switches on and off under continuous excitation, with blinking time scales ranging from nanoseconds[15] to more than a minute.[14] The blinking periods with no emission, or "dark" states, complicate the retrieval of information provided by fluorescent markers and degrade sample emission performance over time.[13] Many efforts were made to study the cause of intermittency and explain the characteristic power laws of on and off times, which display a ubiquitous exponent of −1.5, with several proposed mechanisms that are still debated.[14] Intermittency is often accompanied by spectral diffusion, which is caused by fluctuations of local surface charges in the dielectric environment of a quantum emitter, resulting in a red shift in the emission energy proportional to the square of the electric field arising from the quantum confined Stark effect.[16,17]

Clearly, to create efficient quantum light sources based on SWCNTs, blinking and spectral diffusion must be controlled or suppressed altogether. To this end, Kiowski *et al*. demonstrated that spectral diffusion is a property of the surfactant wrapping the SWCNT, rather than intrinsic to SWCNTs, and that free-standing SWCNTs without a surfactant display drastically reduced spectral diffusion.[18] However, devices with free-standing SWCNTs are brittle and integration of air-bridged SWCNTs with cavities to enhance the light extraction seems quite challenging. Recently, it was also shown that blinking was stopped in nanodiamonds by

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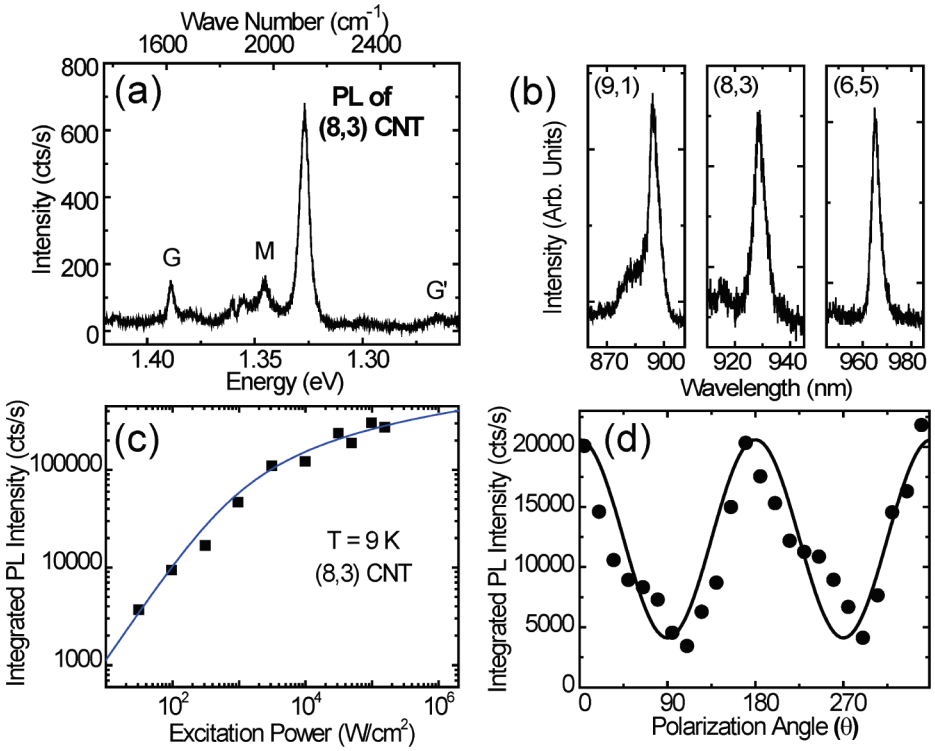

Figure 1. (a) μ-PL spectrum of an individual (8,3) SWCNT excited (nonresonantly) at 780 nm. (b) Examples for various SWCNT chirality types present in sample solution. (c) Integrated PL intensity from an (8,3) CNT; the solid line is a guide to the eye. (d) Sinusoidal polarization dependence of excitation on PL intensity. Data are recorded at 7 K.

embedding the sample in a polyvinyl film[19] as well as in ternary core−shell quantum dots.[20]

Here, we report an experimental demonstration of strong suppression of blinking and spectral diffusion of individual SWCNTs, which was achieved by capping them with poly(methyl methacrylate) (PMMA) and subjecting samples to a baking procedure. As a result, 5-fold enhanced exciton emission was observed. Furthermore, it was found that the characteristic slopes of the blinking power laws are largely independent of the dielectric environment in the limit of a large number of switching events. In contrast, the on/off ratio determined from statistical occurrence analysis is found to be 3 orders of magnitude more sensitive, making the on/off ratio an important measure for charge transfer from/into the local dielectric environment of a quantum emitter.

RESULTS AND DISCUSSION

In order to study light emission from individual semiconducting SWCNTs, chirality purified (6,5) SWCNTs grown by the CoMoCat technique were dissolved in sodium dodecylsulfate (SDS) solution and deposited onto a 90 nm SiO_2 substrate, resulting in a tube density of less than one per square micrometer after dry out. Figure 1 shows the results of a microphotoluminescence (μ-PL) study carried out at 7 K base temperature under laser excitation at 780 nm with a CW laser diode. Figure 1a shows a typical plot of a μ-PL spectrum of an individual SWCNT, which displays a dominant PL emission at 1.375 eV and several Raman sidebands. The excitation at 780 nm is nonresonant with respect to the E_{22} transition or phonon (Raman) sidebands.[21] When scanning the relative position between sample and the laser, it is found that the emission is strongly localized; that is, emission signatures are more than 20 μm apart, indicative of emission from individual CNTs. Although the SWCNTs have been purified to yield predominantly tubes with (6,5) chirality, the spectra in Figure 1b taken across the wafer also display optical signatures of SWCNTs with (9,1) and (8,3) chiralities, which were assigned following refs 22−24. The spectral line widths of 10 different SWCNTs were found to vary between 2.5 and 8.4 nm, some of them slightly asymmetric to longer wavelength. The integrated PL intensity as a function of pump power follows a linear increase in photo counts up to about 100 kcts/s at 50 μW excitation power, corresponding to 3 kW/cm^2, followed by the onset of a saturation behavior at higher pump powers (Figure 1c). This observation is similar to results from Matsuda *et al.*[9] and was attributed to efficient exciton−exciton annihilation processes on a 1 ps time scale acting as a nonradiative Auger recombination path in the presence of strong Coulomb correlations and more than one exciton.[25]

Another indication that the PL emission stems predominantly from an individual SWCNT is the

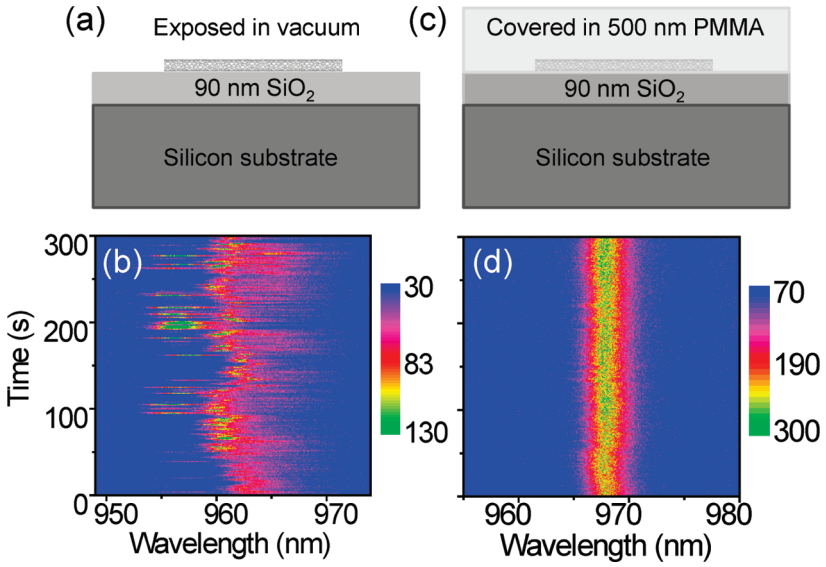

Figure 2. Schematic diagrams of an individual (6,5) SWCNT deposited on SiO_2 substrate without (a) and with (c) a PMMA capping layer. The PMMA-capped samples were furthermore subject to a baking procedure to remove trap states. The corresponding spectral trajectories of the PL emission are shown for SWCNTs without (b) and with (d) PMMA capping/baking. Data are recorded at 7 K. The intensity scale is counts per 200 ms recording time.

pronounced sinusoidal dependence between the polarization axis of the excitation laser and the integrated PL intensity, which is maximized when the electric field component of the laser lies along the tube axis, as shown in Figure 1d. The extracted polarization extinction ratio of about 7 falls within the range found previously for individual SWCNTs[26] and would be washed out if several CNTs contribute to the signal.

To study spectral diffusion, we investigated samples with individual SWCNTs dispersed directly onto SiO_2 without changing their dielectric environment, *i.e.*, directly exposed to the vacuum environment in the cryostat, as illustrated in Figure 2a. Spectral trajectories were obtained by continuously recording spectra every 200 ms. Figure 2b shows a typical example for SWCNTs displaying large variations in the spectral emission wavelength over time. In addition, as indicated by the intensity scale, the exciton emission displays strong intensity fluctuations, which are attributed to fluorescence intermittency, further indicating that emission in our samples stems from individual SWCNTs rather than bundles, since these quantum emitter effects are washed out in the ensemble emission.

In order to eliminate spectral diffusion, we followed the idea that the photo-created charges cannot tunnel in or out of the SWCNTs if charge trap states are removed from the local environment. As is well-known, CNTs integrated in field-effect transistors suffer from gate hysteresis effects due to local charge trapping by the SiO_2 surface-bound water, which is attached to the silane groups proximal to the nanotubes.[27−30] This gate hysteresis effect can be removed by elevating CNTs bridging an air gap,[28] similar to the elimination of spectral diffusion in free-standing CNTs.[18] However, since this approach is not compatible with integration of SWCNTs into cavities,[6] we instead capped the samples with 500 nm PMMA (see Figure 2c) followed by baking on a hot plate at 150 °C for several days in ambient air to remove surface-bound water molecules. Using the same technique, initially introduced by Kim *et al.*,[27] we have recently demonstrated that gate hysteresis can be fully eliminated in in-plane grown SWCNT field effect transistors.[30]

Figure 2d shows the corresponding spectral trajectory for a PMMA-covered/baked (6,5) SWCNT displaying remarkably less spectral diffusion and blinking. Spectral diffusion can be eliminated if local charge fluctuations in the SWCNT environment are removed, which would otherwise lead to local electric-field-induced Stark effects in the PL emission. On the microscopic scale there are several possible mechanisms to account for spectral diffusion or changes to the exciton oscillator strength, such as protonation at the sidewalls of the SWCNTs during local chemical reactions with acids and bases,[22,31] modified band gap values caused by oxygen doping upon ozone/light treatment,[32] or removal of surface-bound water molecules proximal to the nanotubes.[27−30] Interestingly, we also observe that after about two weeks of exposure to ambient air the PMMA-covered samples degrade and, as a result, again display spectral diffusion and blinking. Subsequent heat treatment again results in strong suppression of spectral diffusion and blinking signatures, demonstrating that the effect is reversible. Furthermore, we also observe suppression of spectral diffusion and enhanced light emission when coating the SWCNTs with polyvinyl alcohol (PVA) dissolved in water instead of PMMA (see Supporting Information),

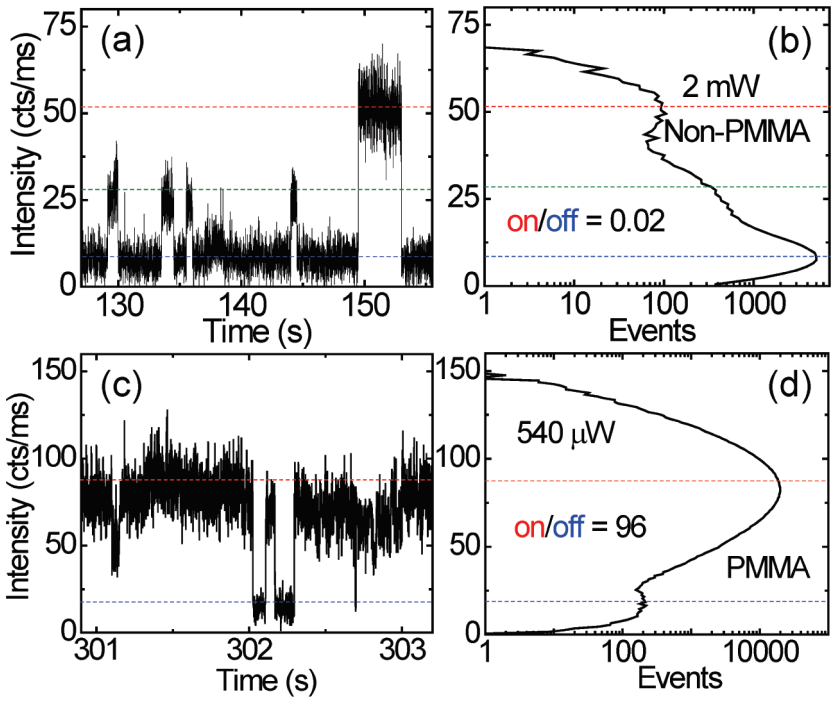

Figure 3. (a, c) TCPC plots for individual SWCNTs without (a) and with (c) a PMMA capping layer showing blinking-fluorescence intermittency for a small segment of the entire TCPC trace recorded for 30 min. (b, d) Plots of occurrence of a count value within 1 ms time bins for the entire TCPC trace without (b) and with (d) PMMA. Dotted lines indicate the peak maximum of the on (upper) and off (lower) states.

indicating that the effect does not depend on the specific chemistry of the chosen polymer. The observed rehydration in an ambient environment suggests that the presence/absence of physisorbed water molecules, which can act as charge trap states in the PMMA layer,[27−30] are responsible for the suppression of spectral diffusion.

To further quantify the achieved suppression of blinking, we increased the temporal resolution of spectral intensity measurements, at the cost of losing spectral energy information, by sending the PL emission through a 10 nm wide interference filter and coupling into a silicon avalanche photodiode with 400 ps timing jitter. Time-correlated photon counts (TCPC) were tagged in 1 ms time bins. To illustrate a consistent improvement in device performance, we repeated the same set of photon count measurements for 10 samples, five samples with and five without PMMA (non-PMMA). Plots of intensity counts per millisecond during a small time segment of a TCPC trace are shown in Figure 3a for a non-PMMA sample. Individual blinking times range from 1 ms to several seconds, with emission jumping between typically two, sometimes three, defined levels. To better illustrate the percentage of time spent in the on and off emission states, we plotted occurrence of a count value within 1 ms for the entire TCPC trace in Figure 3b for the non-PMMA sample. For this particular SWCNT a trimodal distribution is visible, which peaks at about 5000 events in the off state, 400 events in the intermediate state, and about 100 events in the on state, as illustrated by the dotted lines. Clearly, the noncoated SWCNTs spent most of their time in the off state and are thus fairly inefficient quantum emitters.

The TCPC traces can also be analyzed by plotting the probability distributions $P(t_{on})$ and $P(t_{off})$ for the on and off periods, respectively. This leads to the characteristic power-law distributions $P(t_{off})=At_{off}^{-n}$ and truncated power laws $P(t_{on})=At_{on}^{-m}\exp(-t_{on}/\tau_{on})$ for blinking quantum emitters,[13,14,33] where A is a constant, $m(n)$ is the on-state (off-state) slope, and τ_{on} is the saturation time. In contrast to most studies on "zero-dimensional" quantum dots, the $P(t_{on})$ dependence of investigated "one-dimensional" SWCNTs displays no truncation, *i.e.*, follows the simple power law $P(t_{on})=At_{on}^{-m}$ (see Supporting Information). As was recently pointed out by Crouch *et al.*, care must be taken when extracting the slopes m, which can depend significantly on the binning time, threshold level, and total number of blinking events.[33] Indeed, when the number of blinking events was low (100−5000), the values for m scatter in a large range between 1.35 and 2.3 (see Supporting Information), *i.e.*, outside of the range $1.5 \leq m \leq 2$ expected for a Cole−Davidson dielectric medium.[34] When the event count was above 5000 and the threshold value was set slightly higher than the center threshold value of the bimodal distribution, slope values were found in a much narrower range of 1.5 to 1.88. However, no clear trend was found between PMMA and non-PMMA samples, indicating that the

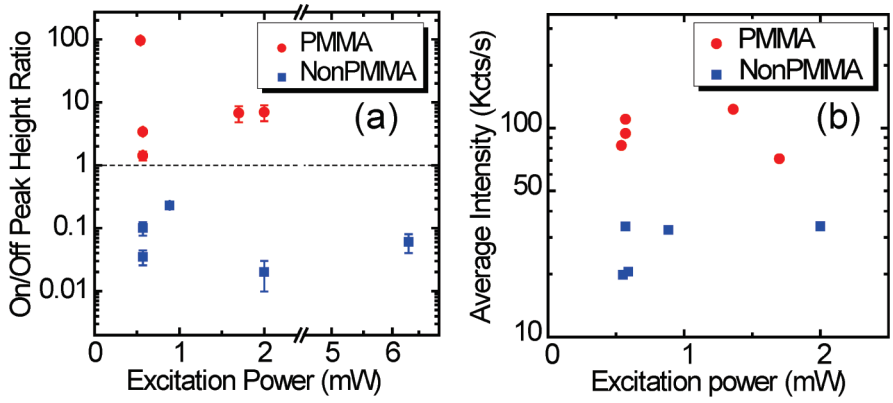

Figure 4. Statistical plot showing suppression of blinking and enhancement of exciton emission after PMMA capping/baking of individual SWCNTs (dots) compared to non-PMMA-coated samples (squares). (a) On/off ratio distribution for 10 samples recorded at various pump powers, with error bars for different time binning. (b) Average SWCNT emission intensity (kcts/s) for 10 samples recorded at various pump powers.

on-time slope is not very sensitive to the dielectric environment, similar to a previous investigation on organic molecules.[35]

To better quantify the improvement achieved by the PMMA coating technique, we introduce here the on/off ratio as defined by the peak ratio between on and off event counts, which is a measure for the percentage of time spent in the on state relative to the off state. For the non-PMMA sample a value of 0.02 is found, as indicated in Figure 3b. Note that the binning time (1−10 ms) has a minor influence on the extracted on/off ratio (see supptorting Information). The corresponding plots for the PMMA sample in Figure 3c show that emission remains in the on state a majority of the time, with occasional blinks to the off state. The occurrence plot in Figure 3d shows a bimodal distribution with an on/off ratio of 96, which is 3 orders of magnitude larger than the value for the non-PMMA sample, demonstrating successful suppression of blinking. The appearance of either a bimodal or trimodal distribution is random depending on the particular SWCNT chosen and does not significantly depend on the laser excitation power (see Supporting Information). While the off state is rather narrow and well-defined in both cases, the on state appears as a rather broad distribution due to the one-dimensional nature of the SWCNT, causing a variation in the on-state intensity level over time, as seen in previous studies of nanorods and nanowires.[14]

To further demonstrate that the improvement in on/off ratio does not depend significantly on excitation power, we compare the on/off ratios for five PMMA and five non-PMMA samples, as shown in Figure 4a. The data demonstrate that all non-PMMA blinking samples have an on/off ratio well below 1, even at the highest excitation powers up to 6 mW, where the PL emission saturates. In contrast, the PMMA samples have on/off ratios well above 1, with highest values up to 96. Therefore, the experimental on/off ratio introduced above, which varies by 3 orders of magnitude, is a much better indicator for changes in the charge configuration of the dielectric environment than the slopes from the power-law model.

In principle, the ratios determined from the occurrence histogram and the slopes of the power laws should be related. To this end, we calculated the expected on/off ratio using the estimated power-law slopes and total number of events and simulated the length of blinking intervals with random numbers, as further detailed in the Supporting Information. While the general trend of systematically larger on/off ratios for PMMA-coated samples is reproduced, the magnitude of the simulated on/off ratio is much smaller than the experimental one. This indicates that the SWCNTs with their inherent one-dimensional density of states do not obey a simple blinking model, in contrast to zero-dimensional structures, which are well explained by this blinking model.

Finally, Figure 4b demonstrates that all samples subjected to the PMMA coating and baking procedure display a factor 3 to 5 improvement in the integrated exciton emission rate as compared to non-PMMA samples recorded at similar or even at higher excitation powers, where the PL emission saturates. This improvement in exciton emission intensity was achieved by effectively removing trap states in the local environment, leading to a strong suppression of charge tunneling effects in and out of the SWCNTs, with the effect that emission occurs predominantly from the on state, which effectively enhances the exciton emission rate. The PMMA layer furthermore encapsulates the SWCNTs from the environment and can be utilized as a dielectric spacer medium in Fabry−Perot-type cavities.[6]

CONCLUSIONS

We successfully suppressed fluorescence intermittency and spectral diffusion of individual SWCNTs by utilizing PMMA capping and baking. As a result, up to 5-fold enhancement of the exciton emission was observed due to the removal of charge trap states. In

addition, we have shown that the on/off ratio from statistical occurrence analysis is very sensitive to changes in the local environment, while the often used slope of the blinking power laws is largely independent of the environment. Our approach is compatible with integration into cavities, in contrast to previous demonstrations of spectral diffusion suppression achieved in free-standing SWCNTs. This opens up possibilities to couple the exciton emission of nonblinking SWCNTs to cavity modes to further benefit by the Purcell effect and to enhance the light extraction efficiency, in order to ultimately demonstrate efficient photonic devices such as nanolasers[36] or quantum light sources[5] operating at room temperature.

METHODS

Experimental Setup. Measurements of microphotoluminescence (μ-PL) were taken by placing samples inside a liquid helium cryostat with a 7 K base temperature and exciting them with a CW laser diode operating at 780 nm. A spot size of about 1.5 μm was achieved using a microscope objective of NA 0.55. The relative position between sample and laser spot was positioned with a piezoelectric actuator (Attocube Systems) mounted directly inside the cryostat, which actuates the sample holder. Spectral emission from the sample was dispersed using a spectrometer with a 0.75 m focal length and spectral resolution of 0.05 nm and imaged by a liquid nitrogen cooled CCD camera. Laser stray light was rejected combining a 630 nm notch filter and 800 nm high-pass filter. In order to collect more light from individual SWCNTs, we used an index-matching solid immersion lens (S-LAH79 with refractive index 1.97) on top of the wafer to enhance the light extraction from the high-index medium, which is limited by total internal reflection.

Sample Preparation. Commercial CoMoCat SWNTs were prepared with bath sonication for 1 h in a vial containing 0.4 wt % sodium dodecylsulfate (SDS) solution. The product was poured through a 5 μm filter to form a concentration of 0.2 mg/mL and deposited directly onto a 90 nm SiO_2 substrate on top of a p++ type Si wafer. To effectively remove charge trap states, SWCNT samples were spin coated with poly(methyl methacrylate) (PMMA) and baked on a hot plate for several days at 150 °C.

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Supporting Information Available: Further details of analysis of number of events, threshold, and binning time on extracted on/off ratios and power-law slopes, influence of excitation power and capping material on blinking behavior, and numerical simulations of on/off ratios expected from the power-law model are provided. This material is available free of charge via the Internet at http://pubs.acs.org.

REFERENCES AND NOTES

1. Wang, F.; Dukovic, G.; Brus, L. E.; Heinz, T. F. The Optical Resonances in Carbon Nanotubes Arise from Excitons. *Science* **2005,** *308*, 838–841.
2. Misewich, J. A.; Martel, R.; Avouris, Ph.; Tsang, J. C.; Heinze, S.; Tersoff, J. Electrically Induced Optical Emission from a Carbon Nanotube FET. *Science* **2003,** *300*, 783–786.
3. Avouris, P.; Freitag, M.; Perebeinos, V. Carbon-Nanotube Photonics and Optoelectronics. *Nat. Photonics* **2008,** *2*, 341–350.
4. Högele, A.; Galland, C.; Winger, M.; Imamoğlu, A. Photon Antibunching in the Photoluminescence Spectra of a Single Carbon Nanotube. *Phys. Rev. Lett.* **2008,** *100*, 217401/1–217401/4.
5. Strauf, S.; Stolz, N. G.; Rakher, M. T.; Coldren, L. A.; Petroff, P. M.; Bouwmeester, D. High-Frequency Single-Photon Source with Polarization Control. *Nat. Photonics* **2007,** *1*, 704–708.
6. Xia, F.; Steiner, M.; Lin, Y; Avouris, P. A Microcavity-Controlled, Current-Driven, On-Chip Nanotube Emitter at Infrared Wavelengths. *Nat. Nanotechnol.* **2008,** *3*, 609–613.
7. Moerner, W. E.; Orrit, M. Illuminating Single Molecules in Condensed Matter. *Science* **1999,** *283*, 1670–1676.
8. Nirmal, M.; Dabbousi, B. O.; Bawendi, M. G.; Macklin, J. J.; Trautman, J. K.; Harris, T. D.; Brus, L. E. Fluorescence Intermittency in Single Cadmium Selenide Nanocrystals. *Nature* **1996,** *383*, 802–804.
9. Matsuda, K.; Kanemitsu, Y.; Irie, K.; Saiki, T.; Someya, T.; Miyauchi, Y.; Maruyama, S. Photoluminescence Intermittency in an Individual Single-Walled Carbon Nanotube at Room Temperature. *Appl. Phys. Lett.* **2005,** *86*, 123116/1–123116/3.
10. Htoon, H.; O'Connell, M. J.; Cox, P. J.; Doorn, S. K.; Klimov, V. I. Low Temperature Emission Spectra of Individual Single-Walled Carbon Nanotubes: Multiplicity of Subspecies within Single-Species Nanotube Ensembles. *Phys. Rev. Lett.* **2004,** *93*, 027401/1–027401/4.
11. Matsuda, K.; Inoue, T.; Murakami, Y.; Maruyama, S.; Kanemitsu, Y. Exciton Fine Structure in a Single Carbon Nanotube Revealed through Spectral Diffusion. *Phys. Rev. B* **2008,** *77*, 193405/1–193405/4.
12. Cichos, F.; von Borczyskowski, C.; Orrit, M. Power-Law Intermittency of Single Emitters. *Curr. Opin. Colloid Interface Sci.* **2007,** *12*, 272–284.
13. Stefani, F. D.; Hoogenboom, J. P.; Barkai, E. Beyond Quantum Jumps: Blinking Nano-Scale Light Emitters. *Phys. Today* **2009,** *62*, 34–39.
14. Frantsuzov, P.; Kuno, M.; Jankó, B.; Marcus, R. A. Universal Emission Intermittency in Quantum Dots, Nanorods, and Nanowires. *Nature* **2008,** *4*, 519–522.
15. Sher, P. H.; Smith, J. M.; Dalgarno, P. A.; Warburton, R. J.; Chen, X.; Dobson, P. J.; Daniels, S. M.; Pickett, N. L.; O'Brien, P. Power Law Carrier Dynamics in Semiconductor Nanocrystals at Nanosecond Timescales. *Appl. Phys. Lett.* **2008,** *92*, 101111/1–101111/3.
16. Empedocles, S. A.; Bawendi, M. G. Quantum-Confined Stark Effect in Single CdSe Nanocrystallite Quantum Dots. *Science* **1997,** *278*, 2114–2117.
17. Müller, J.; Lupton, J. M.; Rogach, A. L.; Feldmann, J.; Talapin, D. V.; Weller, H. Monitoring Surface Charge Movement in Single Elongated Semiconductor Nanocrystals. *Phys. Rev. Lett.* **2004,** *93*, 167402/1–167402/4.
18. Kiowski, O.; Lebedkin, S.; Hennrich, F.; Kappes, M. M. Single-Walled Carbon Nanotubes Show Stable Emission and Simple Photoluminescence Spectra with Weak Excitation Sidebands at Cryogenic Temperatures. *Phys. Rev. B* **2007,** *76*, 075422/1–075422/8.
19. Bradac, C.; Gaebel, T.; Naidoo, N.; Sellars, M. J.; Twamley, J.; Brown, L. J.; Barnard, A. S.; Plakhotnik, T.; Zvyagin, A. V.; Rabeau, J. R. Observation and Control of Blinking Nitrogen-Vacancy Centres in Discrete Nanodiamonds. *Nat. Nanotechnol.* **2010,** *5*, 345–349.
20. Wang, X.; Ren, X.; Kahen, K.; Hahn, M. A.; Rajeswaran, M.; Maccagnano-Zacher, S.; Silcox, J.; Cragg, G. E.; Efros, A. L.; Krauss, T. D. Non-blinking Semiconductor Nanocrystals. *Nature* **2009,** *459*, 686–689.
21. Htoon, H.; O' Connell, M. J.; Doorn, S. K.; Klimov, V. I. Single Carbon Nanotubes Probed by Photoluminescence Excitation Spectroscopy: The Role of Phonon-Assisted Transitions. *Phys. Rev. Lett.* **2005,** *94*, 127403/1–127403/4.

22. O'Connell, M. J.; Bachilo, S. M.; Huffman, C. B.; Moore, V. C.; Strano, M. S.; Haroz, E. H.; Rialon, K. L.; Boul, P. J.; Noon, W. H.; Kittrell, C.; *et al.* Band Gap Fluorescence from Individual Single-Walled Carbon Nanotubes. *Science* **2002,** *297*, 593–596.
23. Bachilo, S. M.; Strano, M. S.; Kittrell, C.; Hauge, R. H.; Smalley, R. E.; Weisman, R. B. Structure-Assigned Optical Spectra of Single-Walled Carbon Nanotubes. *Science* **2002,** *298*, 2361–2366.
24. Tan, P. H.; Rozhin, A. G.; Hasan, T.; Hu, P.; Scardaci, V.; Milne, W. I.; Ferrari, A. C. Photoluminescence Spectroscopy of Carbon Nanotube Bundles: Evidence for Exciton Energy Transfer. *Phys. Rev. Lett.* **2007,** *99*, 137402/1–137402/4.
25. Murakami, Y.; Kono, J. Nonlinear Photoluminescence Excitation Spectroscopy of Carbon Nanotubes: Exploring the Upper Density Limit of One-Dimensional Excitons. *Phys. Rev. Lett.* **2009,** *102*, 037401/1–037401/4.
26. Lefebvre, J.; Fraser, J. M.; Finnie, P.; Homma, Y. Photoluminescence from an Individual Single-Walled Carbon Nanotube. *Phys. Rev. B* **2004,** *69*, 075403/1–075403/5.
27. Kim, W.; Javey, A.; Vermesh, O.; Wang, Q.; Li, Y.; Dai, H. Hysteresis Caused by Water Molecules in Carbon Nanotube Field-Effect Transistors. *Nano Lett.* **2003,** *3*, 193–198.
28. Ong, H. G.; Cheah, J. W.; Chen, L.; Tangtang, H.; Xu, Y.; Li, B.; Zhang, H.; Li, L.; Wang, J. Charge Injection at Carbon Nanotube-SiO_2 Interface. *Appl. Phys. Lett.* **2008,** *93*, 093509/1–093509/3.
29. Vijayaraghavan, A.; Kar, S.; Soldano, C.; Talapatra, S.; Nalamasu, O.; Ajayan, P. M. Charge-Injection-Induced Dynamic Screening and Origin of Hysteresis in Field-Modulated Transport in Single-Wall Carbon Nanotubes. *Appl. Phys. Lett.* **2006,** *89*, 162108/1–162108/3.
30. Ai, N.; Sul, O.; Begliarbekov, M.; Song, Q.; Kumar, K.; Choi, D. S.; Yang, E. H.; Strauf, S. Transconductance and Coulomb Blockade Properties in In-Plane Grown Carbon Nanotube Field Effect Transistors. *Nanosci. Nanotechnol. Lett.* **2010,** *2*, 73–78.
31. Cognet, L.; Tsyboulski, D. A.; Rocha, J. R.; Doyle, C. D.; Tour, J. M.; Weisman, R. B. Stepwise Quenching of Exciton Fluorescence in Carbon Nanotubes by Single-Molecule Reactions. *Science* **2007,** *316*, 1465–1468.
32. Ghosh, S.; Bachilo, S. M.; Simonette, R. A.; Beckingham, K. M.; Weisman, R. B. Oxygen Doping Modifies Near-Infrared Band Gaps in Fluorescent Single-Walled Carbon Nanotubes. *Science* **2010,** *330*, 1656–1659.
33. Crouch, C. H.; Sauter, O.; Wu, X.; Purcell, R.; Querner, C.; Drndic, M; Pelton, M. Facts and Artifacts in the Blinking Statistics of Semiconductor Nanocrystals. *Nano Lett.* **2010,** *10*, 1692–1698.
34. Tang, J.; Marcus, R. A. Diffusion-Controlled Electron Transfer Processes and Power-Law Statistics of Fluorescence Intermittency of Nanoparticles. *Phys. Rev. Lett.* **2005,** *95*, 107401/1–107401/4.
35. Hoogenboom, J. P.; Hernando, J.; van Dijk, E. M. H. P.; van Hulst, N. F.; García-Parajó, M. F. Power-Law Blinking in the Fluorescence of Single Organic Molecules. *ChemPhysChem.* **2007,** *8*, 823–833.
36. Strauf, S.; Hennessy, K.; Rakher, M. T.; Choi, Y.-S.; Badolato, A.; Andreani, L. C.; Hu, E. L.; Petroff, P. M.; Bouwmeester, D. Self-Tuned Quantum Dot Gain in Photonic Crystal Lasers. *Phys. Rev. Lett.* **2006,** *96*, 127404/1–127404/4.