Export Citatio

View Onlin

Mechanism of quantum yield enhancement in Si quantum dots by high-pressure water vapor annealing from single-dot studies

Cite as: Appl. Phys. Lett. **125**, 071110 (2024); doi: 10.1063/5.0223989 Submitted: 18 June 2024 · Accepted: 2 August 2024 · Published Online: 15 August 2024

Xi Lu,¹ 🛅 Jingjian Zhou,² 🛅 Bernard Gelloz,³ 🍺 and Ilya Sychugov^{1,a)} 🛅

AFFILIATIONS

¹Department of Applied Physics, KTH Royal Institute of Technology, Stockholm 11419, Sweden ²Department of Physics and Astronomy, Uppsala University, SE-75121 Uppsala, Sweden ³Graduate School of Science, Nagoya University, Furo-cho, Chikusa, Nagoya, Aichi 464-8602, Japan

^{a)}Author to whom correspondence should be addressed: ilyas@kth.se

ABSTRACT

High-pressure water vapor annealing (HWA) was recently demonstrated as a method that can substantially improve the photoluminescence quantum yield (PLQY) of silicon quantum dots (Si QDs) with the oxide shell. In this Letter, the mechanism of this enhancement is studied optically on a single-dot level. HWA treatment is performed on Si QDs formed on a silicon-on-insulator wafer, and their photoluminescence (PL) properties were examined before and after the treatment. Our experiments show a 2.5 time enhancement in the average blinking duty cycle of Si QDs after 2.6 MPa HWA treatment without changing the average ON-state PL intensity. This observation proves the carrier trapping process is suppressed on the HWA-built Si/SiO₂ interface. We also discussed the mechanism behind the PLQY enhancement of HWAtreated Si QDs by comparing single-dot-level data to reported ensemble PL Si QDs results. HWA treatment is found to mainly brighten "grey" (not 100% efficient) QDs, a mechanism different from changing dark (non-emitting) to bright (100% efficient) Si QDs by ligand passivation.

© 2024 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution-NonCommercial 4.0 International (CC BY-NC) license (https://creativecommons.org/licenses/by-nc/4.0/). https://doi.org/10.1063/5.0223989

The silicon/silicon dioxide (Si/SiO₂) interface plays a crucial role in silicon bulk and nano device technologies, such as quantum dots (QDs), metal-oxide-semiconductor field-effect transistors (MOSFETs), solar cells, etc. The Si/SiO2 interface often contains a small number of defects, which can be vacancies or Si dangling bonds, subject to preparation conditions.¹⁻⁶ These defects generate localized states within the energy bandgap, where electrons and/or holes are trapped, thereby limiting the electrical and optical performance of devices. For instance, in MOSFETs, this trapping-detrapping phenomenon can result in random telegraph signal (RTS) noise, which is recognized as a critical issue in analog circuits.⁷⁻¹⁰ In QDs, trappingdetrapping can cause blinking,4,11-14 significantly reducing the photoluminescence quantum yield (PLQY).

High-pressure water vapor annealing (HWA) has been reported as an effective method for reducing the density of defect states on the Si/SiO₂ surface, thereby improving the electrical performance, and reducing trapping-related behaviors of MOSFETs.^{15–19} Similarly, HWA has also been recently suggested and successfully used as a method to enhance the PLQY of porous Si^{20-22} and Si QDs up to QY > 50%, 23,24 which is of large interest for CMOS-compatible technology applications.^{25}

In general, HWA can both reduce the number of interface defects, such as P_b centers,²¹ and form a more relaxed and stable bulk part of the oxide network. The latter was shown from Fourier-transform infrared spectroscopy (FTIR) data through bond lengths and angles being closer to that of perfect quartz for Si nanostructures^{22,23} and bulk films.¹⁵ However, although reported Fourier-transform infrared spectroscopy (FTIR) and electron spin resonance (ESR) analyses indicate a reduction in density of defect states at the Si/SiO₂ interface of Si QDs in HWA treatment, the mechanism of PLQY enhancement in nano-silicon remains unclear.

The PLQY measured in ensemble QD cannot differentiate between the contribution to the QY enhancement of "bright" (100% efficient), "dark" (non-emitting), and "grey" (not 100% efficient) QDs. Those contain non-radiative centers with the recombination rate negligible, much faster, and comparable to the radiative recombination, respectively. They originate from different type of defect states.^{26–28} In dark QDs a single defect on the surface or in the core would render nanoparticle non-emissive. Indeed, the non-radiative recombination rate in bulk semiconductors is limited by the diffusion time of carriers to the non-radiative center. In nanocrystals, carriers are strongly localized, and the non-radiative process in such QDs would be instantaneous. Grey QDs may contain a trap state, leading to QD charging and subsequent non-radiative Auger recombination. Trap position and the carrier tunneling rate will then define a typical switching frequency between bright and dark states and the average level of grey QD PL intensity.

In this Letter, we performed HWA on individual Si QDs with an SiO_2 shell formed on Si wafers to elucidate the mechanism of the PLQY enhancement. We studied the photoluminescence (PL) blinking properties of the Si QDs before and after HWA treatment. Our results demonstrate that HWA improves the average duty cycle of the Si QDs while maintaining the average ON-state PL intensity for up to moderate HWA treatment. Therefore, we provide direct experimental evidence of the mechanism of PLQY enhancement under HWA treatment as being related to the reduction of the carrier trapping at the interface. Results are compared to previous ensemble data and to the ligand passivation method for Si QDs.

Methods for fabricating Si QDs and their properties are detailed elsewhere.^{29,30} In brief, Si QDs were created from the silicon-oninsulator device layer through RIE etching followed by rapid thermal annealing (RTA). As depicted in Fig. 1(a), after the RIE, the chip ($\approx 1 \text{ cm}^2$) exhibited a device (silicon) layer thickness gradient, with some areas (closer to the edge) already exposing buried oxide, while other areas (closer to the center) still featuring a thick device layer. Importantly, in between these two a fraction of the surface ($\sim 100 \ \mu \text{m}$ wide) is covered by a mixture of Si quantum wells (QWs) and well-dispersed, isolated Si QDs. The structure of Si QDs was characterized using scanning transmission electron microscopy (STEM, JEOL



FIG. 1. (a) Scheme of QDs on chip fabricated by RIE etching, (b) TEM, and (c) PL image of the sample area with single QD.

JEM-2500SES), with samples prepared using a conventional focused ion beam (FIB) milling technique with a carbon protection layer. Examples of cross-sectional TEM image of an Si QD and Si QW postthermal annealing are shown in Fig. 1(b) and Fig. S1, respectively. Silicon lattice fringes are visible. The Si QDs were non-spherical, with a darker contrast region surrounding the Si QD, indicating the presence of an oxide shell as a result of RTA at 900 °C (10 s) in oxygen atmosphere. These Si QDs were studied in detail previously by singledot PL spectroscopy in luminescence, absorption, and lifetime modes, including low-temperature measurements, as well as in conjunction with theory.³¹

For HWA treatment, Si QD chips and a pre-calculated amount of de-ionized water were loaded into a stainless-steel chamber at room temperature. The tightly closed chamber was then placed on a hot plate pre-heated at 260 °C. After 4 h, the chamber was allowed to cooldown to room temperature and opened. The amount of water set in the chamber was determined using the Van der Walls gas equation for water to produce the desired pressure at 260 °C. In this report, pressures of 1.3, 2.6, and 3.9 MPa were used, corresponding to typical values studied in previous reports on ensembles.^{21,22}

Optical properties of the treated Si QDs at the single-particle level were studied using a micro-PL setup. The excitation beam was directed to the sample from outside the microscope at an incident angle of approximately 45° (dark-field configuration). Emission light from the sample was collected by an inverted optical microscope (Zeiss Axio Observer Z1) with a $100 \times$ objective lens (Nikon, NA = 0.73) and filtered by a 442 nm long-pass filter (Semrock) to remove excitation light. The emitted light was then captured by a thermoelectrically cooled EMCCD camera (Andor iXon3 888) with a frame rate of 1 Hz. Figure 1(c) (micro-PL image) confirms that the surface density of Si QDs on the chip was low enough to allow single-dot measurements under a far-field microscope. It was also confirmed by single-dot spectroscopy revealing different luminescence peak positions for different nanodots as a result of the quantum size effect (Fig. S2).

PL intensity of the QD luminescence over time was extracted using a built-in plugin in ImageJ. Figure 2 shows the PL intensity trace of several typical individual Si QDs, where the blinking phenomenon can be clearly observed both before and after the HWA treatment.



FIG. 2. PL intensity time trace and the corresponding intensity histograms of one example of single QD (a) before HWA treatment, (b) after 1.3Mpa HWA treatment, and (c) after 2.6Mpa HWA treatment.



Both blinking traces and intensity histograms demonstrate distinct ON and OFF states in PL intensity. To define the ON state, we initially fit the OFF-state intensity distribution with a Gaussian fitting, then points with intensities greater than $X_c + 3\sigma$ were considered as the ON state, where X_c represents the mean (around zero) and σ signifies the standard deviation of the Gaussian fitting. We define duty cycle as a fraction of time spent in the ON-state for any given trace.

Unlike in II–VI QD counterparts with a power-law distribution of ON- and OFF-times, blinking in Si QDs is of purely random character.^{11,32} It implies a Markov process with a mono-exponential distribution of ON- and OFF-times. Therefore, average quantities, such as average time spent in each state $\tau_{\rm ON}$ and $\tau_{\rm OFF}$, exist as a first moment of corresponding distributions. The duty cycle is then directly related to these average quantities as $d = \tau_{\rm ON}/(\tau_{\rm ON} + \tau_{\rm OFF})$ and can be used for monitoring modifications introduced by different treatments.

We employed a statistical method to analyze changes in the QD behavior. As shown in Fig. 3(a), the average ON-state intensities exhibited no significant changes following both HWA treatments. At the same time, an approximately 2.5 time enhancement was observed

in the average duty cycle after the 2.6 MPa HWA treatment, and a 1.5 time enhancement was observed after the 1.3 MPa HWA treatment.

We start the discussion by the observation of unchanged intensity in the ON-state for HWA-treated Si QDs. The excitation power density used in the experiment was below the saturation level of Si QDs.³³ The ON-state intensity can be described as being proportional to³⁴

$$I \sim \eta \alpha I_{\text{ex}},$$
 (1)

where η is the internal quantum efficiency (IQE, determined by radiative and non-radiative recombination rate), α is the absorption crosssession, and I_{ex} is the excitation intensity. Quantum dots before and after HWA treatment have similar optical characteristics (intensity and emission spectral range), indicating a similar absorption cross section. The fact that the average ON-state intensities remain the same after 1.3 and 2.6 MPa HWA treatment indicates that there were also no changes to their IQE because of HWA treatment (other parameters kept the same). That would be expected when the luminescence is governed by the Si QD core properties only.

However, this does not imply that the QD core remains unchanged. Based on previous ensemble studies of HWA-treated Si NCs it is established that HWA treatment can build additional oxide layers rather than only modifying the existing shell.^{22,24,35,36}

Experimentally, we could not track the same Si QDs before and after the treatment. One possible explanation is that HWA treatment simultaneously shrinks all the quantum dots, where "old" Si QDs shrink below the size threshold and their PL quenched, while larger Si islands are oxidized to the appropriate size, thereby becoming Si QDs with an improved oxide shell. An indication is, as shown in Fig. 4, after the 2.6 MPa HWA treatment, a thinner QW side disappeared (red circles), being presumably completely oxidized. At the same time, after the 1.3 MPa HWA treatment, the thinner QW edge only became blurred in the PL image rather than disappearing entirely. This observation suggests the presence of Si core oxidation, and this oxidation process is pressure dependent. Due to the substantial difference in the interface curvature between the QDs and QWs, the oxidation rate for QDs is considerably different from that for QWs.^{24,35,37} Therefore, even when most of the QWs remain, old QDs are highly likely to shrink beyond PL detection, while new ones with very similar corerelated PL properties are formed.



FIG. 4. PL images of QWs and QDs before and after (a) 1.3 MPa HWA treatment and (b) 2.6 MPa HWA treatment; red circles indicate disappeared QW after the treatment.

Next, we turn to the QD property, which was modified, namely the blinking duty cycle. According to the well-established chargingblinking model, the OFF-state of a blinking QD occurs when a charged carrier tunnels into a trap state on the Si/SiO₂ interface, leading to luminescence quenching via nonradiative Auger processes. Luminescence resumes only after the trapped carrier thermally returns to the QD after a certain time. Therefore, the increase in the blinking duty cycle suggests that the probability of carrier trapping is reduced, providing evidence of a lower defect density on the HWA-treated Si/SiO₂ surface. As Fig. 3(b) demonstrates, the duty cycle enhancement is pressure dependent, showing that the ability of the HWA treatment of building/modifying the oxide shell can be controlled by adjusting the pressure.

However, as was also previously found in ensemble measurements, these two processes of the core and shell modification cannot be entirely decoupled. In other words, it is unlikely to remove all the interface defects by increasing reaction time or pressure. In a control experiment, we found that HWA cannot improve the duty cycle of Si QDs, which already have a thick and relaxed³⁰ (annealed at 1100 °C in Argon) oxide shell. As shown in Fig. S3(b), the average duty cycle of this type of single-dot sample before and after 3.9 MPa HWA treatment remained largely unchanged (80% and 75%, respectively). In contrast, the 900 °C oxidation is known to be self-limiting^{38,39} due to the building up of the interface stress. This results in the formation of trap sites, which HWA can, at least partially, eliminate.

Now we can summarize observations from single-dot studies in relation to previous reports on standard characterization of such QD ensembles. In a blinking QD, QY depends on not only IQE but also on the duty cycle. The former can represent very fast (unresolved) blinking or other charge-related processes on a shorter timescale.^{40,41} Therefore, when comparing with the ensemble behavior of blinking QDs, the IQE, the duty cycle, and the fraction of dark QDs (QY = 0) should be all considered. Since dark QDs cannot be seen and analyzed, we can only compare single-dot data with integrated ensemble PL intensity instead of PLQY, where dark QDs contribute through absorption. The luminescence intensity *I* of ensemble sample can be expressed as

$$I \propto \left(\eta_{\rm b} \cdot d_{\rm b} \cdot p_{\rm b} + \eta_{\rm g} \cdot d_{\rm g} \cdot p_{\rm g}\right),\tag{2}$$

$$p_{\rm b} + p_{\rm g} + p_{\rm d} = 1,$$
 (3)

where $\eta_{\rm b}$, $\eta_{\rm g}$ are the relative IQEs, $d_{\rm b}$, $d_{\rm g}$ are the relative duty cycles, $p_{\rm b}$, $p_{\rm g}$, $p_{\rm d}$ are fractions of bright Si QDs, gray Si QDs, and dark Si QDs, respectively. According to the results discussed above regarding uniform and unchanged ON-state intensity, $\eta_{g} \approx \text{const.}$ Next, p_{b} can be considered as 0 from experimental results, as Si QDs with a 100% duty cycle are rarely found in these samples and luminescence decays are not mono-exponential in such single Si QDs.⁴¹ Then, Eq. (2) can be simplified to $I \propto d_{\rm g} \cdot p_{\rm g}$ here. As shown in Fig. 5, the reported PL intensity enhancement in porous silicon²² matches well with the relative duty cycle from this work ($I \propto d_g$). So, the p_g quantity remains unchanged under HWA treatment. In the experiment, the quantity of Si QDs observed by PL did not increase (it was estimated from the QD surface density count from PL images before and after the treatment), which is consistent with this conclusion. One can speculate that relaxed oxide may not be able to passivate dangling bonds or affect core-related defects in the dark QDs.



FIG. 5. Relative duty cycle enhancement from single-dot studies (this work, red) and that of ensemble of porous silicon²² (blue) under different HWA pressure treatments.

Therefore, summing all the discussions above, one can draw the conclusion that the HWA treatment only enhances the quality of gray Si QDs by suppressing trapping at the Si/SiO₂ interface for moderate HWA treatment. At higher pressure with prolonged treatment, a substantial shrinkage of the Si core or even complete oxidation occurs, resulting in a reduced light emission [Fig. S3(a)]. Another reference sample with a very long HWA treatment (24 h at 1.3 MPa) resulted in the complete oxidation of Si nanodots (Fig. S4) and no observable PL. In general, the HWA oxide thickness depends on treatment time, pressure, geometry of the Si surface, and initial surface conditions. For porous Si and Si nanocrystals, 1–2 nm oxide thickness can be grown after 3–4 h, where the growth is fast in the beginning and then slowly saturates.^{23,24} For large Si nanowires (~100 nm in diameter), as much as ~20 nm oxide was grown in 3 h at a similar pressure.³⁷ In any case, extending HWA time inexorably extends the oxide thickness.

In conclusion, we have studied the mechanism of PLQY enhancement in QDs following HWA treatment at the single Si QD level. A 2.5 time enhancement of duty cycle is observed after 2.6 MPa HWA treatment, which indicates that, compared to the oxide layer formed by RTA, the trapping process due to defects on the Si/SiO₂ interfaces is suppressed on the improved oxide layer formed by HWA. Furthermore, the single Si QD-level data are compared with reported ensemble porous Si data, showing that the enhancement of HWA treatment mainly originates from gray QDs becoming brighter as opposed to the conversion of dark nanocrystals to bright ones. The latter is an established mechanism for quantum yield enhancement in ligand-passivated Si QDs with similar PL properties of red, near-infrared emission and $\sim \mu s$ lifetime.⁴² Thus, while the reported QY values in both cases are comparable (>50%), the mechanism behind improved efficiency is different for these two passivating shells.

See the supplementary material for additional microscopy and spectroscopy data.

X.L. acknowledges funding support from the China Scholarship Council (CSC). The work is supported by the Swedish Energy Agency Project No. P2022-00859.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xi Lu: Data curation (equal); Formal analysis (equal); Methodology (equal); Writing – original draft (equal); Writing – review & editing (equal). Jingjian Zhou: Methodology (supporting); Writing – review & editing (equal). Bernard Gelloz: Formal analysis (supporting); Methodology (supporting); Resources (equal); Writing – review & editing (equal). Ilya Sychugov: Formal analysis (equal); Methodology (equal); Resources (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

- ¹S. Nunomura, T. Tsutsumi, I. Sakata, and M. Hori, J. Vac. Sci. Technol. B **41**(5), 052202 (2023).
- ²E. H. Poindexter and P. J. Caplan, Prog. Surf. Sci. 14(3), 201 (1983).
- ³R. A. B. Devine, D. Mathiot, W. L. Warren, D. M. Fleetwood, and B. Aspar, Appl. Phys. Lett. **63**(21), 2926 (1993).
- ⁴N. P. Brawand, M. Voros, and G. Galli, Nanoscale 7(8), 3737 (2015).
- ⁵W. Fussel, M. Schmidt, H. Angermann, G. Mende, and H. Flietner, Nucl. Instrum. Methods Phys. Res., Sect. A **377**(2–3), 177 (1996).
- ⁶M. Zacharias, D. Hiller, A. Hartel, and S. Gutsch, Phys. Status Solidi A 209(12), 2449 (2012).
- ⁷M. J. Kirton and M. J. Uren, Adv. Phys. 38(4), 367 (1989).
- ⁸W. K. Chim, K. K. Leong, and W. K. Choi, Jpn. J. Appl. Phys., Part 1 40(1), 1 (2001).
- ⁹D. Veksler, G. Bersuker, S. Rumyantsev, M. Shur, H. Park, C. Young, K. Y. Lim, W. Taylor, and R. Jammy, in *International Reliability Physics Symposium* (IEEE, 2010), p. 73.
- ¹⁰Y. Yu, Z. Zhang, and S. Chen, "Analysis of low frequency noise in Schottky junction trigate silicon nanowire FET on bonded SOI substrate," IEEE Trans. Electron Devices 69, 4667–4673 (2022).
- ¹¹B. Bruhn, F. Qejvanaj, I. Sychugov, and J. Linnros, J. Phys. Chem. C 118(4), 2202 (2014).
- ¹²A. L. Efros and M. Rosen, Phys. Rev. Lett. 78, 1110 (1997).
- ¹³C. Galland, Y. Ghosh, A. Steinbrueck, M. Sykora, J. A. Hollingsworth, V. I. Klimov, and H. Htoon, Nature 479(7372), 203 (2011).
- ¹⁴ M. E. Pistol, P. Castrillo, D. Hessman, J. A. Prieto, and L. Samuelson, Phys. Rev. B 59(16), 10725 (1999).

- ¹⁵T. Sameshima and M. Satoh, Jpn. J. Appl. Phys., Part 2 36(6a), L687 (1997).
- ¹⁶Z. Zhang, M. D. Liao, Y. Q. Huang, X. Q. Guo, Q. Yang, Z. X. Wang, T. Gao, C.
- H. Shou, Y. H. Zeng, B. J. Yan, and J. C. Ye, Sol. RRL 3(10), 1900105 (2019).
- ¹⁷Y. Uraoka, M. Miyashita, Y. Sugawara, H. Yano, T. Hatayama, T. Fuyuki, and T. Sameshima, Jpn. J. Appl. Phys., Part 1 45(7), 5657 (2006).
- ¹⁸P. Punchaipetch, M. Miyashita, Y. Uraoka, T. Fuyuki, T. Sameshima, and S. Horii, Jpn. J. Appl. Phys., Part 2 45(4–7), L120 (2006).
- ¹⁹K. K. Selvi, N. DasGupta, and K. Thirunavukkarasu, Thin Solid Films 531, 373 (2013).
- ²⁰B. Gelloz, A. Kojima, and N. Koshida, Mater. Res. Soc. Symp. Proc. 832, 141 (2005).
- ²¹B. Gelloz, A. Kojima, and N. Koshida, Appl. Phys. Lett. **87**(3), 031107 (2005).
- ²²B. Gelloz and N. Koshida, J. Appl. Phys. **98**(12), 123509 (2005).
- ²³K. Q. Loh, H. P. Andaraarachchi, V. E. Ferry, and U. R. Kortshagen, ACS Appl. Nano Mater. 6(7), 6444 (2023).
- ²⁴B. Gelloz, F. B. Juangsa, T. Nozaki, K. Asaka, N. Koshida, and L. H. Jin, Front. Phys. 7, 47 (2019).
- ²⁵C. Zhang, P. Zeng, W. J. Zhou, Y. C. Zhang, X. P. He, Q. Y. Jin, D. C. Wang, H. T. Wang, S. Y. Zhang, M. Lu, and X. Wu, IEEE J. Sel. Top. Quantum Electron. 26(2), 1500107 (2020).
- ²⁶F. Sangghaleh, I. Sychugov, Z. Yang, J. C. Veinot, and J. Linnros, ACS Nano 9(7), 7097 (2015).
- 27 Y. Shu and B. G. Levine, J. Phys. Chem. C 118(14), 7669 (2014).
- ²⁸S. Miura, T. Nakamura, M. Fujii, M. Inui, and S. Hayashi, Phys. Rev. B 73(24), 245333 (2006).
- ²⁹I. Sychugov, Y. Nakayama, and K. Mitsuishi, Nanotechnology 21(28), 285307 (2010).
- 30 J. Zhou, F. Pevere, H. Gatty, J. Linnros, and I. Sychugov, Nanotechnology 31, 505204 (2020).
- ³¹I. Sychugov, F. Pevere, J. Luo, A. Zunger, and J. Linnros, Phys. Rev. B **93**, 161413 (2016).
- ³²I. Sychugov, R. Juhasz, J. Linnros, and J. Valenta, Phys. Rev. B 71(11), 115331 (2005).
- ³³F. Pevere, I. Sychugov, F. Sangghaleh, A. Fucikova, and J. Linnros, J. Phys. Chem. C 119(13), 7499 (2015).
- ³⁴J. Valenta and I. Pelant, *Luminescence Spectroscopy of Semiconductors* (Oxford University Press, US, 2012).
- ³⁵B. Gelloz and N. Koshida, Thin Solid Films **508**(1–2), 406 (2006).
- ³⁶B. Gelloz, R. Mentek, and N. Koshida, Jpn. J. Appl. Phys., Part 1 48(4), 04c119 (2009).
- ³⁷B. Salhi, B. Gelloz, N. Koshida, G. Patriarche, and R. Boukherroub, Phys. Status Solidi A 204(5), 1302 (2007).
- ³⁸H. I. Liu, N. I. Maluf, R. F. W. Pease, D. K. Biegelsen, N. M. Johnson, and F. A. Ponce, J. Vac. Sci. Technol. B **10**, 2846 (1992).
- ³⁹A. Fujiwara and Y. Takahashi, Nature **410**(6828), 560 (2001).
- ⁴⁰N. J. Orfield, J. R. McBride, F. Wang, M. R. Buck, J. D. Keene, K. R. Reid, H. Htoon, J. A. Hollingsworth, and S. J. Rosenthal, ACS Nano 10(2), 1960 (2016).
- ⁴¹F. Pevere, F. Sangghaleh, B. Bruhn, I. Sychugov, and J. Linnros, ACS Photonics 5(8), 2990 (2018).
- ⁴²J. Zhou, J. Huang, H. Chen, A. Samanta, J. Linnros, Z. Yang, and I. Sychugov, J. Phys. Chem. Lett. **12**, 8909 (2021).

Supporting information

Mechanism of quantum yield enhancement in Si quantum dots by highpressure water vapor annealing from single-dot studies

Xi Lu,¹ Jingjian Zhou,² Bernard Gelloz,³ and Ilya Sychugov^{1,*}

¹Department of Applied Physics, KTH Royal Institute of Technology, Stockholm, 11419, Sweden

² Department of Physics and Astronomy, Uppsala University, SE-75121 Uppsala, Sweden

³ Graduate School of Science, Nagoya University, Furo-cho, Chikusa, Nagoya, Aichi 464-8602, Japan

* ilyas@kth.se

1. TEM image of QW



Fig.S1 TEM image of a QW, including silicon lattice (darkest region) and surrounding oxide shell

2. Emission spectrum of Si QDs after HWA treatment



Fig.S2 a) PL spectrum of the sample after HWA treatment (horizontal axis as wavelength and vertical axis as vertical position), showing the Si QDs can be detected individually b) PL spectra of several typical Si QDs showing each Si QD has different emission peak

3. PL intensity and duty cycle before and after HWA treatment of the sample with a relaxed oxide shell under 3.9MPa.



Fig.S3 a) PL intensity before and after HWA treatment under 3.9MPa, with 10s exposure time b) Duty cycle before and after HWA treatment under 3.9MPa, with 2Hz frame rate

4. Wafer after 24h HWA treatment under 1.3MPa



Fig.S4 White light image a) before and b) after 24h 1.3MPa HWA treatment in same area, most of Si (brighter part) is completely oxidized as can be seen from the front position shift. Two spots in the right top part were used as reference points. No QD PL was observed from this sample.