

1. Quantification of grain size

PbS quantum dot (QD) size statistics were gathered from plane view transmission electron microscopy (TEM) images of PbS on TEM grids. Selection of individual QDs was done manually using ImageJ 1.43U image processing software. Each QD was selected using the elliptical selection tool. The area of each QD was calculated in pixels and converted to nm by setting the scale in ImageJ. Once all the QDs were selected an approximate diameter for each QD could be determined based on the area, assuming a circular shape for the QDs. Standard deviation in diameter and the area fraction could also be determined using this data. Figure S1 shows 30 cycles of PbS with 10 flashes per cycle before and after the grain selection process.

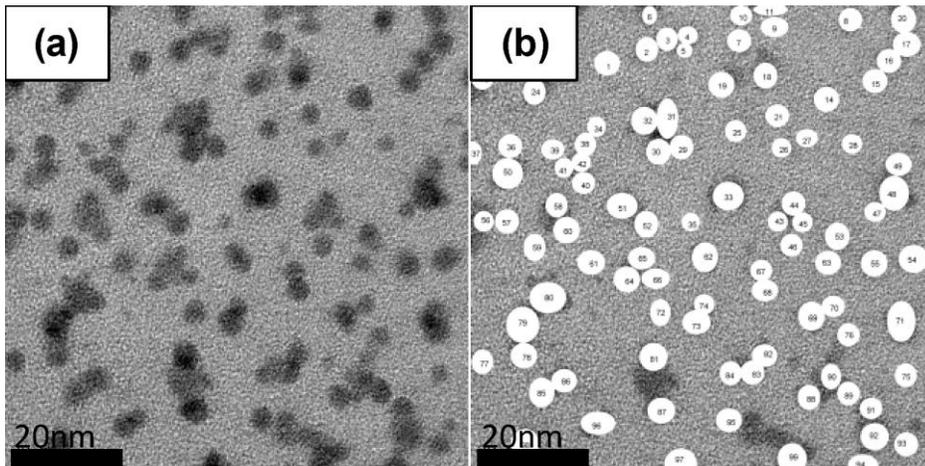


Figure S1. (a) 30 cycles of PbS on SiO₂ TEM grid with 10 flashes per cycles. (b) 30 cycles of PbS on SiO₂ TEM grid with 10 flashes per cycles after grain selection process.

2. Thermal model for power supply design

Prior to fabrication of a setup capable of flash annealing, it was desirable to estimate the energy needed per flash in order to design a flash bulb and power supply capable of achieving annealing temperatures. While the exact desired annealing temperature was application specific, it was estimated from previous *ex situ* annealing experiments an increase of substrate temperature to approximately 500 °C would be an appropriate target. Since the actual experimental setup would be a complicated design with many bodies and complicated geometries that would take significant effort to model, it was first decided to simplify the setup into its essential thermal components so a thermal model could be constructed. It was determined the surfaces that needed to be modelled were the xenon flash bulb, the wafer with a thin atomic layer deposition (ALD) film and the environment. It was decided to model the setup as two parallel discs. Figure S2 shows a schematic of the model that was used.

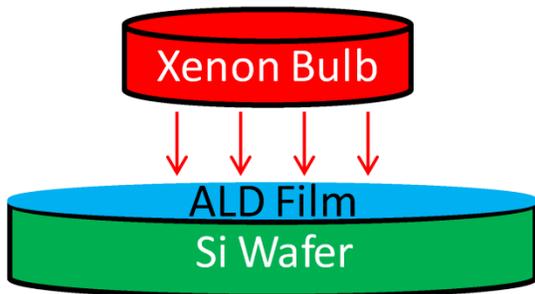


Figure S2. Simplified schematic used for thermal model.

To solve for the wafer temperature as a function of time, a lumped capacitance assumption for the wafer and film was made. The light was modelled as a black body at 5800K which is an appropriate approximation based on the manufacturer's specifications. Based on this model, a differential equation was then constructed using a simple energy balance shown in Equation S1 where ρ_{Film} is film density, c_{Film} is thermal capacitance of the film, V_{Film} is film volume, T_{Film} is

film temperature, F_{Lamp_Film} is the view factor from the lamp to film, σ is the Boltzmann constant, α_{Film} is the absorptivity for the film wafer surface, $A_{Flash\ Lamp}$ is the area of the xenon lamp, $T_{Flash\ Lamp}$ is the flash bulb temperature, F_{Film_Env} is the view factor between the film and the environment, ε_{Film} is the emissivity of the film wafer surface, A_{Wafer} is the area of the wafer surface, T_{Env} is the environment temperature, $T_{Substrate}$ is the ALD chamber temperature, and $R_{Contact}$ is the thermal contact resistance between the wafer and the ALD chamber. In the model convection was neglected as the ALD chamber operates in vacuum.

$$\begin{aligned} \frac{dE}{dt} = \dot{E}_{in} - \dot{E}_{out} &= \rho_{Film} c_{Film} V_{Film} \frac{\partial T_{Film}}{\partial t} \\ &= F_{Lamp_Film} \sigma \alpha_{Film} A_{Flash\ Lamp} (T_{Flash\ Lamp}^4 - T_{Film}^4) \\ &\quad - \left(F_{Film_Env} \sigma \varepsilon_{Film} A_{Wafer} (T_{Film}^4 - T_{Env}^4) + \frac{T_{Film} - T_{Substrate}}{R_{Contact}} \right) \end{aligned}$$

Equation S1. Energy balance equation for surface film temperature.

Equation S1 was solved numerically in Matlab using the RK method. To get the desired annealing effects it was determined the lamp would have to be flashed multiple times. The power supplies for flash lamps are mainly used for pumping lasers and have a standard repetition rate of approximately 100 hertz. The flash times can range from μ s to ms based on the size of the capacitor bank in the power supply. The number of flashes will also depend on the capacitor bank as well as the flash repetition rate. Assuming 20 flashes, and a repetition rate of 100 hertz, it was found 2ms flashes would raise the film temperature from 200 °C to 512 °C as shown in Figure S3.

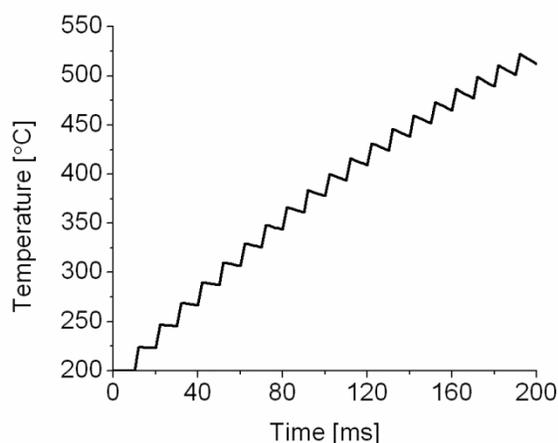


Figure S3. Result of thermal model for film temperature as a function of time with 2 ms flashes pulsed at 100 hertz.

Using the results of this model the specifications of the power supply and bulb were then calculated and the parts were fabricated.

3. TEM and SAD of TiO₂

High-resolution TEM (HRTEM) images and selected-area electron diffraction (SAD) patterns were taken using a FEI Tecnai G2F20 X-TWIN microscope operated at 200 kV. The SAD analysis of the ALD TiO₂ film (control sample) and the FLA ALD TiO₂ film flashed 40 times were performed over an area of $5.72 \times 10^5 \text{ nm}^2$. The SAD of ALD TiO₂ films deposited with 200 cycles (Figure 3c) consists of highly diffused rings typical of amorphous phase. The SAD ring pattern of FLA ALD TiO₂ (Figure 3d) film indicates random orientation of the as-formed TiO₂ crystallites. The polymorphic modification of the observed crystallites was determined by comparing the experimental SAD pattern with a calculated one based on an anatase crystal structure (JCPDS 86-1157) with a unit cell parameter $a = 3.783 \text{ \AA}$. The SAD ring pattern simulation was calculated

using JEMS software (Electron Microscopy Software Java version, P. Stadelmann, 1999-2004). The input parameters were crystal structure data, acceleration voltage of the microscope, camera length and average crystal size. The sharpness of the simulated diffraction rings is influenced by crystallite lateral parameter and becomes significant when particle size is below 10 nm. The experimental SAD pattern matches well with the calculated one for the anatase TiO₂ crystal structure. In addition, the experimental SAD pattern of FLA TiO₂ sample indicates that besides primary anatase phase, a small amount of brookite phase is also present in the film.

4. TEM and SAD of TiO₂ at 200 °C

As there may have been a small, approximately 15 °C ambiguity in the 160 °C sample temperature, a control ALD TiO₂ sample was fabricated with a substrate temperature of 200 °C. The plane-view and SAD TEM of this sample are shown in Figure S4. From Figure S4, it appears the 200 °C control ALD TiO₂ sample is amorphous as-deposited. As this control temperature is higher than the ambiguity in the thermocouple measurement of substrate temperature during growth, we conclude that the crystallization occurred during the annealing step rather than during the ALD reaction.

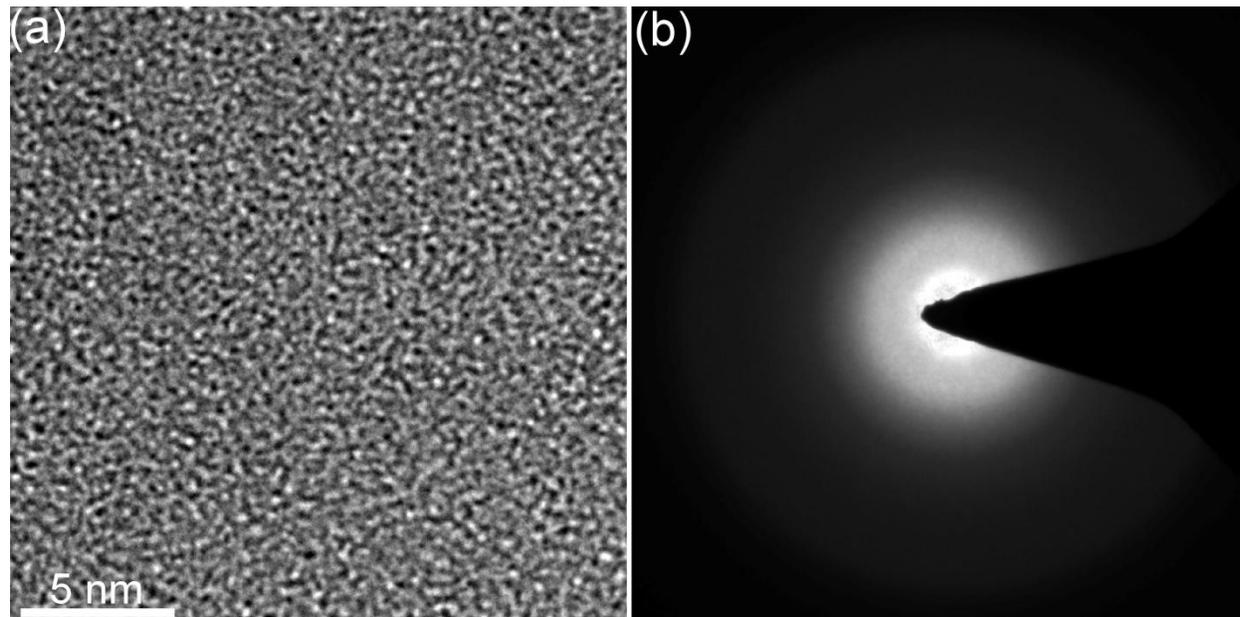


Figure S4. (a) Plane-view and (b) SAD TEM analysis of ALD TiO₂ deposited at 200 °C.