

Supplemental Material

Derivations and explanations of equation (3)

During the initial stage of the epitaxial deposition, the 2D wetting layer grows on substrate, which is due to the lower surface energy of the epitaxial wetting layer as compared to that of substrate. We assume there is an existing wetting layer with the average thickness of h' on the substrate, as shown in Fig. S1 (a). The further growth has only two possibilities: one is keeping further 2D growth (Fig. S1 (c)) and the other one is forming 3D quantum dots on the wetting layer (Fig. S1 (b)).

For the formation of a 3D QD, the change of energy, ΔE_{3D} is mainly determined by two factors, the increase of QDs' side face and elastic relaxation caused by QDs formation. Therefore, ΔE_{3D} can be expressed as

$$\Delta E_{3D} = [\gamma_s A_1 V^{2/3} - \gamma(h) A_2 V^{2/3}] - \omega A_3 \varepsilon_0^2 V \quad (S1)$$

where γ_s is the surface energy density of the QD's side facet, A_1 , A_2 and A_3 are the shape factors which are determined by the shape of QD, ω is a elastic constant which $\omega = Y(1+\nu)/(1-\nu)$ in which Y and ν are Young's modulus and Poisson ratio, ε_0 is the strain mismatch between epitaxial material and substrate, V is the volume of the QD. When the surface of substrate is not planar, the thickness of wetting layer is usually uneven. We defined the wetting layer which is covered by the QD, i.e. underneath the QD, has the thickness of h , as shown in Fig. S1 (b). In this case, $\gamma(h)$ represent the surface energy density of the wetting layer covered by the QD. The first term of Eq. (S1) represents the change of surface energy caused by the QD formation, and the second term is the elastic relaxation energy caused by the QD

formation.

For the further 2D growth, the change of energy, ΔE_{2D} is mainly caused by the increase of thickness of wetting lay. We assume the further growth 2D layer reaches the average thickness of h'' , and the surface area of the wetting layer is A . Therefore, ΔE_{2D} can be expressed as

$$\Delta E_{2D} = A[\gamma(h'') - \gamma(h')] \quad (\text{S2})$$

In order to compare the two energy changes caused by further 2D growth and the 3D QD formation, the deposited amounts of the two cases should be equal to each other, i.e. the volume of QD should be equal to the volume of the wetting layer caused by further 2D growth. In other words,

$$V = A(h'' - h') \quad (\text{S3})$$

i.e.

$$h'' = h' + V/A \quad (\text{S4})$$

Likewise, in order to compare the two energy changes caused by further 2D growth and the 3D QD formation, the surface area of the wetting layer in Eq. (S2) is determined by the density of QDs (the number of QDs per unit area). If we assume that the density of QDs is k , the surface area of the wetting layer in Eq. (S2) should have the relation that $A = 1/k$. Therefore, $1/A$ represents the density of QDs, and A is the area of occupancy by a single QD, as shown in Fig. (S2).

Combining Eqs. (S1), (S2) and (S3), we can obtain that the total energy difference between the two growth modes can be written as

$$\Delta E = [\gamma_s A_1 V^{2/3} - \gamma(h) A_2 V^{2/3}] - \omega A_3 \varepsilon_0^2 V - A[\gamma(h' + V/A) - \gamma(h')] \quad (\text{S5})$$

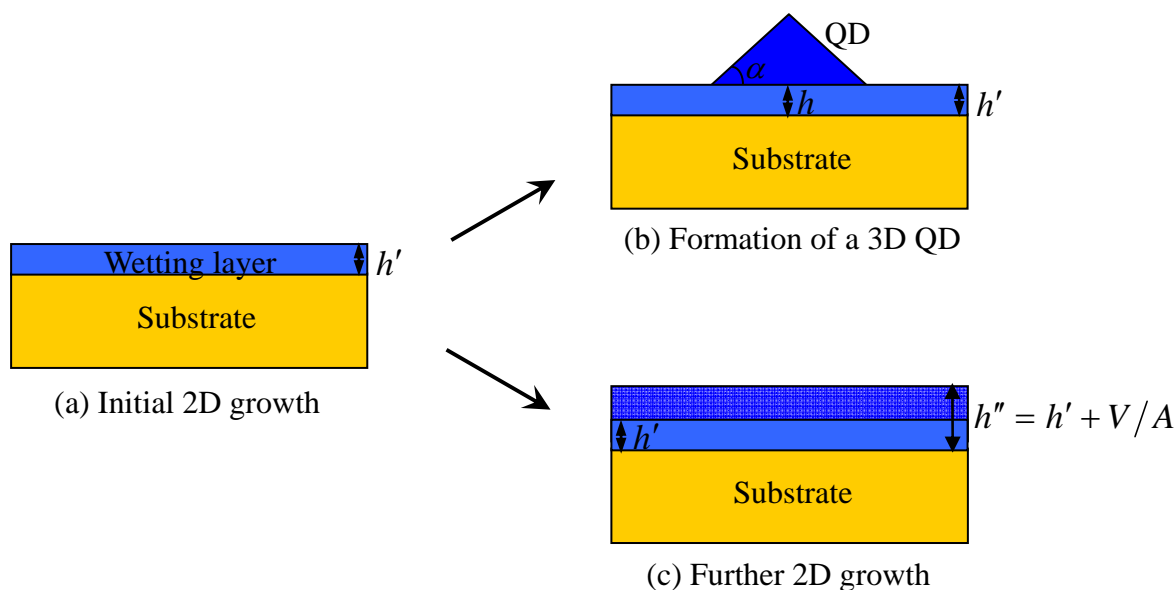


Fig. S1 Schematic illustration of two growth modes on initial 2D wetting layer growth (a): formation of a 3D QD (b) and Further 2D growth (c). In order to compare the two energy changes caused by the two growth modes, the deposited amounts of the two cases should be equal to each other. Therefore, $h'' = h' + V/A$.

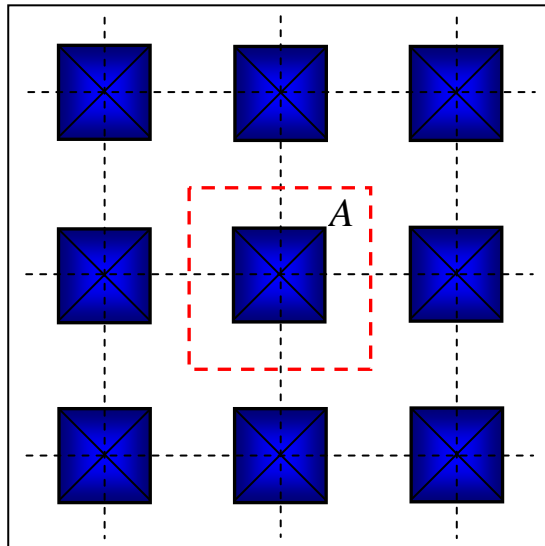


Fig. S2 Schematic illustration of the QDs' array. In order to compare the two energy changes caused by the two growth modes, the surface area of the wetting layer in Eq. (S2), A , should be equal to the area of occupancy by a single QD, as shown by the regions inside the red dashed line. Therefore, $A = 1/k$, where k is the density of QDs (the number of QDs per unit area).