RESEARCH ARTICLE

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Dynamical evolution of Ge quantum dots on Si(111): From island formation to high temperature decay

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Abstract

Heteroepitaxial growth is a process of profound fundamental importance as well as an avenue to realize nanostructures such as Ge/Si quantum dots (QDs), with appealing properties for applications in opto- and nanoelectronics. However, controlling the Ge/Si QD size, shape, and composition remains a major obstacle to their practical implementation. Here, Ge nanostructures on Si(111) were investigated in situ and in real-time by low energy electron microscopy (LEEM), enabling the observation of the transition from wetting layer formation to 3D island growth and decay. The island size, shape, and distribution depend strongly on the growth temperature. As the deposition temperature increases, the islands become larger and sparser, consistent with Brownian nucleation and capture dynamics. At 550°C, two distinct Ge/Si nanostructures are formed with bright and dark appearances that correspond to flat, atoll-like and tall, faceted islands, respectively. During annealing, the faceted islands increase in size at the expense of the flat ones, indicating that the faceted islands are thermodynamically more stable. In contrast, triangular islands with uniform morphology are obtained from deposition at 600°C, suggesting that the growth more closely follows the ideal shape. During annealing, the islands formed at 600°C initially show no change in morphology and size and then rupture simultaneously, signaling a homogeneous chemical potential of the islands. These observations reveal the role of dynamics and energetics in the evolution of Ge/Si QDs, which can serve as a step towards the precise control over the Ge nanostructure size, shape, composition, and distribution on Si(111).

KEYWORDS
epitaxial growth, GeSi heterostructures, low energy electron microscopy

1 | INTRODUCTION

Since the early 90s, the growth of germanium quantum dots (QDs) on silicon surfaces has been a classical example of semiconductor heteroepitaxial growth.[1,2] Once a critical thickness of a few monolayers (ML) of Ge has been deposited, the 4.2% lattice mismatch between the two elements causes a strain-induced transition of the Ge wetting layer, which is replaced by three-dimensional (3D) islands following a Stranski–Krastanow growth model.[3–6] The

limited size of these Ge/Si nanostructures confers interesting properties that are promising for a variety of device applications in opto- and nanoelectronics.[1,7–10]

To achieve full control of the QD properties means being able to tailor their shape, size, composition, and positioning, which in turn requires a comprehensive understanding of the effect of the growth parameters and the interplay between kinetic and thermodynamic factors.[1,11–16] Typically, insights on the evolution of Ge/Si islands were pieced together by observation through traditional surface science techniques, such as scanning probe microscopy (SPM)[5,6,11] and electron diffraction,[17–19] which were used to acquire snapshots of the Ge/Si islands at different growth steps. The Si surface orientation was found to play a primary role, as it defines the QD shape and structure.[3] For instance, Ge islands formed on Si(111) present various morphologies, including truncated pyramids that are faceted and tall, and atoll-like shapes that are flat. The faceting of the truncated pyramidal islands is attributed to strain release mechanisms via the introduction of dislocations, defects, or progressive Ge/Si intermixing and alloying.[3,14,20,21] The distribution of Ge QDs on Si(111) has also been extensively investigated, showing a notable tendency towards self-ordering.[1,22] However, previous studies involving measurements at specific stages of growth were unable to address questions such as the kinetics of the QD growth and the stability of the resulting nanostructures, which are crucial to tailor the QD size and shape. Therefore, we conducted a real-time investigation of the growth process of Ge QDs on Si(111) using low energy electron microscopy (LEEM), which combines high resolution and fast acquisition to make it possible to study the evolution of nanostructures at high temperatures.[16,22,23]

Here, we studied the growth of Ge/Si(111) QDs at three different deposition temperatures, between 450°C and 600°C, and observed their subsequent decay during annealing up to 800°C. Our investigation confirms that surface temperature is the determining factor in defining QD size, with larger and sparser islands obtained at higher substrate temperature, in accordance with a predominant mechanism of Brownian nucleation and capture dynamics. The real-time investigation of Ge/Si(111) at 550°C revealed the formation of two distinct kinds of islands, distinguished by their contrast in LEEM images, areal growth rates, and decay mechanisms. The darker, faceted islands exhibited slower areal growth than the bright, flat islands. During annealing, the faceted islands were observed to grow at the expense of the flat ones, implying higher thermodynamic stability of the faceted islands. At 600°C, a single type of island with a largely triangular shape was obtained, suggesting that the metastable flat islands are not favored at 600°C. The formation of a single morphology is consistent with the similar thermodynamic stability of these triangular islands observed in the annealing experiments, where the islands initially showed no change in size and morphology before rupturing together. This study provides an improved understanding of the interplay of kinetics and thermodynamics in the growth and decay of Ge QDs on Si(111), enabling greater control over their morphology, size, and distribution through the choice of deposition and annealing temperatures.

2 | RESULTS

To follow the evolution of the Ge/Si QD growth, LEEM images were recorded during deposition of Ge on Si(111) at three different temperatures, 450°C, 550°C, and 600°C, using the same deposition rate of ∼0.2 ML/min for ∼43 min. These images were obtained at start voltages (which defines the incident electron energy with respect to the sample surface) in the range of 0–10 V in bright-field mode. Two mechanisms can contribute to the imaged electron intensity, with the LEEM mode dominant at higher electron energies and the mirror electron microscopy (MEM) mode dominant at the low end of this range. In LEEM, the image contrast results from electron diffraction at the sample surface, showing surfaces parallel to the substrate as bright and faceted steps as dark, whereas the MEM image contrast results from electron reflectivity which depends on the local work function and the topography.[16,24–26] We refer to images acquired with electron energies between 0 and 2 eV as LEEM/MEM images because both modes contribute to image formation in this range. During the initial phase of deposition, Ge forms a uniform wetting layer with no evident features on the surface in the LEEM/MEM movies (see Movie S1–S3).[27] As the film thickness increases, small dots start to appear after 20 min, corresponding to the nucleation of 3D Ge/Si islands (Figure S1, Movie S1–S3). The sizes of the dots, as well as their shapes and distribution, are strongly dependant on the Si surface temperature.

LEEM/MEM images were obtained near the end of deposition at three different temperatures (Figure 1). Similar features were exhibited in LEEM images acquired at higher start voltages after the end of the deposition, as shown in Figure S2. At 450°C, the surface is punctuated by a dense array of QDs (Figure 1A, Figure S2A, and Table 1), which range in appearance from dark to bright, probably due to variation in the heights and shapes of the islands. When the sample temperature is increased to 550°C, the QDs observed in LEEM/MEM images become much less dense and larger in area by one order of magnitude compared to 450°C (Figure 1B, Figure S2B, and Table 1). Two types of islands with slightly irregular shapes were observed at this temperature: darker features indicative of faceted, tall islands and brighter, larger features corresponding to flatter, atoll-like islands.[16] Increasing the deposition temperature to 600°C yields even larger and sparser islands with dark edges and largely triangular shapes (Figure 1C, Figure S2C, and Table 1). Our observations of the temperature dependence of the island size, morphology, and density are consistent with a previous scanning reflection electron microscopy (SREM) study.[28]

Plotting the area of a selection of QDs against time for the samples prepared at 450°C, 550°C, and 600°C (Figure 2) shows that the growth rates of the islands are temperature dependent. The growth rate is uniformly low at 450°C (Figure 2A,B). Two types of islands at 550°C, faceted and flat, nucleate simultaneously and follow different growth rates (Figure 2C,D), with the flat ones growing considerably faster in area. At 600°C, the islands exhibit a similar, fast growth rate, with most of them retaining a triangular shape during growth (Figure 2E,F, Movie S3). A growth law of the
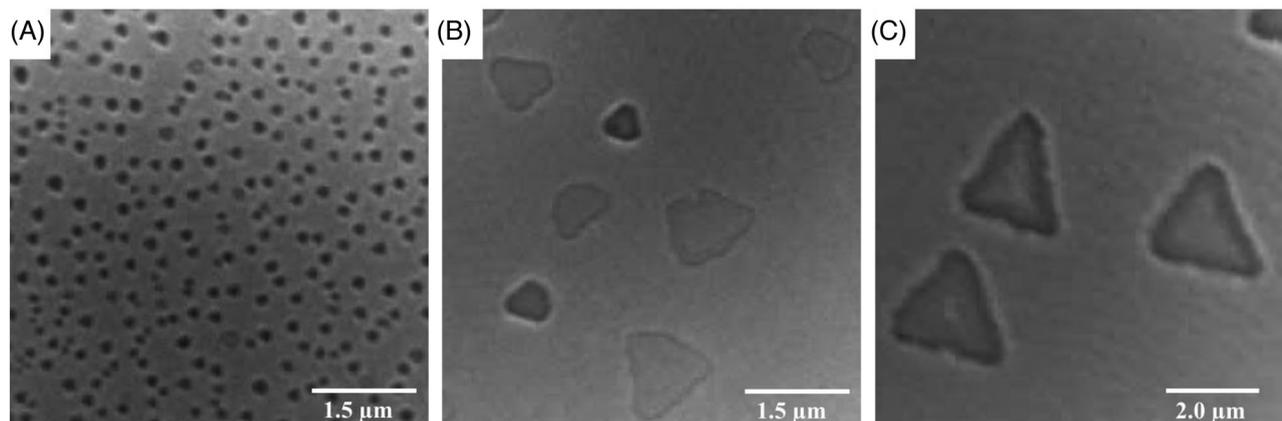
FIGURE 1 Ge/Si QDs obtained by the end of deposition at different surface temperatures: (A) QDs formed at 450°C showing smaller features with varied contrasts. Start voltage: 0.4 V. (B) QDs formed at 550°C showing bigger and sparser islands than in (A) with two different types of islands identifiable by their sizes and contrasts. Start voltage: 0.1 V. (C) QDs formed at 600°C showing bigger islands with lesser density and variations in contrast compared to (A) and (B). Start voltage: 0 V

TABLE 1 Details on the samples investigated

Deposition temperature (°C)	Average island size (μm)	Average island density (μm⁻²)
450	0.18 ± 0.04	7.3 ± 1.3
550	0.98 ± 0.32	0.15 ± 0.025
600	2.48 ± 0.50	0.027 ± 0.004

kind $A = Bt^x$ with $x < 1$ can be inferred from the data, where A is the area of the island, t is the time, x is a function of temperature and island thickness, and B is a pre-factor.[29]

To gain insights into the relative stabilities of the Ge/Si QDs, annealing experiments were performed for each sample after deposition. Samples were heated from an initial temperature around 200°C to a maximum temperature of 800°C (see temperature profiles in Figure S3). Animations of the LEEM/MEM observations during annealing of the three samples are shown in Movie S4–S6. In general, the disintegration of islands during annealing is associated with an increase in surface diffusion of Si and Ge atoms and the strain relaxations that occur with an increase in surface temperature.[30]

For the sample prepared at 450°C, raising the surface temperature caused some of the islands to increase in size at the expense of their neighbors in an Ostwald-ripening process (Figure 3A). Once the surface temperature crossed over 650°C, the larger islands started to disintegrate into less compact structures and the smaller islands vanished completely (Figure 3B,C). For the sample prepared at 550°C, the two types of Ge/Si islands followed different decay pathways. Starting at 600°C, the faceted islands became slightly larger, whereas the larger flat islands started to shrink and leave a darkened footprint on the surface, which matches their former dimensions (Figure 3D). At 700°C, holes began to form in the faceted islands, whereas the footprints left by the flat islands started to disappear (Figure 3E). At 780°C, only the footprints of the faceted islands remained, with no sign of the previous flat islands (Figure 3F). Finally, for the sample prepared at 600°C, the largely triangular islands did not show any change in size and morphology until 750°C, above which they started to rupture (Figure 3G–I).

3 | DISCUSSION

The clear observation of an increase in the island area and a decrease in island density at higher growth temperatures (Figure 1, Table 1) is consistent with previous studies using LEEM, SREM, and SPM.[22,28,31] The growth of Ge/Si QDs is often considered in terms of a model of Brownian nucleation and captures dynamics in which random diffusion of Ge and Si adatoms drive nucleation and growth. In particular, the smaller diffusion lengths of adatoms at lower temperatures lead to a higher concentration and smaller size of the QDs.[1,22,32,33] However, this kinetic model does not fully describe the QD growth for Ge/Si(111), as the size of individual nanostructures correlates weakly with the area around each island (Voronoi cell area), suggesting that energetic factors may also influence the process of mass-sharing among coexisting nanostructures.[1,22]

The real-time LEEM/MEM characterization of the sample prepared at 550°C provides two key insights into the Ge/Si QD areal growth and stability. First, the faceted and flat islands nucleated simultaneously and grew at distinct rates (Figure 2C,D). The simultaneous nucleation of both kinds of islands is consistent with the earlier SREM study by Shklaev et al., and a similar critical island size was measured at different temperatures indicating a comparable nucleation process for both.[28] The branching of the islands into two distinct morphologies could arise from the random inclusion of dislocations and defects during the initial stages of growth.[28] To evaluate whether the flat and faceted islands exhibit different *volumetric* growth rates, the difference in height must be taken into account. Therefore, SPM measurements of islands produced under comparable conditions could provide additional insights into the island growth, for example whether the faceted islands grow more slowly due to an Ehrlich-Schwoebel barrier to adatom diffusion onto the facets.[34,35] Annealing of the sample prepared at 550°C (Figure 3D–F) showed that the faceted islands had higher thermal stability than the flat islands, and the faceted islands grew at the expense of the flat islands during the initial stages of annealing. These observations indicate the faceted islands have a lower chemical potential compared to the flat islands, and that the formation of the flat islands is the result of kinetic factors.
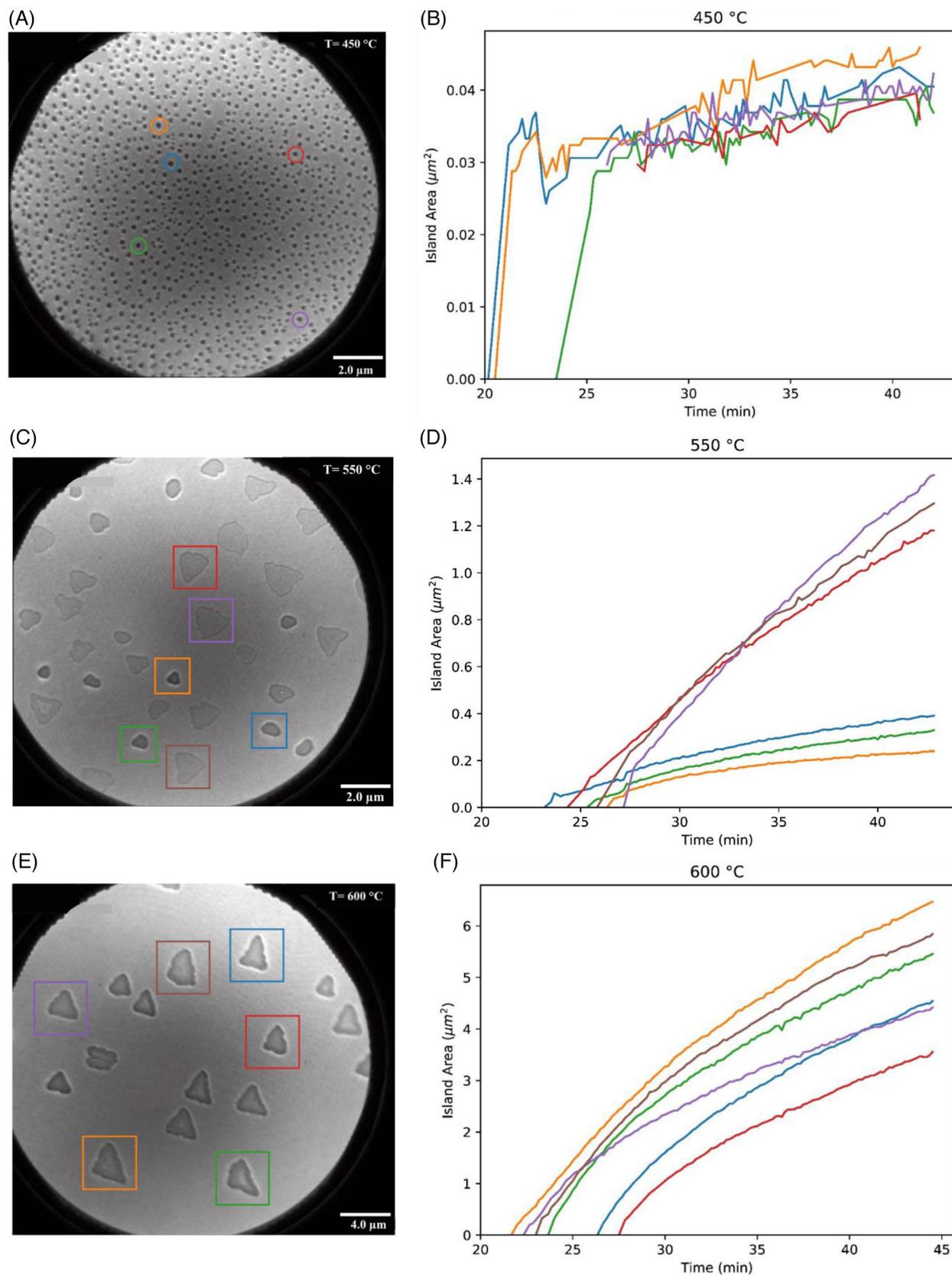
FIGURE 2 Area of Ge/Si QDs during deposition. LEEM/MEM images on the left show the Ge/Si QDs near the end of growth at each temperature, and line graphs on the right show the growth of the outlined QDs prior to each image. (A, B) LEEM/MEM image and areal growth plot for deposition at 450°C. Start voltage: 0.4 V. (C, D) LEEM/MEM image and areal growth plot for deposition at 550°C, showing distinct growth rates for each type of island. Start voltage: 0.1 V. (E, F) LEEM/MEM image and areal growth plot for deposition at 600°C. Start voltage: 0 V
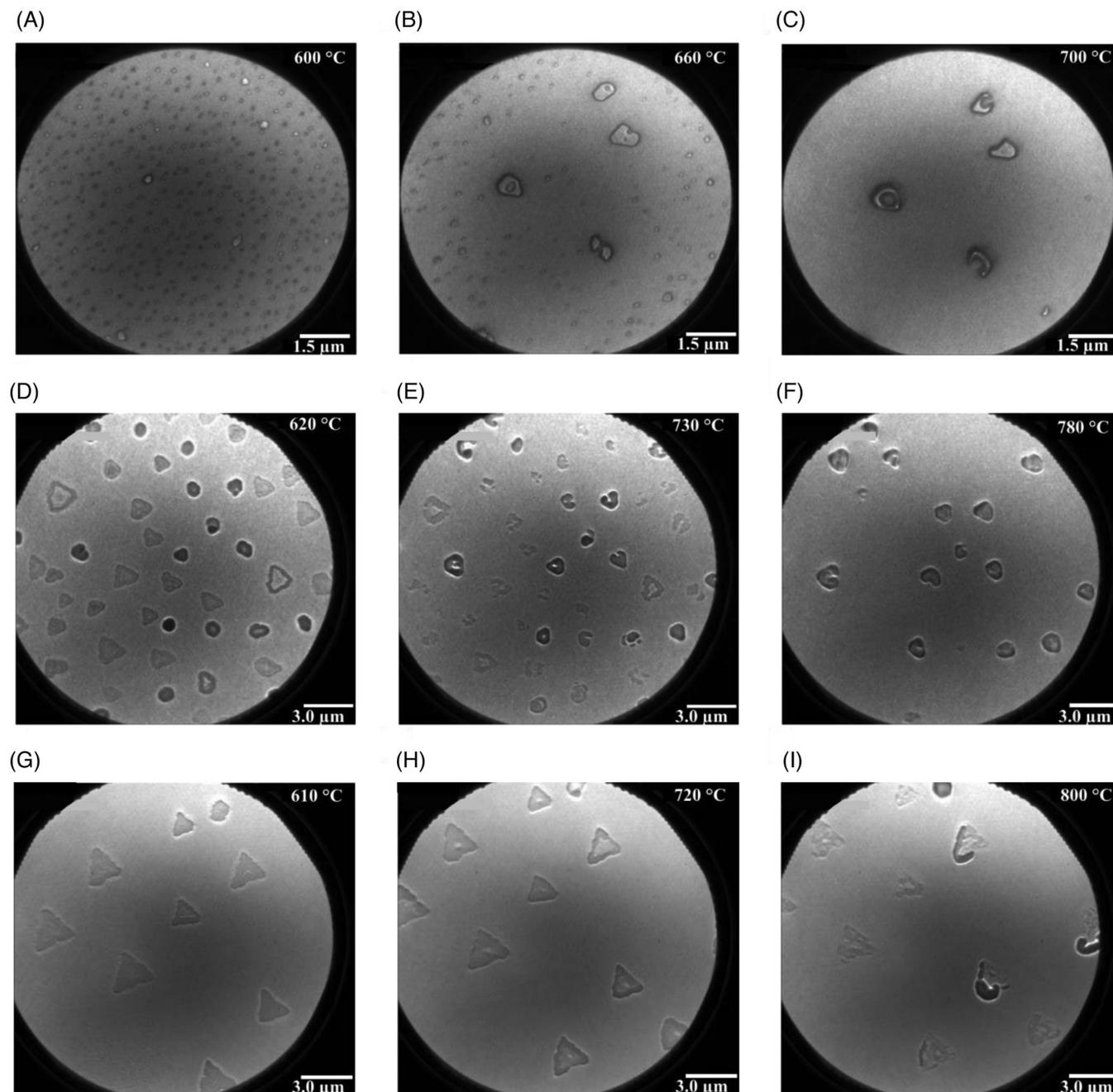
FIGURE 3 LEEM/MEM images during the annealing of Ge/Si QDs prepared at 450°C (A–C, start voltage: 7.2 V), 550°C (D–F, start voltage: 0.1 V), and 600°C (G–I, start voltage: 0.2 V). Surface temperature is given at the top right corner of each image

Islands with more homogenous morphology were obtained at 600°C (Figures 1C and 2E), whereas the formation of the metastable flat islands is suppressed. A higher temperature increases the rate of disaggregation from the flat islands to the wetting layer, which is likely to favor the growth of the more stable faceted islands. The predominance of the faceted islands at 600°C could also be influenced by the intermixing of Ge and Si, which previous studies have shown is greater at higher temperatures.[12,16,36,37] An increase in the extent of intermixing would promote diffusion between the islands, wetting layer, and Si substrate. This could disfavor the formation of dislocations, thereby preventing the flat islands formation at higher temperatures.[28] In addition, during the annealing experiments of this sample, the islands remained stable up to 750°C and then ruptured at the same time (Figure 3G–I), showing higher stability of the islands compared to the sam-

ples prepared at lower temperatures. This also shows that the Ge/Si islands grown at 600°C have similar chemical potentials and that the QD growth occurs in a regime of predominant thermodynamic control. In contrast, for the samples prepared at 450°C and 550°C, certain islands gained mass from other islands during the early stages of annealing (Figure 3A–F), implying more heterogeneous chemical potentials of the islands.

Our observations suggest that the size and number density of the QDs can be tuned by varying the growth temperature and deposition time (Figure 2, Table 1, and Figure S1). Islands of more uniform size, shape, and stability were obtained at 600°C, although they are substantially larger than the scale yielding size-dependent effects. To avoid the undesirable formation of multiple island morphologies, one strategy is to use a high growth temperature. In addition to the QD

size, shape, and density, the growth temperature also affects the Ge–Si composition of the QDs.[16,36] A second strategy is to reduce the Ge deposition rate, which was not examined in the present study but is expected to yield more uniform and less-dense islands.[6]

4 | CONCLUSIONS AND PERSPECTIVES

We carried out a detailed real-time LEEM/MEM investigation of the evolution of Ge/Si QDs during the growth at three different deposition temperatures and their subsequent annealing. The Ge/Si islands become larger and sparser as the deposition temperature increases, showing the influence of Brownian nucleation and capture dynamics. Both faceted and flat islands form at 550°C, which our real-time measurements reveal differ in their areal growth rates. The faceted islands have higher thermodynamic stability since they were observed to grow at the expense of vanishing flat islands during annealing experiments. Triangular islands were formed at 600°C, whose uniformity is indicative of growth following closer to the ideal shape. The homogenous chemical potential of the islands formed at 600°C is evidenced by the simultaneous rupturing of islands without prior changes in shape or size during annealing experiments. Thus, our results reaffirm that both dynamic and energetic factors play a significant role in determining the size, morphology, and self-ordering of the QDs at different growth and annealing temperatures. Overall, the use of higher growth temperatures is recommended to obtain Ge/Si islands with more homogenous size and morphology, although further optimization would be required to produce smaller Ge/Si islands exhibiting size-dependent effects.

To gain insights into the origin of the two distinct islands at 550°C and their growth rates calls for real-time studies using higher resolution techniques such as scanning tunneling microscopy and atomic force microscopy, capable of monitoring the initial growth stages when the branching occurs. In addition, real-time investigations of Ge/Si QD evolution using composition-sensitive techniques such as photoemission electron microscopy[36,38] provides an improved understanding of the intermixing and alloying of Si and Ge atoms during growth and annealing at different temperatures, taking the community one step closer to the predictive control over the synthesis of Ge/Si QDs.[14]

5 | EXPERIMENTAL SECTION

The experiments were performed using a LEEM from Elmitec company. The surface imaging can be done with a lateral resolution of ∼10 nm and a time resolution of a few tens of milliseconds in LEEM and MEM operating conditions. The microscope was operated in a bright-field mode to obtain the images of Ge growth and decay at start voltages in the 0–10 V range. At higher start voltages (LEEM mode), the island tops and the wetting layer which are parallel to the substrate surface plane appear bright and the highly stepped facets at the sides of the Ge/Si islands appear dark due electron diffraction. At lower voltages (MEM mode), the contrast difference arises from electron reflectivity, which is a function of topography and local work function.[16,24–26]

The Si(111) samples were degassed at 600°C for several hours. Then they were flashed up to 1200°C repeatedly until a sharp 1×1 to 7×7 transition was observed while cooling down the surface to room temperature. Three different samples were prepared by in situ deposition of Ge on Si(111) by molecular beam epitaxy (MBE) at a growth rate of ∼0.2 ML/min for ∼43 min, after stabilizing the sample temperature at 450°C, 550°C, and 600°C.

CONFLICT OF INTEREST
The authors declare no conflict of interest.

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

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