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Ali Oguz Er
Old Dominion University

Wei Ren
Old Dominion University

Hani E. Elsayed-Ali
Old Dominion University, helsayed@odu.edu

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Low temperature epitaxial growth of Ge quantum dot on Si(100)-(2×1) by femtosecond laser excitation

Ali Oguz Er,[1] Wei Ren,[2] and Hani E. Elsayed-Ali[2,a)]
[1]*Department of Physics, Old Dominion University, Norfolk, Virginia 23529, USA*
[2]*Department of Electrical and Computer Engineering and the Applied Research Center, Old Dominion University, Norfolk, Virginia 23529, USA*

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Low temperature epitaxy of Ge quantum dots on Si(100)-(2×1) by femtosecond pulsed laser deposition under femtosecond laser excitation was investigated. Reflection high-energy electron diffraction and atomic force microscopy were used to analyze the growth mode and morphology. Epitaxial growth was achieved at ~70 °C by using femtosecond laser excitation of the substrate. A purely electronic mechanism of enhanced surface diffusion of the Ge adatoms is proposed. © *2011 American Institute of Physics*. [doi:10.1063/1.3537813]

Growth of Ge on Si is a classical model of the Stranski-Krastanov growth-mode, also known as layer-plus island, where growth starts in a uniform layer-by-layer up to ~3 monolayers (ML). In Ge on Si growth, one way to suppress misfit dislocations is by lowering the substrate temperature.[1] To lower the epitaxial growth temperature, extrinsic assistance by energetic particles, such as ions, electrons, and photons, have been used to promote the migration of adsorbed atoms at the surface.[2] Pulsed laser induced electronic processes leading to surface structural modifications have been shown to occur when the laser intensity is significantly below the melt threshold.[3–5] We have recently studied the effects of nanosecond pulsed laser-induced electronic excitations on the self-assembly of Ge quantum dot (QD) on vicinal Si(100)-(2×1) grown by pulsed laser deposition.[2,6] Electronic excitations, due to laser irradiation of the Si substrate and the Ge film during growth, were shown to decrease the roughness of films grown at room temperature and ~120 °C. At this temperature, the grown films were nonepitaxial. Electronic excitation resulted in the formation of an epitaxial wetting layer and crystalline Ge QD at ~260 °C, a temperature at which no crystalline QD formed without excitation under the same deposition conditions.[2,6]

Ge quantum dots on Si(100) were grown in an ultrahigh vacuum (UHV) chamber (base pressure $\sim 7.0\times 10^{-10}$ Torr) by femtosecond pulsed laser deposition. The Si(100) substrates (dimensions of 2.0 mm×10 mm×0.5 mm *p*-type boron doped, and resistivity 0.060–0.075 Ω cm, miscut angle 0.38°) were chemically etched by using a modified Shiraki method before being loaded into the UHV chamber.[2] The Ge target was a 2 in. disk, 0.5 mm thick, undoped *n*-type, with a resistivity of 45–58.7 Ω cm. The vacuum system was then pumped down, baked at 150 °C for at least 24 h, and sample was degassed overnight at ~650 °C and then flashed to 1200 °C for ~60 s by direct heating to remove native oxides and carbon and to form (2×1) reconstructed surface. The target was rotated at 5 rpm to reduce the particulates formation. The surface temperature was initially measured using a combination of a type K (chromel-alumel) thermocouple mechanically attached to the substrate surface and a pyrometer. The deposition was carried out with a chirped pulse amplified Ti:sapphire laser. Output pulse has ~60 fs pulse width with center wavelength ~800 nm operating at 1 kHz repletion rate. The femtosecond laser was split into ablation and excitation beams of nonequal powers by means of half-wave plate and a thin film polarization beam splitter. The p-polarized ablation beam was focused on the rotating Ge target, resulting in a laser energy density of ~ 0.2 J/cm^2. The s-polarized excitation passed through another half-wave plate, used to change its polarization, and then was directed onto substrate. This laser was used to excite the substrate during deposition with an energy density of ~30 mJ/cm^2. A well-collimated 20 keV electron beam with a spot size <90 μm diameter was used to probe the growth dynamics. A phosphor screen displayed the electron diffraction pattern which was recorded by a charge-coupled device camera. Sample-to-target distance was ~10 cm. The final film thickness measurement was done by a spectroscopic ellipsometer.

To study the effect of using excitation laser, a series of Ge samples was prepared at different substrate temperatures. The film growth was accomplished with and without the excitation laser, but otherwise, under the same conditions.

We have first deposited Ge on Si(100)-(2×1) with the excitation laser and constantly decreased the substrate temperature until the point where the reflection high-energy electron diffraction (RHEED) diffraction spots decayed completely with coverage. Figure 1(a) shows the reconstructed Si(100)-(2×1) surface, consisting of spots aligned on Laue circles. Upon initiating growth with the substrate temperature at 70 °C, the intensity of the RHEED spots drops immediately due to formation of many small two-dimensional islands and the pattern indicates that the growth starts epitaxially, as shown in Fig. 1(b). At ~4 ML coverage, the intensity of the diffraction spots starts to decrease and an elongated transmission pattern evolves with further Ge growth, corresponding to formation of hut clusters, as shown in Fig. 1(c). With further coverage, the elongation decreases and round transmission spots start to form at ~6 ML coverage, shown in Fig. 1(d). Those elongated streaks became shorter, shown in Fig. 1(e), due to transformation of huts into domes at ~8 ML coverage. The deposition was stopped at ~12 ML coverage. The RHEED pattern at that coverage,

a)Author to whom correspondence should be addressed. Electronic mail: helsayed@odu.edu.

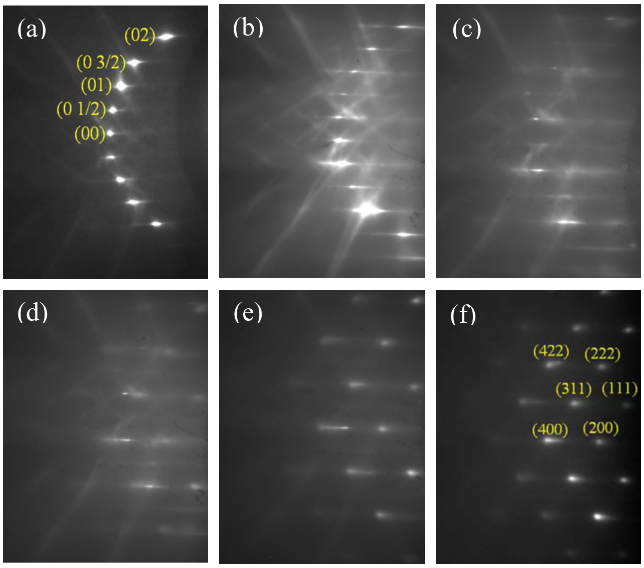

FIG. 1. (Color online) RHEED patterns during Ge deposition on Si(100)-(2×1) at ~70 °C with excitation laser of ~30 mJ/cm^2. (a) Si(100)-(2×1) reconstructed surface, (b) during formation of Ge wetting layer, (c) the RHEED spot intensity decreased at ~4 ML, (d) some round transmission spots started to form at ~6 ML, (e) elongated streaks transformed into round spots at ~8 ML, and (f) final transmission pattern at ~12 ML.

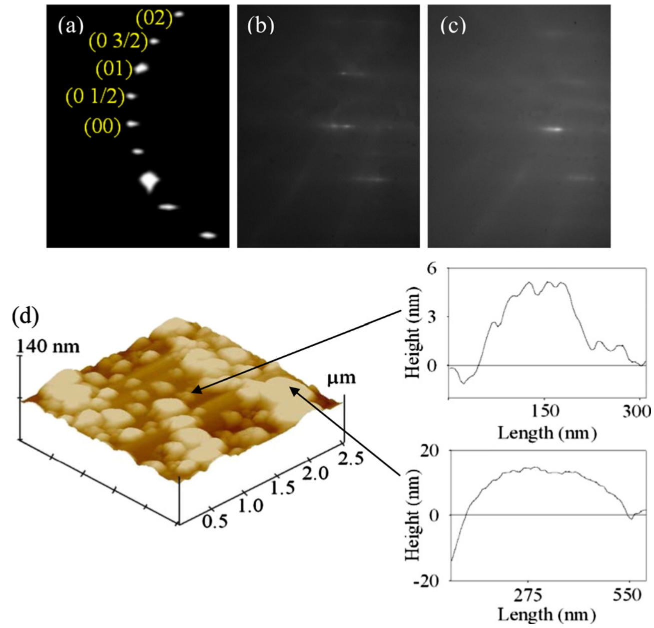

FIG. 3. (Color online) RHEED pattern and AFM image of Ge quantum dots on Si(100)-(2×1) grown at 70 °C without the excitation laser. (a) Clean Si(100)-2×1 surface (b) after ~7 ML, (c) after ~12 ML, and (d) topographic AFM image and line scans of two clusters.

shown in Fig. 1(f), obtained ~12 ML, shows well-defined spotty transmission pattern indicative epitaxial QD growth. These spots are not falling on Laue circles and result from transmission of electrons through faceted islands. The AFM image in Fig. 2(a) shows islands with different sizes. Most of the islands are dome shaped with well-defined facets. Line scans of two islands, taken along their major axes, are shown

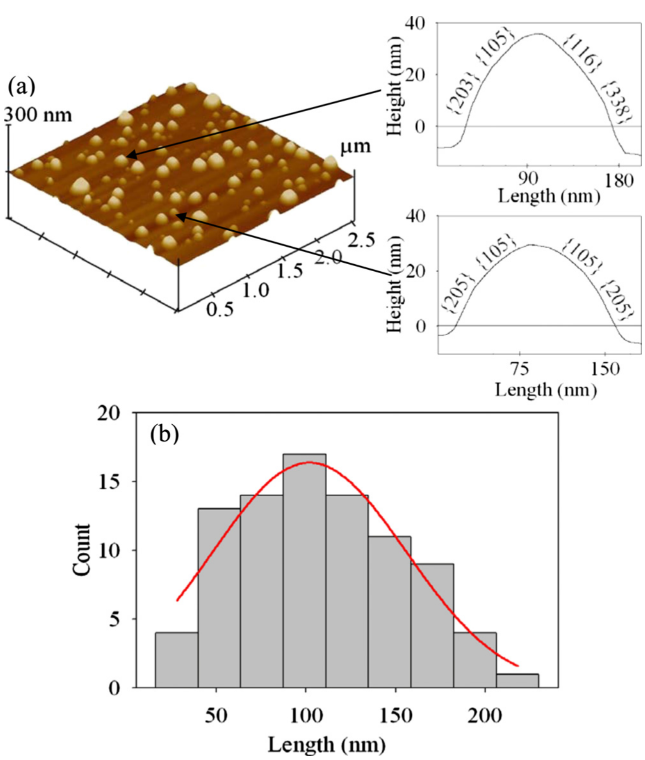

FIG. 2. (Color online) (a) AFM image and line scans of two islands, and (b) its corresponding Ge quantum dot size distribution for film grown at 70 °C with excitation.

in Fig. 2(a). The facets were identified by the angle they make with {100} plane. The average height for this AFM image is ~30 nm and island density is ~1.5×10^9 cm^{-2}. Size distribution graph, shown in Fig. 2(b), indicated that the average QD length is ~110 nm while the most expected size is ~100 nm.

For fs-PLD of Ge on Si(100)-(2×1) at 70 °C without laser excitation, some of the diffraction spots became dim after ~7 ML coverage, and almost no pattern appeared after 12 ML, indicating loss of long-range order on the surface as shown in Fig. 3. At this low temperature, formation of a rough disordered surface is expected due low surface diffusion coefficient. The AFM image in Fig. 3(d) shows a collection of nonuniform clusters as generally expected for low temperature heteroepitaxial growth. Line scans of two clusters show irregular shape with no defined facets. For deposition at a substrate temperature of 150 °C, while applying the same excitation laser energy density, AFM observations showed decrease in island density and increase in average QD size and height compared to that at 70 °C. The same trend was observed as the substrate temperature is further increased to 280 °C.

When the Si sample is irradiated by the 800 nm (1.55 eV) femtosecond laser, the initial effect is to generate electron-hole pairs with excess energies of no more than 0.43 eV. The energy density of the fs laser pulses used in the present work is well below the damage threshold of Si, which is ~200 mJ/cm^2.[7] Thermal effects can be readily ruled out by the fact that the used energy densities are almost an order of magnitude lower than that for ablation and, more importantly, is that the temperature excursion occurs only in a subnanosecond time scale due to heat diffusion to the bulk. Since the surface processes affecting growth occur at a much slower time scale, thermal effects of the laser irradiating the substrate are negligible. Also, the temperature build-up on the surface due to the repetitive nature of excitation is too small to cause any measurable effect on Si growth. We have used a spectroscopic ellipsometer to measure Ge film coverage with and without laser excitation for all other deposition conditions kept the same. For samples, without laser excitation at 70 °C, the Ge thickness was 16.7 ± 1.0 Å, while with excitation the Ge thickness it was 17.4 ± 0.5 Å. Therefore, within the experimental error, there appears to be no effect on Ge film thickness due to the excitation laser, and any atom desorption by electronic excitation is too small to affect the grown film.

The interaction of electrons or photons with semiconductor surfaces can cause emission of ions or neutrals due to electronic excitation leading to surface bond breaking.[8] A two-hole localization (THL) model was proposed for this mechanism.[9] The primary assumption in the THL model is that surface bond rupture leading to neutral-atom desorption can be induced by strong lattice relaxations associated with localization of two valence holes on the same surface bond.[10] The localization of the second hole causes strong vibrations of the surface atom, which could lead to bond breaking. Due to this transient strong lattice vibration (phonon kick), these atoms could be ejected with a distribution of translational energies that starts from a given threshold.[5] Vibrational relaxation after electronic excitation would lead to emission of many phonons. As the energy of the surface atoms increases, the bonded atoms vibrate more strongly. If the phonon kick perpendicular to the surface imparted to a surface atom is not sufficient to cause desorption, the enhanced vibrational motion could lead to increased surface diffusion. In fs PLD, plume pulse width is on the order of microseconds, similar to ns PLD.[11] The longest lifetime of holes in n-type germanium at 300 K is longer than 1 ms. Therefore the holes generated by the excitation pulse are present during the nucleation and growth processes.

The THL mechanism followed by the phonon-kick could occur on semiconductor surfaces, in general. For surface bond breaking, the phonon-kick has to transfer enough energy to the top atom along the bond direction to break that bond. If that energy transfer is not sufficient for bond breaking, then, the atom will have a vibrational excitation that can lead to increased surface diffusion. The process of THL was found to preferentially occur on surface defect sites.[12] THL on the Ge surface can lead to selective energy transfer to the Ge atoms that landed on the surface from the ablation plume since these atoms constitute a surface defect site. The energy that is preferentially given to these adsorbed atoms can result in their hopping to settle epitaxially on the surface.

In summary, epitaxial Ge QDs was grown on Si(100)-(2×1) by fs pulsed laser excitation during growth. The growth was studied by *in situ* RHEED and *ex situ* AFM. The results show that excitation laser reduces the epitaxial growth temperature to ~70 °C. This result could lead to nonthermal method to achieve low temperature epitaxy which limits the redistribution of impurities, reduces intermixing in heteroepitaxy, and restricts the generation of defects by thermal stress.

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