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Comparative STM and RHEED studies of Ge/Si(001) and Si/Ge/Si(001) surfaces

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Abstract

Although epitaxial growth has been traditionally monitored by reciprocal space techniques, such as reflection high-energy electron diffraction (RHEED), the development of surface-sensitive imaging techniques, such as scanning tunneling microscopy (STM), allows for monitoring in the real-space. RHEED averages information over relatively large sample regions, and is thus more representative of the dominant surface morphology, which, however, makes it less sensitive than STM to the presence of small amounts of scatterers, or to local surface modifications. Compressively strained Ge/Si(001) surfaces exhibit a rich variety of two- and three-dimensional structures, which are interesting from a phenomenological standpoint, and may have far-reaching implications for the semiconductor industry. A comparison between these two techniques in the specific case of a Ge–Si heterosystem emphasizes how the complementary information provided by RHEED and STM is essential for a correct interpretation of the data.

Keywords: Epitaxy; Germanium; Reflection high-energy electron diffraction; Scanning tunneling microscopy; Silicon; Surface structure, morphology, roughness and topography

1. Introduction

Owing to its compatibility with the existing Si-based technology, the Si–Ge heterosystem is attractive for semiconductor devices. Although the strain in a heteroepitaxial layer, e.g. due to the 4.2% of lattice mismatch in Si–Ge case, can relax by misfit dislocations [1], in Stranski–Krastanow growth mode, it can also, at least initially, relax elastically via formation of three-dimensional (3D) pyramidal or dome-like islands [2,3], pits [4], or ripples [5]. Many of the structural details and growth characteristics of these features can only be revealed by atomic-resolution STM. As this roughness should be avoided, or at least minimized, for application in planar devices, or maximally controlled for applications in low-dimensional quantum confined devices [6,7], the ability to detect various stages of surface evolution, preferably as they occur during growth, is essential.

An important advantage of RHEED over low-energy electron diffraction (LEED), is that while the normal-incidence LEED picks out the well-ordered parts of the surface with orientations close to that of the average surface, the grazing-angle RHEED electrons will interact with asperities on the surface, producing a streaky or transmission diffraction pattern [8]. Static RHEED has been routinely used to estimate the quality of semiconductor surfaces [8–13], surface lattice constants [9], and, with sufficiently precise apparatus, even

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strains [11,14]. As the sample is not obstructed by the remote RHEED geometry, patterns can be acquired during growth (this so-called 'dynamic' RHEED is beyond the scope of this paper).

Since the interpretation of diffraction data without additional information from real-space can be ambiguous [12,15], a combination of a diffraction technique, e.g. RHEED, with a real-space technique, e.g. STM, is invaluable for reliable interpretation of the results. As RHEED is an integral part of any standard set-up for epitaxial growth in ultra-high vacuum (UHV), this paper is intended to serve as a kind of manual, by correlating between STM and RHEED patterns for the most common types of Ge/Si(001) and Si/Ge/Si(001) surface.

2. Experimental

Details of our sample preparation, growth, and STM acquisition are given in our previous work [16]. Briefly, chemically treated n-type Si(001) substrates were degassed in UHV, mounted in the STM stage (JEOL JSTM-4500XT), flashed at 1150°C, quenched to $T \leq 600$°C, slowly cooled to the desired growth temperature (RT–500°C), and exposed to germane (GeH_4) and/or disilane (Si_2H_6) in the 10^{-7}–10^{-5} Pa range. STM images were acquired in situ and in real time, i.e. during the exposure to germane and disilane fluxes, in constant-current mode, with currents around 0.1 nA and voltages in the ± 3 V range. [To avoid metal-induced $(2 \times N)$ reconstruction, the samples were handled with ceramic tweezers and clamped to the Ta support on the holder by Ta clamps. Sample cleanliness was routinely checked by Auger electron spectroscopy (AES), to yield a contamination-free surface. During the sample flashes and anneals, the pressure was kept below 10^{-7} Pa. Such treatment has generally proved effective in producing well-ordered Si-(2×1) surfaces.] RHEED patterns, obtained in situ with a 12-kV primary beam, were photographed from the screen (located 33 cm from the sample), scanned, and computer-analysed using conventional image-processing software. The accuracy of the interatomic-spacing measurements was ± 0.10 Å [corresponding to $\pm 2.60\%$ of the Si (001) surface lattice constant, $a = 3.84$ Å]. The sample mounting in the STM stage allowed patterns to be obtained only in a single [110] azimuth.

3. Results and discussion

A RHEED pattern from a Si(001) surface is shown in Fig. 1a, with the corresponding STM image in Fig. 2a. Although the double periodicity is only shown in a single crystallographic direction, LEED (not shown) exhibits a symmetrical appearance of the half-order reflections in perpendicular $\langle 110 \rangle$-directions, consistent with the equally populated (2×1) and (1×2) terraces, separated by monoatomic steps (Fig. 2a). The 3.83 ± 0.10 Å surface lattice constant calculated from the interstreak distance [9] in Fig. 1a is in excellent agreement with the value expected for Si(001), as well as with the value measured directly from high-resolution STM images. Two important observations follow from the comparison between Figs. 1a and 2a.

1. Although a certain degree of streaking is apparent in the diffraction pattern, this pattern is characteristic of a high-quality 2D surface (cf. patterns in Ref. [10] and Fig. 1e and f). This streaking results from the combination of instrumental and sample broadening [8]. As all the patterns discussed in this work were obtained within the same experimental apparatus under identical conditions, the differences between the patterns arise only from the latter. An imperfect, but still 2D, surface consisting of ordered domains of size L, e.g. terrace width, broadens the reciprocal lattice rods into cylinders of $a \sim 1/L$ thickness. The intersections of such cylinders with the Ewald sphere are ellipses, i.e. streaks (rather than spots in the idealized experiment). Thus, it can be concluded that the streaking in Fig. 1a is mainly due to the 30–40-nm terraces in Fig. 2a.
2. Fig. 1a also demonstrates that RHEED is insensitive to the short segments of dimer vacancy lines (DVLs) in Fig. 2a. A small amount of dimer vacancies is inherent to clean Si(001)-(2×1) surfaces, and coalesces into DVLs upon

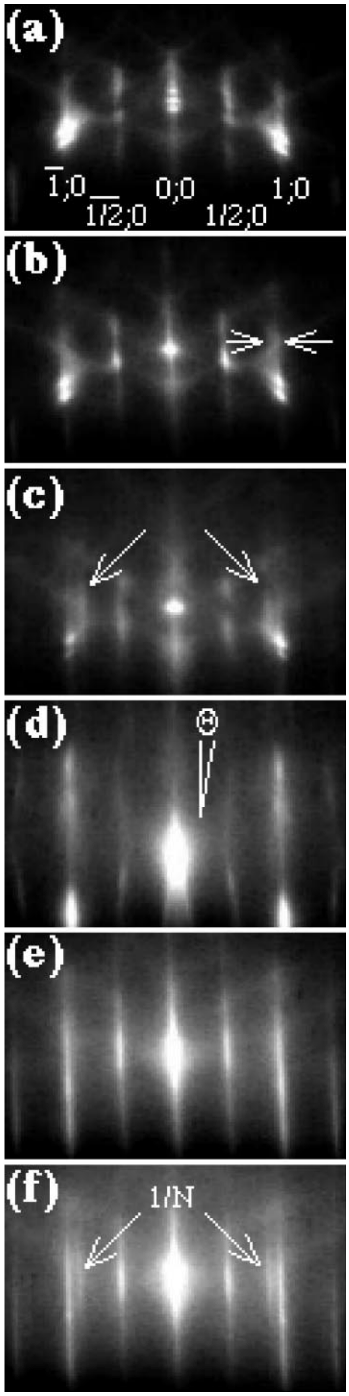

Fig. 1. Magnified RHEED patterns from Si(001), Ge/Si(001), and Si/Ge/Si(001) surfaces shown in Fig. 2 (⟨110⟩ azimuth, 12 kV). (a) pre-growth Si(001) surface at 500°C, (b) Si/Ge/Si(001) surface after anneal at 450°C, (c) 7.5 ML of Ge/Si(001) grown at 500°C, (d) 7 ML of Ge/Si(001) grown at 430°C, (e) Si/Ge-huts/Si(001) layer grown just to cap the apices of the Ge-huts at 430°C, after a short anneal at 430°C, and (f) the same Si-capping layer after a night anneal at 430°C. Arrows in (c) and (f) point to the $1/N$ satellites.

annealing [17], unless a buffer layer is grown. Such 2–3-dimer-wide DVLs can also form by sputtering, etching, or metal contamination [17], but, as we take special precautions to avoid metal contaminations [16], and, indeed, no metals were detected at the surface at any time, the DVLs shown in Fig. 2a are the intrinsic ones. Even when a higher density of DVLs, almost forming $(2 \times N)$ reconstruction, e.g. to accommodate segregated Ge [15], is present at the surface (arrowed in Fig. 2b), the only effect that can be seen in the corresponding RHEED pattern in Fig. 1b is a very slight broadening (arrowed as well) of the integer order reflections, which is inversely proportional to the interdefect spacing.

At the Ge/Si(001) surface, where DVLs are formed to relieve the homogeneous compressive stress of the Ge layer, the corresponding degree of order is sufficiently high, resulting in a well-resolved doublet of the integer-order RHEED reflections. Neither such Ge-$(2 \times N)$/Si(001) surface nor its corresponding RHEED pattern is shown here in isolation, as they also appear in the next figures (Figs. 1c and 2c). In accordance with the Ge/Si(001) growth diagram [16], further Ge deposition leads to a Ge-$(M \times N)$/Si(001) reconstruction, formed by an overlap of DVLs with dimer row vacancies (DRVs) [4,16,18]. The RHEED pattern in Fig. 1c exhibits well-resolved integer-reflection doublets resulting from the long periodicity of the vacancy trenches $N \approx 9a$, corresponding to the period between DVLs in Fig. 2c. Additional satellites reflecting the M-periodicity of DRVs are not resolved in this pattern, apparently due to a higher degree of meander and significantly larger period (see inset in Fig. 2c). However, the most striking observation is that the diffraction pattern in Fig. 1c is not indicative in any way of the ⟨100⟩-oriented, {501}-faceted hut clusters [2] seen in the STM image (Fig. 2c)! Clearly, the

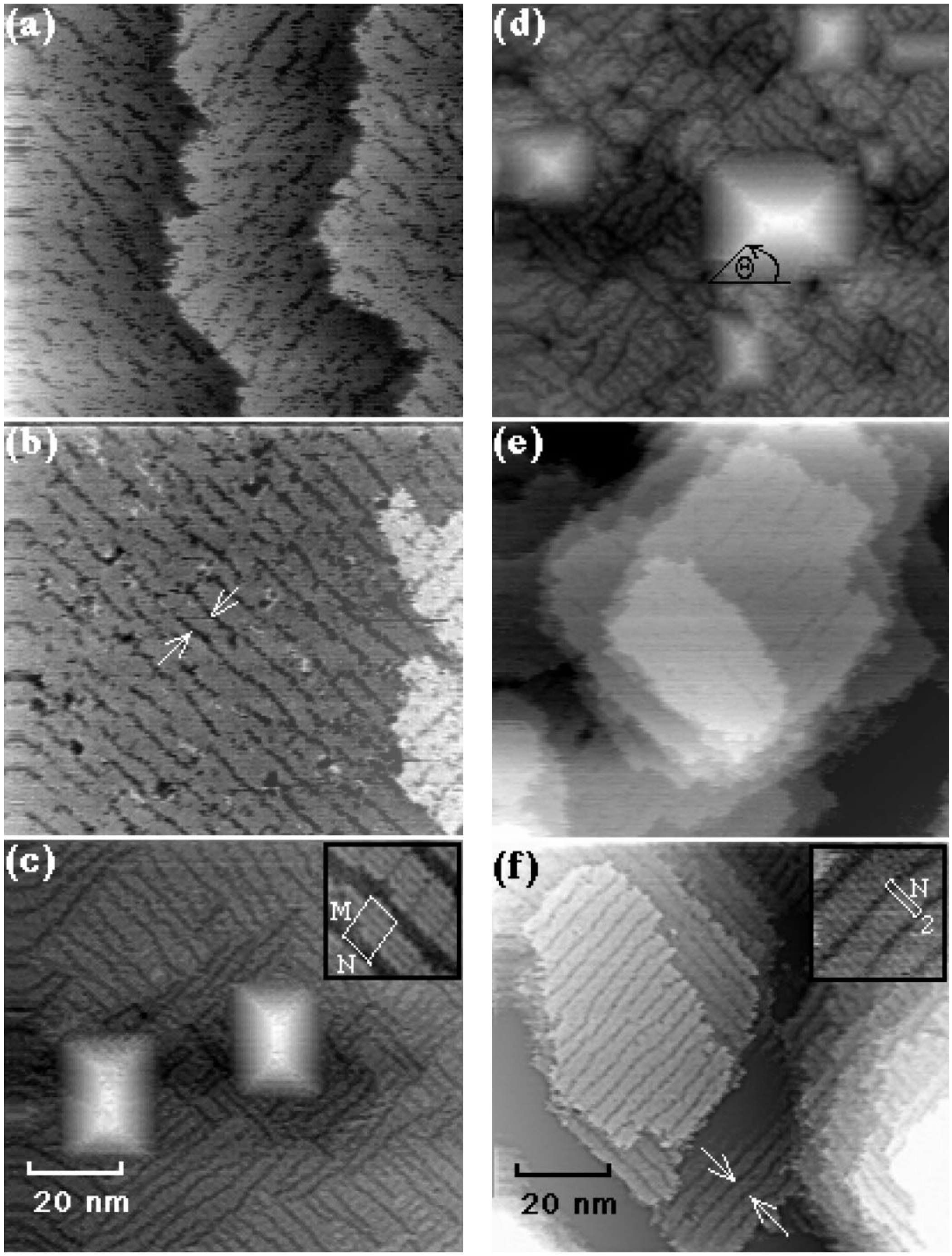

Fig. 2. (a)–(f) Constant-current filled-states (negative surface bias) STM image of the surfaces, from which Fig. 1a–f RHEED patterns were obtained. $(M \times N)$ and $(2 \times N)$ unit cells are outlined in (c) and (f), respectively.

initial low density of these small-sized 3D islands does not constitute sufficient active volume to be sensed by the electron beam, DRVs are 'unseen' as well, and the beam samples only the (2×1) and $(2\times N)$ superstructure in the $(M\times N)$-reconstructed intercluster wetting layer. It can be concluded, therefore, that not only the $(2\times N)$-to-$(M\times N)$ transition remains undetected by RHEED, but it is also impossible to determine the onset of the 2D-to-3D growth transition! Only when a sufficient number density of the huts is present at the surface (Fig. 2d) does the RHEED pattern change into a well-known chevron type [11–13] in Fig. 1d where the inclination Θ of the lines relative to the (0,0) line is caused by rotation of the reciprocal lattice of {501}-facets relative to the reciprocal lattice of (001) layer [8]. Thus, when diffracted near zone-axis, along the $\langle 100\rangle$-type azimuth $\Theta\approx11.2^\circ$, but along one of the $\langle 110\rangle$-directions, as in Fig. 1d, it is only $\sim8^\circ$ [12]. A certain degree of relaxation can be expected, mostly concentrated at the hut apices, but the measured 3.90 ± 0.10 Å, corresponding to about 1.6% relaxation, is still within the limits of measurement error (0.10 Å or 2.6%), and thus cannot be determined with certainty. Higher density and/or larger huts correspond to a higher volume fraction in the beam path and can result in a more precise measurement.

Capping the huts with Si is required to produce a quantum dot device. Figs. 1e and 2e show the corresponding RHEED and STM data, respectively, from such a Si/Ge-huts/Si(001) surface, after a short anneal. The source of the intense streaking in Fig. 1e is apparent in the STM image of Fig. 2e: the Si surface consists of several-monolayer height islands and holes. Such a discontinuous, so-called 'atomically rough' or 'multilevel' surface, combined with short terrace size, causes homogeneous intensity distribution along the streak length [8], as in Fig. 1e. The surface does not recover even after a very long, 12-h anneal at the growth temperature, as the unchanged amount of streaking in Fig. 1f reflects the same scale length of the ordered regions in Fig. 2f as in Fig. 2e. However the long annealing time resulted in Ge-segregation to the surface, followed by the formation of strain-relieving DVLs, the ordering of which is quite perfect, as can be judged from the STM image in Fig. 2f, as well as from the sharp and well-resolved $1/N$ satellites in Fig. 1f. It is well known [19] that, due to its lower surface energy, Ge tends to segregate to the Si layers grown on top of it, causing the formation of DVLs to relieve the resulting inhomogeneous stress at the Si(Ge)(001)-(2×1) surface [15]. Thus, the homogeneous surface strain yielding the $(2\times N)$ and/or $(M\times N)$ structures, following initial coverage of Ge on Si(001), gives DVLs that are one dimer wide, as in Fig. 2c, whereas in Fig. 2f, the inhomogeneous surface strain, resulting from segregation to the capping layer, gives DVLs that are two-three dimers wide [17]. This difference, however, is too subtle to be picked-up by RHEED.

4. Conclusions

The aim of this work was to estimate the capability of RHEED to detect the initial stages of various surface transitions (in the 2D state, as well as 2D-to-3D states) during strained-layer growth of alternating Si and Ge layers on (001) substrates. To achieve this, we have conducted a detailed correlation between RHEED patterns and corresponding STM images from these surfaces at various transitional stages.

It has been found that, relative to STM, RHEED is insensitive to the initial transition stages. For example, only at the final stages of the (2×1)-to-$(2\times N)$ transition are the DVL trenches sufficiently ordered and with a sufficiently short periodicity to produce well-resolved doublets of the integer-order reflections. The resolution is not sufficient to reflect the larger (and less ordered) M-periodicity, created by the DRV trenches, even at the final stages of $(2\times N)$-to-$(M\times N)$ transition. Furthermore, as RHEED beam bypasses even the 3D hut clusters, until sufficiently high cluster density develops at the surface, the initial, nucleation stage of the 2D-to-3D Stranski–Krastanow transition is completely missed-out by RHEED.

However, RHEED is very sensitive to the size of ordered domains on the surface, and discontinuous, atomically rough surfaces manifest themselves in RHEED as intense continuous streaking.

Segregation of Ge into the Si layer can be indirectly detected by RHEED, via $1/N$ satellites resulting from stress-relieving DVLs, provided that DVLs are sufficiently well ordered and their periodicity is sufficiently short to be resolved.

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