Adatom and Addimer-Mediated Exchange Diffusion in the Ge/Si(001)Surface Alloy

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Motivation—In Si-Ge thin films, the influence of the composition on the morphology, electronic, and optical properties is widely exploited for technological purposes. At the Si(100) surface, Ge alloying is used to tune the bandgap and charge mobility for micro- and opto-electronic device applications. The Ge-Si(100) surface is also a model system for studying alloy growth, the role of strain in epitaxy, and self-organization of nanostructures. In the initial stage of Ge-Si(100) epitaxy, Ge embeds into the surface at apparently random substitutional sites to form a surface alloy, driven by the resulting reduction in surface stress. Many studies have examined the early stages of the growth process, making inferences about the most likely mechanisms of surface alloy formation and structure, however, the atomistic processes involved in Ge embedding have not been observed directly by an atomic-resolution technique.

Accomplishment—Using variable-temperature STM, we investigated the initial formation and kinetics of the Ge/Si surface alloy. We observe the diffusion of the embedded Ge atoms at elevated temperature (>100°C). The diffusion events have the distinct character of defect-mediated diffusion. That is, rapid sequential diffusion of neighboring atoms separated by longer time intervals. We discovered that there are two defects responsible for embedded Ge atom diffusion: adsorbed dimers and adsorbed monomers.. Adsorbed dimers on this surface come in two flavors: pure Si-Si dimers and mixed Si-Ge dimers. In previous atom-tracking studies of pure and mixed dimer diffusion, we observed rare events in which mixed Ge-Si dimers exchange their Ge atom with a surface Si atom to become a pure Si-Si dimer. We also observed the reverse – a pure Si-Si dimer picking up an embedded Ge atom through an exchange event – leading to a mixed Ge-Si dimer. In the current work, we find that this process occurs much more frequently than previously thought leading to long-range diffusion and rearrangement of embedded Ge atoms in the surface alloy, as adsorbed dimers continually pick up and remotely deposit Ge atoms over the surface. Dimer-mediated Ge diffusion is shown in [Figure 1.]

Unlike dimers, adsorbed monomers on this surface are not imaged by STM at room temperature because they diffuse too fast. Nevertheless, there are configurations on the Ge/Si(001) surface in which a trapped monomer can be observed at the ends of adatom chains separated by a dimer row. Monomer diffusion is inferred by disappearance of the adatom at the end of one chain and the almost instantaneous appearance at the other. Exchange events between the diffusing monomer and alloyed Ge are detected as the monomer moves back and forth with an activation barrier of 0.8 eV.

Significance—Since the barrier for monomer diffusion (0.7 eV) is only slightly smaller than barrier to exchange into the surface, Ge-Si mixing will occur for any growth condition in which diffusion is operative. This is important because it makes atomically abrupt interfaces between Si and Ge, by Ge deposition alone, unlikely. Under growth conditions, in which there may be nonequilibrium concentrations of adsorbed monomers and dimers, the dominant process will depend on the populations of monomers and dimers, determined by kinetics, as well as thermodynamics.

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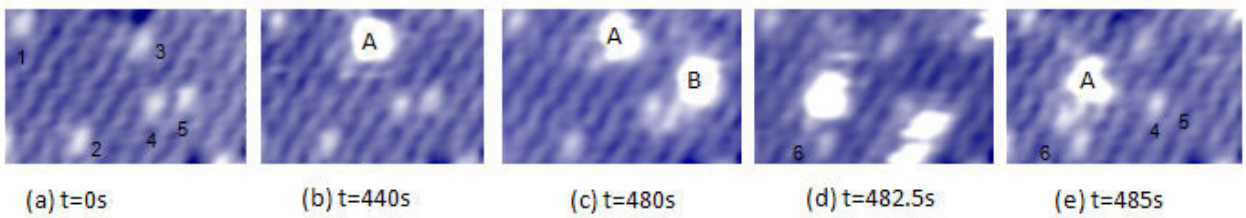

Figure 1. An image (4x8 nm^2) sequence showing the rearrangement of embedded Ge by mobile addimers on the dilute Ge-Si(100) surface at T=200°C. (a) The initial configuration of embedded Ge sites (numbered 1,2,3…). Ge does not move until addimers diffuse into the region. (b) After 440s (170 STM frames) an addimer labeled A is observed in the area. The image is deceptive: addimers are moving sufficiently fast that in most images, such as (d), the addimers appear as blurs because they hop several times during the STM image (c) At t=480s, a second addimer, B, arrives in the area. (d) A new Ge atom, 6, appears to have been exchanged into the surface, and dimer B passes over Ge site 5. A subsequent image (e) shows that the site 5 Ge atom is no longer present after the addimer B has passed, in addition, the new Ge atom at site 6 is now evident.