Mesoscopic Relaxation in Homoepitaxial Metal Growth

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We demonstrate that a size-dependent mesoscopic mismatch exists in homoepitaxy, which has a strong impact on the morphology of the islands and the substrate. Atomic scale calculations for double layer Cu islands on Cu(111) reveal that mesoscopic strain relaxations in both islands and the substrate strongly influence the shape of islands and can effect the details of atomic motion near the island.

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The microscopic understanding of film growth is of great importance for developing advanced microelectronic, optical, and magnetic devices. As device miniaturization reaches the nanometer-length regime, the control of the individual atomic events involved in the formation of stable nanostructures becomes crucial. The fast-growing area of nanotechnology requires experimental and theoretical studies of ways to manipulate the growth process on the atomic scale. Despite the considerable progress in this field [1–4], the knowledge of processes which control growth at a fundamental, atomic level is still limited. For example, one of the most intriguing phenomena, the fast island decay in homoepitaxial growth, discovered by Giesen *et al.* [5–8] is far from being understood [9]. The decay of islands and mounds is believed to be the effective channel for interlayer mass transport and can lead to smooth growth morphologies. In contrast to previous experimental and theoretical studies on the interlayer mass transport based on the concept of mass transport with individual adatoms, the decay of mounds is determined by a complex scenario of island and adatom motion, and possibly by local strain fields [8]. Usually strain relaxations are predicted on the basis of the macroscopic lattice mismatch between the two materials. In the case of homoepitaxy, according to the classical rule, no difference between deposited material and substrate has to be made and no effects arise due to a misfit between substrate and the film. The principal drawback of this approach is that mesoscopic and macroscopic islands should adopt their intrinsic bond lengths, which can be different from the bond lengths in bulk. It is well known, for example, that small metallic crystallites display substantially smaller lattice constants as compared to their macroscopic equivalents [10]. Recently we have demonstrated that the island growth in the initial stages of transition metal heteroepitaxy and atomic motion on islands are determined by the size-dependent mesoscopic mismatch [11,12]. Especially for small islands the mesoscopic mismatch may be significantly different from the macroscopic mismatch between bulk materials. Our experiments and atomic scale calculations have shown that the relaxation of the mesoscopic misfit strain in Co islands on Cu(001) leads to a stress oscillation with a period of one atomic layer [13]. The most recent STM experiments of Lin *et al.* [14] have found that mesoscopic strain induces magic Fe clusters on Cu(001). Relaxations of atoms in islands caused by the mesoscopic misfit can lead to in-plane lattice spacing variations [15]. Several experiments of Wang, Ehrlich, and co-workers on atom movement on and near islands in homoepitaxy have suggested that strain relaxations in surface clusters strongly influence dynamics of adatoms [2,16].

The goal of this Letter is to demonstrate that strain relaxations in homoepitaxy are determined by the size-dependent mesoscopic mismatch and can be crucial for understanding the growth process. Motivated by the experiments of Giesen *et al.* [5–8], we concentrate on double layer Cu islands on Cu(111). We reveal that islands and substrate atoms exhibit unexpected strong relaxations. An inhomogeneous distribution of adsorption sites near edges of islands predicted by our calculations may affect interlayer mass transport.

We perform atomic scale simulations by a molecular static method. The system is modeled using many-body potentials developed in the framework of the second-moment approximation to the tight-binding model [17]. The total energy of each atom is presented as the sum of a band contribution and a repulsive term. The form and the parameters of those potentials for copper are given in Ref. [17]. These potentials are known to correctly describe bulk and surface properties. Our system consists of a six layers thick (111) slab. Each layer contains 2250 atoms. Periodic boundary conditions are applied in the surface plane. On the topmost layer we put single or double layer Cu islands.

In mesoscopic islands the relaxation of edge atoms can be the dominating process. These atoms are relaxing in the direction of the center of the island and take other equilibrium positions with shorter bonds than in macroscopic systems. Therefore, the mesoscopic size-dependent mismatch between islands and the substrate in homoepitaxy exists and can locally affect the growth process. In the case of double layer islands or mounds, one can expect that mismatch between islands of different layers exists as well.

First, we present in Fig. 1(a) the results for the mesoscopic mismatch of 2D Cu islands on Cu(111). We determine the mesoscopic mismatch m by measuring atomic bond lengths in Cu islands on Cu(111) in a fully relaxed geometry and calculate it as $m = (r_0 - r_b)/r_0$, (r_b—average first bond length in Cu islands; r_0—first bond length for Cu bulk). One can see that for small Cu islands the mismatch varies strongly with the cluster size. Only for islands larger than 100 atoms does the mesoscopic mismatch approach the macroscopic one, which is zero for homoepitaxy. The above results indicate that strain relaxations in small Cu islands may lead to pronounced structural changes in the substrate and in the shape of clusters.

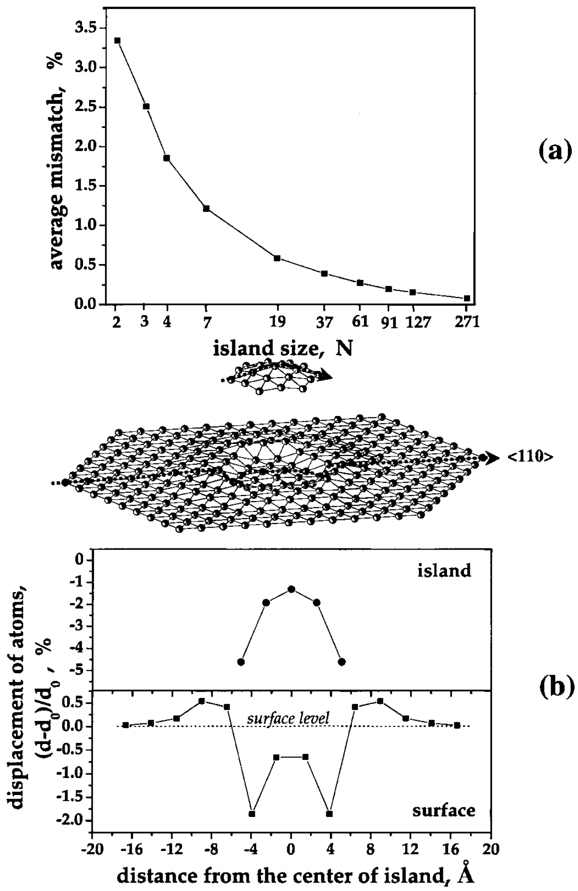

FIG. 1. (a) Evolution of the average mesoscopic mismatch $m = (r_0 - r_b)/r_0$ for Cu islands as a function of island size N (N—number of atoms per island; r_b—average first bond length in Cu islands; $r_0 = 2.556$ Å—first bond length for Cu bulk). (b) The vertical displacement of Cu atoms in the Cu_{19} island and the surface atoms in the topmost layer under the island in $\langle 110 \rangle$ direction; interlayer distance $d_0 = 1.8075$ Å and lattice constant $a_0 = 3.615$ Å.

One very fundamental issue predicted by these results is the possible strong impact of the size-dependent mismatch on the local strain field. In particular, one might expect significant adsorbate-induced structural modifications on the surface, which will be different for small and large islands. In other words, the substrate can dynamically respond to the growth of islands and can exhibit a strong inhomogeneous strain distribution during the growth process.

In Fig. 1(b) we present the atomic displacements in the Cu_{19} island and in the substrate along the $\langle 110 \rangle$ direction. The substrate atoms under the island are pushed down, and the island assumes a convex shape. The islands locally distort the surface and induce a strongly inhomogeneous displacement pattern in the substrate. Such atomic displacements lead to a strongly inhomogeneous stress distribution in the islands and the substrate. To demonstrate this, we perform calculations of the atomic level stress components [18]:

$$\sigma_{\alpha\beta}(i) = -\frac{1}{\Omega_0}\left[\frac{p_i^\alpha p_i^\beta}{m_i} + \frac{1}{4}\sum_j (r_{ij}^\beta f_{ij}^\alpha + r_{ij}^\alpha f_{ij}^\beta)\right],$$

where $(\alpha\beta) \equiv (x, y, z)$, m_i and p_i are the mass and momentum of atom i, r_{ij} means the distance between atoms i and j, $\vec{f}_{ij}$ is the force acting on atom i due to atom j, and Ω_0 defines the average atomic volume.

Figure 2 shows, as an example, the atomically resolved hydrostatic stress $P_\sigma = Tr(\sigma_{\alpha\beta})$ in the Cu_{19} island and in the surface layer. One can see that the mesoscopic mismatch leads to an inhomogeneous stress distribution in the islands and the substrate. At the island edge, the stress in the substrate is tensile (positive), while the substrate layer under the island is seen to exhibit compressive (negative) stress. The tensile hydrostatic stress at the cluster center is different from that at the edge. Thus, similar to heteroepitaxy [11,12] mesoscopic islands in

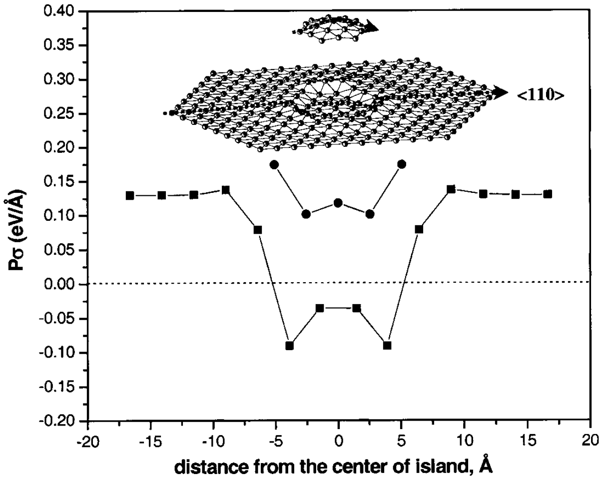

FIG. 2. The hydrostatic stress for the Cu_{19} island and the uppermost Cu atomic layer along the $\langle 110 \rangle$ direction.

homoepitaxial growth introduce stress in the surface region that can vary on an atomic scale. Thus, the principle of coherency between island and substrate is challenged by our calculations, even in homoepitaxy.

Now we turn to the discussion of the effect of mesoscopic mismatch in double layers of Cu islands on Cu(111). In this case, the scenario of mesoscopic relaxations is more complicated compared to the flat substrate. Both the upper and the lower islands exhibit strain relaxations. There are two kinds of step edges: (100) microfaceted step A and (111) microfaceted step B. Because of the relaxation of the edge atoms, the average bond lengths near the island edges at both A and B steps are reduced compared to the center. Therefore, we expect that the mesoscopic mismatch between the upper island and the lower island depends on the distance between the edges and may be different for step A and step B. For example, our calculations for a Cu dimer for different positions on the Cu_{271} island reveal that when the dimer approaches the edge of the island, mismatch between the dimer and the island changes abruptly and differently for A and B steps. These results suggest that the shape of double layer islands and atomic relaxations in islands and the substrate underneath may depend on the distance between the edges of islands. To prove this, we perform calculations for the double layer Cu island when a close contact between the edges occurs. Results shown in Fig. 3 reveal that the atoms at the edge of the lower island and the substrate underneath are pushed up, while atoms of the upper island and the substrate under the large island are pushed down. The strain relief at the edge of islands and in the substrate leads to the shape variation in islands as they approach the edge. We believe that a strongly inhomogeneous displacement pattern in the islands and in the substrate can affect the interlayer mass transport at the edge. Our calculations reveal, for example, that a close contact between island edges leads to local distortions of the adsorption potential around the cluster edge. The results for the adsorption energy for Cu adatoms near the edge of the double layer islands are shown in Fig. 4. One can see that for both steps A and B, variation of the adsorption energy is more than 0.3 eV, i.e., about 10%. An enhanced adsorption energy in some sites should lead to a preferential occupation of these sites by atoms. If we choose an infinite step as a reference [19], the adsorption energy near the island edge can be expressed as $E = E_0 + \Delta E(x, y)$, where E_0 is the adsorption energy near the infinite step; $\Delta E(x, y)$ is determined by the local coordination and by the local strain fields. The occupation probability P of the various sites at a temperature T near the edge of the lower island is given by $P = P_0 \exp(-\Delta E/kT)$, with P_0 denoting the respective value for the infinite step. We found, for example, that P at room temperature near step B where the upper island gets in close contact with the step edge is about 7–8 times larger than for step A [20]. A strong variation of the occupation probability of sites surrounding the island edge can guide Cu adatoms towards certain adsorption sites. Thus, the local coverage near the edge of the double layer island can depend on the position of the upper island.

In summary, while we have used a particular system, Cu islands on Cu(111), to illustrate several points, the points themselves are independent of the specific system. It is true that the size-dependent mesoscopic mismatch exists in homoepitaxy and can have a profound effect on growth modes. The mesoscopic mismatch leads to strongly inhomogeneous stress and strain distributions in islands and substrate. Our results challenge the

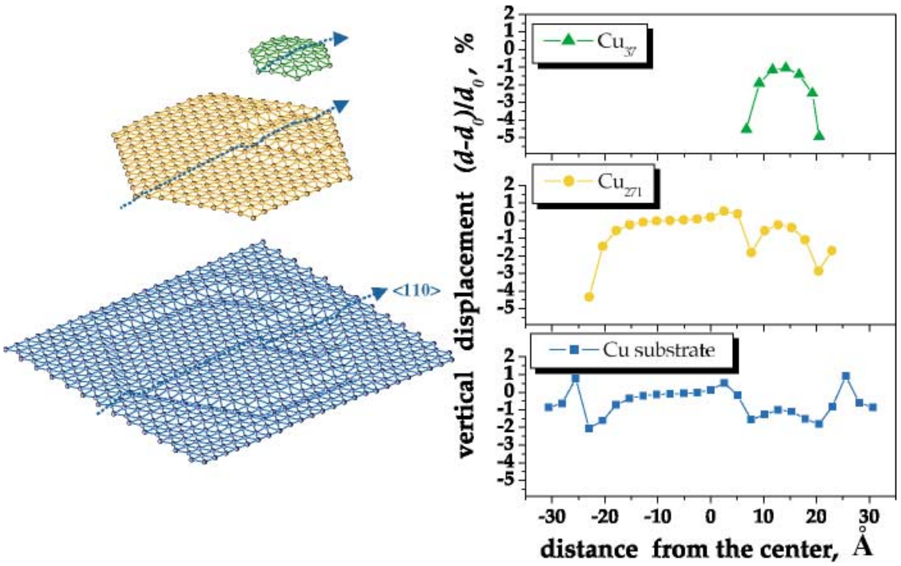

FIG. 3 (color). The shape of the double layer Cu island and the substrate for a close contact between the island edges. The vertical displacement of Cu atoms in the upper and the lower islands, and in the substrate are shown for the ⟨110⟩ direction.

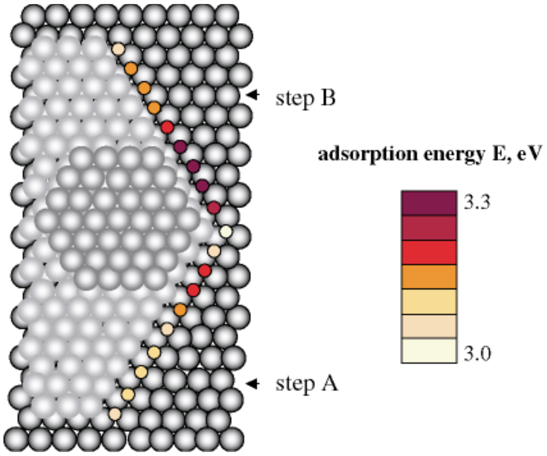

FIG. 4 (color). The adsorption energy for edge sites near the double layer Cu island.

conventional view of the lattice coherence between islands and the substrate in homoepitaxy. Similar to heteroepitaxy, mesoscopic strain makes the island shape size dependent. Our calculations suggest that the mesoscopic strain relaxations at the edges of double layer islands and mounds can be crucial for understanding the interlayer mass transport. The investigations of these effects are underway.

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