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Morphology changes due to AC induced electromigration in Gd islands on W(1 1 0)

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Abstract

Gd islands were grown on W(1 1 0) surface by evaporating Gd on the substrate at room temperature and subsequent annealing. STM images reveal in many cases islands which have a deep hole inside them. The appearance of the hole is associated with the application of an AC field. No such holes appear when the sample is heated by a DC current. We show that this can be explained by the combined affect of the AC field and the barrier to diffusion introduced by steps that can create a nucleus for further growth of an island which includes a hole in the middle. This may be generalized to a technique of tailoring the size, shape and distances of islands by, for example, two orthogonal AC fields with a phase delay of 90° between them.

Keywords: Gadolinium; Scanning tunneling microscopy; Surface thermodynamics; Surface morphology

The transport of matter in metals (mainly) and semiconductors under the influence of a DC current, known as electromigration has a significant (negative) technological importance due to its contribution to failure of electronic circuits. This is becoming even a more important problem due to device miniaturization and increasing current densities. This effect manifests itself as a bias of diffusion of atoms and defects induced by an external electric field [1]. This effect was detected in hydrogen in palladium [2] many years ago. It is understood now, that the electromigration occurs due to the momentum exchange between the conduction electrons and the atoms. This electromigration process, that can be quite significant, can occur both in the bulk (where most studies were performed) and the surface. This process was described as a result of drag forces that were applied by the moving medium of the conduction electrons on the static atoms. Therefore these forces are called wind forces. As one may expect, these forces have a significant influence on the mobility of the electrons too and consequently also on the resistivity of metallic surfaces [3]. Since these two phenomena originate from the momentum exchange between the drifting electrons and the adatoms, it is possible to calculate them simultaneously [13].

The exact estimation of the magnitude of these forces is a very complicated problem and it was discussed by many theoretical works [4–13]. The magnitude of the wind force was estimated by a broad variety of computational techniques. Electromigration is described, using the empirical law $F = eE(Z_d + Z_w)$: In this formula, the force (F) applied on the migrating ion is proportional to the external applied electric field (E). The first contribution to the driving force of the field is related to the valence of the ion (Z_d) and can be considered as the direct interaction of a charged ion with an external applied field. The second, more subtle, force is due to momentum exchange between the electrons and the ion, and it is expressed in terms of the wind valence Z_w.

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The wind force is usually much larger, and is dominant in most electromigration cases.

A straightforward solution to bulk electromigration in a free electron picture is given by the ballistic model [4]. The magnitude of the wind valence can be estimated as $Z_{\mathrm{w}} = -n_0 l \sigma_{\mathrm{tr}}$ where n_0 is the electron concentration, l is the electronic mean free path and σ_{tr} is the transport cross section of the adatom at the Fermi energy. Using typical values for light metals we get $Z_{\mathrm{w}} = -10$. These wind forces have many peculiar properties. As an example, when diffusion on Na(100) was considered, heavier metals such as W have a wind valence which is larger by more than one order of magnitude than light metals. A dependence of the wind valence on the diffusion path was found [10]. Another paper [11] predicts a twofold increase in the wind valence parameter when aluminum atoms are near a single step of aluminum (aluminum is commonly used as an electrical interconnect) – compared to an isolated atom on a terrace. This is a general phenomenon: The shadowing of atom, island, step etc, on a neighboring adatom is expected to modify significantly the wind force valence term Z_{w}, which can be written in these cases, as a tensor, namely drag forces in other directions than the direction of external electric field may appear [12]. In addition to inducing electromigration of adatoms, similar wind forces can induce motion of vacancies as well as other defects or impurities, both in the bulk and on the surface.

In all these cases, only DC voltage was discussed to create diffusion bias in the direction of the field. However, since wind forces can create a relatively significant biased diffusion, we discuss in this paper the induction of electromigration in quasi circular paths as a result of an AC current. This may lead to a possibility to shape the size, the shape and the inter-island spacing with AC current. It should be possible, for example, to connect a (rectangular) crystal with four orthogonal electric contacts and to apply time dependent voltages and currents in such a way that the diffusion is biased each time in a different direction. If we increase the frequency of the AC fields, for example smaller islands might be formed on the surface. In all these cases, the (closed-quasi circular) biased diffusion competes with the random component of the diffusing atom (or vacancy), however, since the applied fields can be large, and there are significant (for example elastic) interactions between the islands, this together with the bias diffusion can provide quite a strong tool for modifying the island's shape, size and inter-island distribution.

The affect of AC current on the morphology of islands is demonstrated in following experiment: We deposited several monolayers of a rare earth material (gadolinium) on top of a metallic crystal (W(110)). After deposition, we applied a strong AC field along the sample. In addition to this the crystal has steps that has a certain angle (namely they are not parallel, nor perpendicular) with the direction of the field. These steps are expected to create a significant barrier to diffusion also, in particular in the downhill directions (the Schwoeble barrier). Thus the combination of the biased diffusion of the field, together with the biased diffusion along the step, can drive the atoms into closed-quasi circular paths, which will alter its final shape. The evidence that this process did indeed occur, is a deep hole, created in the center of the island, which has a very peculiar shape, and appears only when the crystal is heated with a strong AC field. Some of these islands are shown in Fig. 1. We have also done DC heating and did not observe such shapes. A typical image with DC heating is shown in Fig. 2.

The experiment was performed in a UHV system (base pressure: 5×10^{-10} mbar). Gadolinium was epitaxialy grown on a clean W(110) substrate: 2–5 monolayers of Gd were deposited on the substrate at room temperature using an electron gun.

In order to clean the tungsten crystal and to anneal it after Gd was deposited a special heating system was constructed. The heating was made though resistive heating by passing currents close to 100 A through the sample supporting currents for annealing and flashing. The heating stage shown in Fig. 3, is specially designed in order to reach temperatures of several thousands degrees.

Afterwards, the sample was annealed by either an AC current (40 A and a bias voltage of 60 V – at 50 Hz) or similar DC currents to a temperature of 650 °C. In all the steps of this process the sample was characterized by AES to verify that the sample was clean before the deposition and to characterize quantitatively the deposition process, indicating that the islands grow as three-dimensional isolated Gd islands on the tungsten surface. Finally, using an STM [14], the surface of the sample was imaged in constant current mode. It was observed that after annealing by AC, Gd islands were formed with heights 2–14 ML. Fig. 1 shows several STM images in which a bi-modal size distribution of islands is observed (Fig. 1c–e) – small and thin islands and bigger and thicker ones, in which it is possible to identify a peculiar morphology: they appear to contain a relatively deep hole, and a kind of an adjacent balcony, lower than the main island.

The size of the hole is, on the average, 5 nm in diameter and several nm deep. Thus the hole cannot be explained in terms of a difference in the local density of states (as opposite to topography). If one compares the positions of the holes with the positions of the center of mass of the islands (Fig. 4), we see two asymmetric features in the position of the holes. The first is that the holes are distributed to a larger extent along the terraces of the W(110). This can be explained by the fact that the islands themselves are also asymmetric and are longer along the direction of the terraces. Additionally, there is an asymmetry in the positions of the holes in the lateral direction. We recall that in all of our images, the higher terraces are located on the right-hand side of the lower ones. Most of the holes are located closer to the right-hand edge of the island (compared to the center of mass). This asymmetry can be easily understood if one considers the asymmetry of the Schwoeble barrier, which forms a smaller barrier to diffusion in the uphill direction (as compared to downhill direction). This means

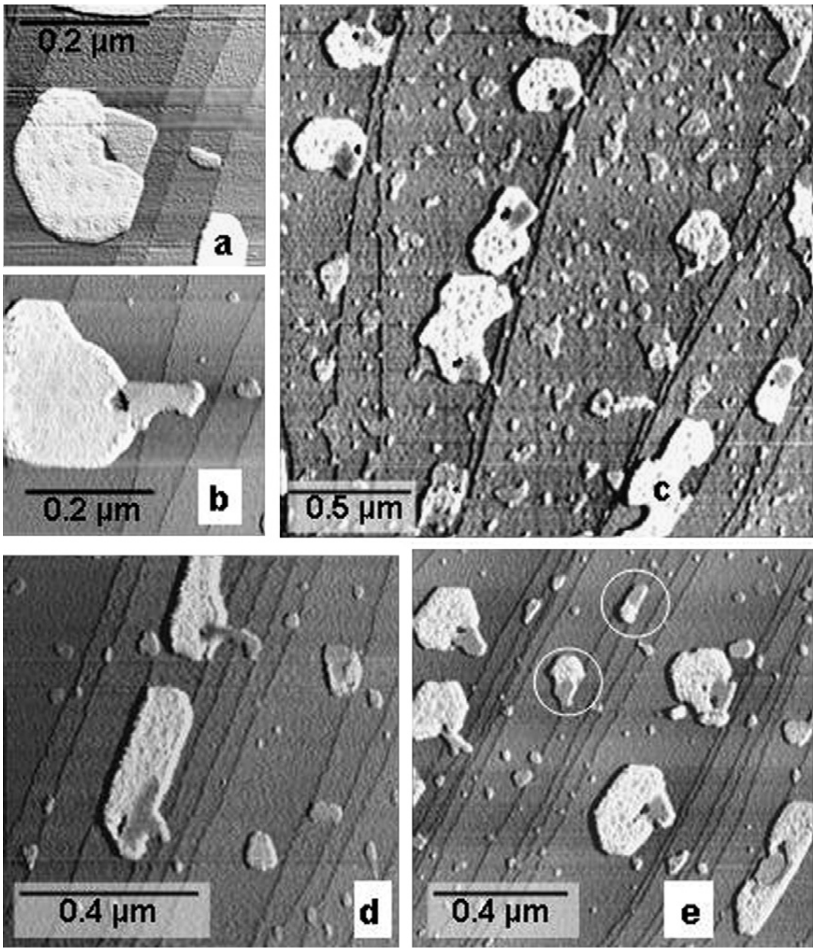

Fig. 1. STM images of islands with the "hole" morphology: The images were taken at a sample bias of 0.8 V and tunneling current of 1 nA. The circles in (e) are around a coalescence of a thick and a thin island without a hole.

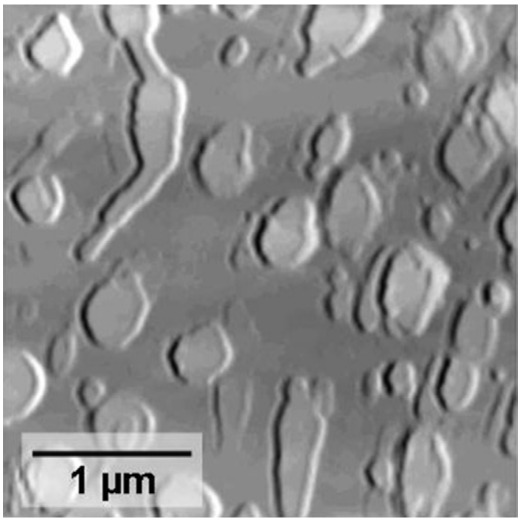

Fig. 2. STM images of 14 monolayers of Gd after annealing at 650 °C with 31 A, 1.8 DC-Volts. The images were taken with bias 0.8 V and 1 nA tunneling current.

that the restriction for diffusion is larger at the downhill step.

These deep holes can be easily explained in terms of circular diffusion. As usual in such growth processes the first step of a nucleation of an island which has a circular shape with a hole, is the most important step. Once a kind of a seed of the island with the hole is formed the diffusion of the atoms into the hole is strongly hindered by the step which surrounds it. In order to show that indeed the path of the Gd atoms under the affect of the electric field (of the same magnitude that was used in the experiment) is similar to the final island morphology, we performed a simple simulation of diffusion based on the Metropolis algorithm [16]. Our first step is to estimate the magnitude of the wind force which is responsible for the biased diffusion. We have no precise values for the parameters that are relevant for this simulation. Namely we do not know the exact value for the wind force valence parameter of gadolinium on W(110). Nevertheless based on the calculation made in [10] we estimate Z_w to be larger than 100 (we also take into account the increase of the wind force parameter in the neighborhood of steps [11]). Thus the wind force is estimated to be eZ_wE. The electric field (E) on the sample is in the order of magnitude of 5×10^3 V/m. and the force is in the order of 10^{-13} N. Since the force is the gradient

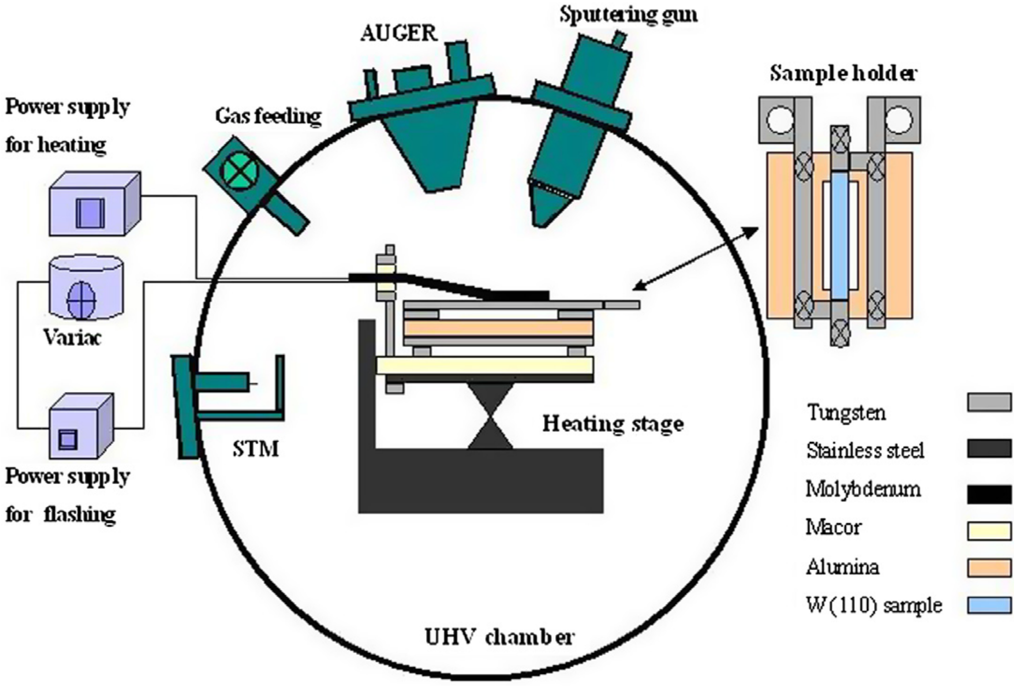

Fig. 3. A schematic presentation of the heating stage of the sample.

of the potential energy, Considering the fact that the periodicity of the diffusion barrier in a W(1 1 0) is in the order of 1 nm we get a difference in barrier to diffusion in the direction of the field in the order of 10^{-22} J. We estimate this to be about 1% of the height of the diffusion barrier of Gd on W(1 1 0).

We have simulated a random walk over a square lattice with a random number generator which gives equal probability (0.25) to move in four directions (Fig. 5 – left). Afterwards, we added the affect of the sinusoidal AC field to induce bias diffusion in the direction of the electric field. We assumed that the diffusion bias induced by the field was $\Delta E/kT = 0.01$. (The relative change in the barrier to diffusion as a result of the field.) The affect of the field was included according to the Metropolis algorithm in the diffusive process by calculating a time dependent factor $F_{\mathrm{E}}(t) = \mathrm{e}^{-0.01\cos(\omega t)}$, when ω is the frequency of the AC field. Thus, a diffusive step in the direction opposite to the field – namely when $F_{\mathrm{E}}(t)$ was smaller than 1 – was performed only when a random number generator has given a value smaller than $F_{\mathrm{E}}(t)$. The probability in all other directions remains the same (Fig. 5 – middle). In this way, a biased diffusion was induced in the direction of the field. In addition, a similar biased diffusion was introduced as a result of the barrier to diffusion at the step. The bias factor at the step was assumed to be $F_{\mathrm{s}} = \mathrm{e}^{-5}$ (the barrier to diffusion in the step was assumed to be 5×10^{-20} J). Again, the diffusion in the direction of the step is performed only when a random number generator gives a number smaller than F_{s}. The probability in all other directions remains the same (Fig. 5 – right). Unlike the diffusion bias of the electric field this bias is applied only at the location of

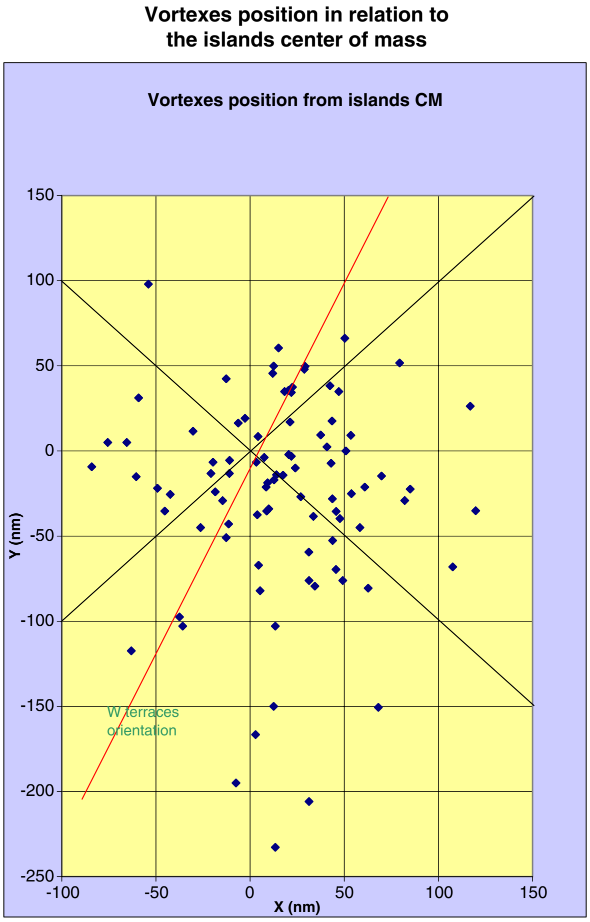

Fig. 4. The position of the holes in the island as compared to the center of mass of the islands. The arrow indicates the average terrace direction.

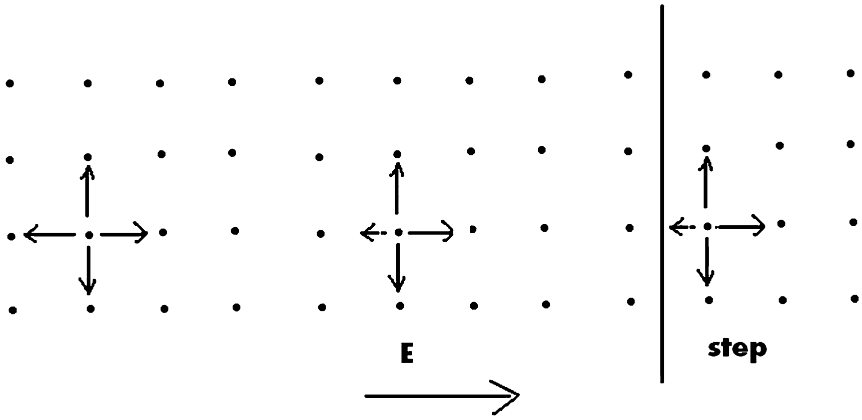

Fig. 5. Calculating probabilities in the Metropolis algorithm: Left – unbiased diffusion: equal probabilities to move in four directions on the square lattice. Middle – biased diffusion as a result of an electric field: the probability to move in the direction *opposite* to the field is reduced. Right – biased diffusion as a result of the presence of a step: the probability to move *in the direction* of the step is reduced.

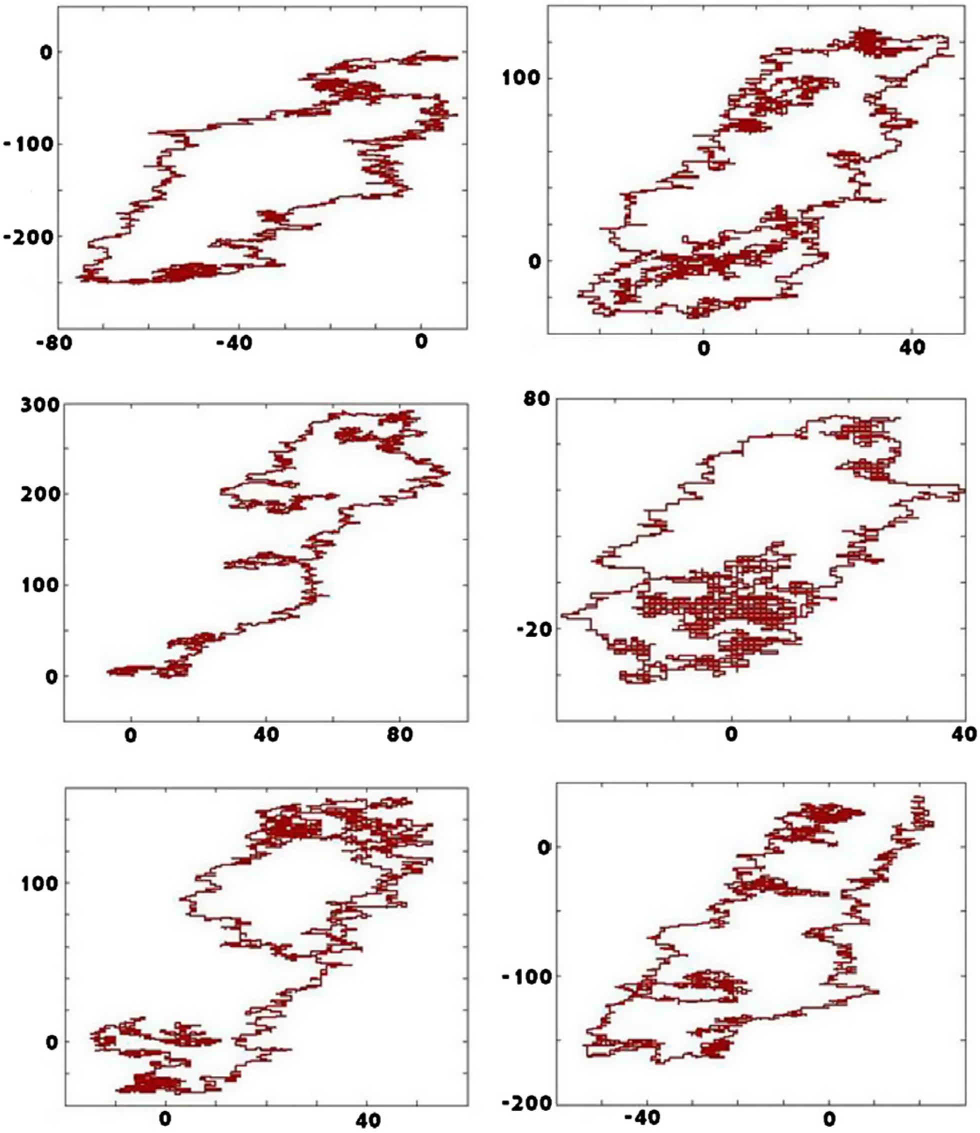

Fig. 6. Six examples for a closed-quasi circular path observed when an atom is under the influence of an AC field and steps. The steps are located at $4X + 50$, and $4X - 50$. The frequency of the AC field was 0.02 Hz.

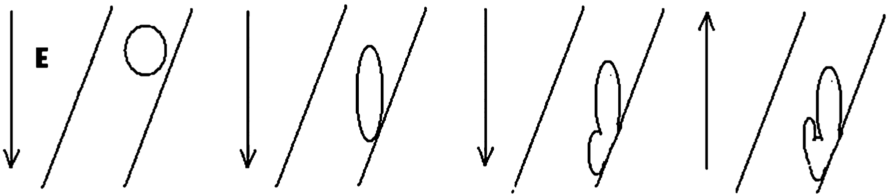

Fig. 7. The evolution of the island morphology to develop a nucleus with a hole from left to right – a circular island is elongated in the direction of the electric field, the island is bent after reaching the step, and elongation of the island in the opposite direction when the field is inverted – can give an empty space in the middle, that can develop into a deep hole when the field is further reversed.

the step. We ignore in the simulation the asymmetry between downhill and uphill diffusion. Anyway, the similar results of the simulation are observed also when the size of the barrier of the steps is reduced by one order of magnitude.

Under the combined influence of the sinusoidal electric field and the barrier of the step, the atom performs a combined random and biased motion. At moments when the electric field is large, the biased diffusion is strong, while at moments when it is small the atom performs a random motion. When the steps are not fully perpendicular to the electric field, If the bias motion is such that the atoms are transferred from one step to the other, diffusive motion may occur along the step before the electric field is reversed to transfer the atoms to the other step. Since the field has also a component parallel to the steps, it will introduce also biased diffusion along the steps in opposite directions. The final result of this scenario is a closed-quasi circular path of the atoms in large number of cases (in the simulations shown in Fig. 6 one of three simulations resulted in a biased diffusion orbit which can be considered closed-quasi circular – six of them are shown in the figure). Of course, when the fields are smaller, the biased diffusion is slower, and it will be necessary to reduce the frequency of the field in order to get such a closed-quasi circular path. As was explained earlier, in such cases once an island with a hole is formed, this will become a very efficient nucleus, which will induce much large islands with a deep hole in the middle, as observed in the experiment. The fact that not even a single island with such a shape was formed when the sample was annealed with a DC current strongly indicates that this scenario is correct (Fig. 2).

This explanation can be correct only if the path made by the diffusing atom in the time scale of one cycle of the electric field is larger (but not much larger) than the distance between the steps. If the path is smaller, the atom will diffuse back and forth without performing a closed-quasi circular path. If the path is too large, the atom will diffuse a large distance along the step. Although no measurements were done on the Gd on W(1 1 0) system, we can take the barrier to diffusion taken from the Pd on W(1 1 0) system – 0.5 eV [15] to perform an order of magnitude estimation. Since the diffusion was done at elevated temperatures, we estimate the diffusion constant in this temperature as $5 \times 10^{-8}\ \mathrm{cm}^2/\mathrm{s}$. This means that the diffusion path for 2×10^{-2} s is of the order of 0.4 μm, which is indeed slightly larger (but not much larger) that the distance between the steps.

There is a complementary plausible way to explain the process in which the islands are nucleated, we recall that it is well known that islands (beyond a certain critical size) that are electromigrating under the influence of a strong electric field and that are, in parallel, change their morphology in such a way that they elongate in the direction of the electric field [17]. Thus a scenario similar to the one of the single atom is possible also in an island as shown in Fig. 7. The island drifts and elongated in the direction of the field. In front of the step the island is bent such that its lower part is parallel to the step. Afterwards, the field direction is inverted and the "broken" island is then elongated in the opposite direction – as a result of the significant "shadowing" due to the ballistic nature of this force [10], this elongation will only occur in the external side of the island, leaving behind a kind of a "hole". This explanation is plausible if the island migrates on a time scale of 0.02 s [18].

This method might be extended, as was explained earlier, to grow islands, with a narrow size, shape and distance distributions, even in the absence of steps, when two orthogonal sinusoidal electric fields 90° out of phase will be applied through four contacts around the sample. By modifying the amplitude and the frequency of these fields, it should be possible to modify the island shape, size and distances and to create, in particular if the islands are interacting with each other, an ordered array of islands.

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