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Aubry transition in a real material: prediction for its existence in an incommensurate gold/gold interface

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Abstract. – Recent studies have shown that a special grain boundary in gold is stable and is an incommensurate interface related to the irrational number $\sqrt{2}$. Using the theoretical framework of quasicrystals, we show that this system can undergo an Aubry transition: the two grains can glide freely along the ideal interface; if we increase the interactions between the grains by applying a compression perpendicular to the interface, there is a threshold after which the grains are pinned to each other. With a numerical analysis of the energy landscape corresponding to the shift of the grains we determine the atomistic mechanism of the grain sliding.

Introduction. – Most of the grain boundaries studied in the literature are periodic structures, *i.e.* the interface structure can be described with a 2D periodicity. However if we imagine, for example, that two grains corresponding to the same crystalline materials are related by a tilt or a twist of a general angle, then the interface can have an incommensurate structure [1–4] and thus it can be non periodic.

Recently, an incommensurate gold grain boundary has been found stable and has been studied by high resolution electron microscopy [5]. An atomistic modeling has been performed and a somehow surprising property has been found and checked numerically [6]: a zero static friction during the relative displacement of the two grains along the ideal interface.

This hypo-friction property has also been discovered for other systems composed of subsystems having incommensurate relationships between them such as a tungsten tip on a silicon surface [7] or multiwalled carbon nanotubes [8].

This property also exists in simple theoretical models that are prototypes to study sliding dynamics: the Frenkel-Kontorova (FK) model [9, 10], the two chain model [11–14], and the Frenkel-Kontorova-Tomlinson model [15]. The Frenkel-Kontorova model consists of a chain of particles with harmonic interactions, embedded in a periodic potential field. The structure is incommensurate when the ratio of the length period of the field to the equilibrium length of the particle interaction is an irrational number.

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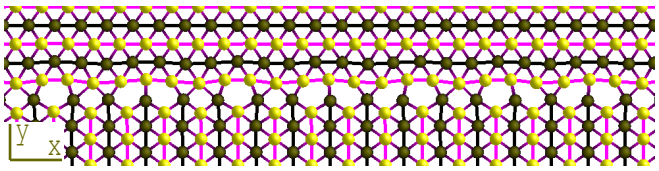

Fig. 1 – Model of the $\sqrt{2}$ gold interface. Inter-atomic distances shorter than 0.32 nm are drawn. Atoms are in two layers at the altitudes 0 (*dark*) and $r_0/2$ (*light*) along the z-direction.

Aubry has shown [9] that, depending on the strength of the interactions, there is a transition between a state in which the incommensurate chain is pinned by the potential field and a state with a Goldstone mode, *i.e.* the potential energy is invariant with respect to a continuous displacement of the chain. In the "$\sqrt{2}$" gold interface, one grain can be seen as the source of a periodic field acting on the other grain. Besides the fact that the grains are 3D bodies, a difference with the FK model is that each grain is distorted by the other. We address here the question of the existence of an Aubry transition in a real material. As we are considering gold, we cannot tune the interactions as in the FK model to get a transition. Instead, we have used the following idea : if we apply an uniaxial pressure perpendicular to the interface, the interatomic distances are shorter along this direction and are in a domain of stiff repulsive interactions. In particular, this is the case between atoms on each side of the interface, resulting in an increase of the inter-grain interactions with respect to the interactions inside of the grains.

Atomic structure. – Gold is a face-centered cubic crystal (fcc) of lattice parameter $\sqrt{2}r_0$, where r_0 is the smallest interatomic distance. Let x be the incommensurate direction parallel to the [100] cubic direction of one crystal (lower grain in figure 1) and parallel to the $[01\bar{1}]$ direction of the other crystal (upper grain). We denote the interface normal as the y direction. The z axis corresponds to a commensurate direction of the interface and is parallel to the [011] direction common to both crystals.

Although the incommensurate interface is not periodic along x, it can be considered as the limit of commensurate interfaces with increasing periods. These structures are called approximants and are useful since they can be studied with periodic boundary conditions. The continuous fraction representing the number $\sqrt{2}$ can be truncated at a given rank, leading to a rational approximation n_s/n_ℓ of this irrational number. This gives us the numbers n_ℓ and n_s of, respectively, [100] and $[01\bar{1}]$ crystal periods ℓ and s ($\ell = \sqrt{2}r_0$ and $s = r_0$ in the ideal incommensurate case), that we have to set along the x axis to get an approximant of the $\sqrt{2}$ interface. In this study, the approximant 17 : 12 has been used for energy barrier calculations while the approximant 239 : 169 has allowed precise atomic structure determinations. In the latter case, the relative error to $\sqrt{2}$ is 10^{-5} and the period of the approximant is 69 nm.

As described with more details in our previous work [6], the construction of the atomic models starts with the crystalline atomic positions of two grains of gold. The positions of all the N atoms are relaxed by minimizing the total potential energy :

$E_{pot} = \sum_{i,j\neq i}^{N,N} a\, e^{-p\, r_{ij}} - \sum_i^N \left(\sum_{j\neq i}^N b\, e^{-2\, q\, r_{ij}} \right)^{1/2}$

where r_{ij} is the distance between atoms i and j. The atomic interactions are based on the tight-binding second moment approximation [16]. This form of N-body potential has been used to analyze the surface stability in noble metals [17]. The parameters $\{a, b, p, q\}$ have been fitted [18] to the lattice parameter, the cohesive energy, and the elastic constants of gold. At the radius 1.2 r_0, the exponential functions are replaced by fifth-degree polynomials that

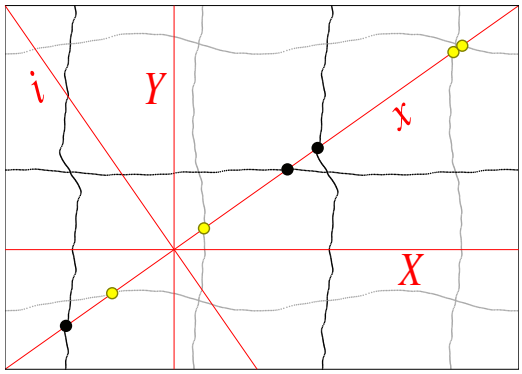

Fig. 2 – Hyper-space view of the $\sqrt{2}$ gold interface. Only lines corresponding to 2 atomic rows on each side of the interface have been drawn. The disks indicate the x coordinates of atoms, which are in two layers at the altitudes $z = 0$ (*dark*) and $z = r_0/2$ (*light*) and which belong to the lower (horizontal lines) or the upper (vertical lines) grains.

vanish at $r_c = 1.5283\ r_0$. Continuity up to the second derivative of the potential is guaranteed everywhere. Several properties of gold such as the vacancy-formation energy and the Rose curve have been successfully reproduced [18].

A zoom on the interface is shown in fig. 1 (239 : 169 approximant). However, with the tools provided by the theory of the quasicrystalline and incommensurate structures [19–21], a better structural description can be given by adding one virtual space dimension i, *i.e.* by embedding the structure in a 4D hyperspace.

Each atom M with coordinates (x_M, y_M, z_M) has simply the 4D coordinates (x_M, y_M, z_M, 0), where the last coordinate is along the new direction i, orthogonal to the physical space. Now, to get a periodic density in the hyper-space, we extend the structure by setting periodic images of M in the following way. The x-axis being the only incommensurate direction, we can restrict our discussion to the (x, i) space. In a basis rotated by $\theta = \arctan(1/\sqrt{2})$ (see fig. 2), we consider a square lattice with the parameter $a = \sqrt{2}r_0/\sqrt{3}$. Atom M and its new periodic images $M_{I,J}$ have the coordinates $X = (x_M\sqrt{2}/\sqrt{3}) + I\,a$ and $Y = (x_M/\sqrt{3}) + J\,a$ where I and J belong to the integer ensemble. These relations come from the fact that the atomic coordinates x in each grain, far from the interface, are periodic. They should be given by the intersection of the x-axis with equidistant straight lines parallel to $Y = 0$ for the lower grain and to $X = 0$ for the upper grain. Figure 2 shows that the "$\sqrt{2}$" interface can be described by a set of continuous "atomic lines", periodically set in the (x, i) space and whose intersections with the x axis give the atomic x coordinates. This demonstrates that the interface structure is indeed not disordered. The curved shapes of the lines correspond to the atomic relaxations that we have calculated and are the results of the subtle distortions of each grain by the other.

If the physical space is translated along the i axis in the hyper-space, the x atomic coordinates goes, respectively, to the negative and positive directions for the lower and upper grain (see fig. 2). This corresponds to the sliding of the grains one against the other. Because this transformation corresponds to a phason displacement, the global structure of the interface is unchanged and in particular the energy is constant. Since the "atomic lines" are continuous, all the atomic moves are continuous and the sliding corresponds to a Goldstone mode. This implies that there is no static friction forces, hence the hypofriction property for an ideal interface of this type.

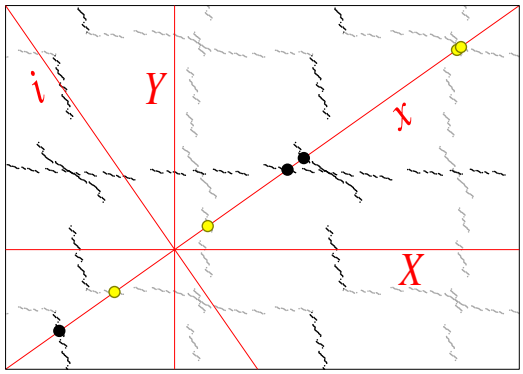

Fig. 3 – Hyper-space view of the strained $\sqrt{2}$ gold interface ($\varepsilon = 3.5$ %). Only lines corresponding to 2 atomic rows on each side of the interface have been drawn as in fig. 2.

Strained structure. – To increase the interaction between the two gold grains, a compression is applied perpendicular to the interface. Thus we shrink the y size of the computational periodic box and we relax again the atomic positions. As we want to strictly keep the $\sqrt{2}$ incommensurability, we have actually applied a strained ε along y with fixed x and z boundary conditions.

The new interface structure, while embedded in hyper-space, is shown in figure 3 for a compression ε equal to 3.5 %. The "atomic lines" undergo a qualitative change: they are now discontinuous. This drastic change corresponds to the "breaking of analyticity" of an Aubry transition [9] and is similar to the transition found in a two-chain model [14] (note that the atomic lines and the Hull functions are slightly different presentations of the same quantities).

The largest gap Δ (parallel to the physical space) of the "atomic lines" can be taken as an order parameter to quantitatively characterize the Aubry transition [22]. In a Frenkel-Kontorova model, it corresponds to the vanishing of the atomic distribution in the vicinity of

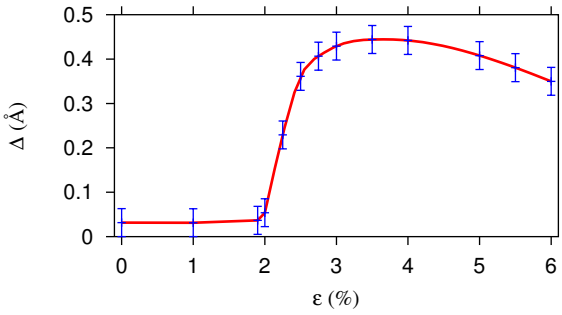

Fig. 4 – Maximum discontinuity Δ (order parameter) of the "atomic lines" , versus the compression strain ε applied perpendicular to the interface. The "atomic lines" are calculated only at discrete points corresponding to atomic positions and the error bars correspond to the maximum separation found in the unstrained structure (approximant 239 : 169).

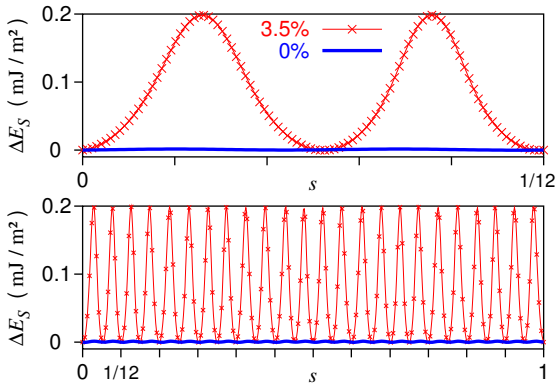

Fig. 5 – Interface energy ΔE_S versus path coordinate s when translating one grain along the other without strain (*thick lines*) and with a 3.5 % compression strain ε applied perpendicular to the interface (*crosses*). *Bottom*: translation by one interatomic distance $d_0 = 0.287$ nm. *Top*: translation by $d_0/12$. Calculation done with a "17÷12" approximant of the "$\sqrt{2}$" interface.

the potential field maxima. Figure 4 shows the gap Δ against the compression strain. This demonstrates that the gold interface undergoes an Aubry transition at a compression equal to 2 % ($\simeq$4 GPa). The curve $\Delta(\varepsilon)$ is continuous and thus the transition is of a second order type as was found in the Frenkel-Kontorova model [9, 22] and related models [14, 15].

If the physical space is now translated along the i axis in the hyper-space, the x atomic coordinates (see fig. 3) go through discontinuous values that correspond to atomic jumps occurring during the grain gliding. Thus the order parameter Δ plotted in fig. 4 is also the length of the longest atomic jump. However, the evolution of the interface structure during the jumps is not described by the hyper-space lines. Only the atomic positions of the interface ground-states are given by the intersection of these lines with the physical space. Thus, to get the complete atomic trajectories corresponding to the gliding, as well as the energy of the transition states, we have performed minimum energy path calculations.

Minimum energy paths. – We have used an algorithm [23] that has been designed to compute the saddle point between two minima. Given two stable configurations of N particles, we compute a path in the configuration space ($3 \times N$ dimensions) that connects these two states by a gradient line that goes through a saddle point, *i.e.* a transition state. This path, also called reaction path, can be viewed as the atomic trajectories at 0 K and the saddle point corresponds to the energetic cost of the transition. On this path, the atomic forces are tangent to the trajectories and this gives access to the force that has to be applied on the atoms to make the transition. In our case, it is related to the static friction force defined [15] as the smallest driving force that could initiate sliding. During the path calculation, the atomic positions at the initial and final states are fixed while the coordinates x, y, and z of each atom along the trajectories are allowed to relax. No atomic displacements have been found in the commensurate z-direction and the interface remains flat without a global y displacement.

These path calculations have been done with the $17 : 12$ approximant of the interface, which means that we have $n_\ell = 12$ segments ℓ facing $n_s = 17$ segments s per period. Let us consider a particle at the interface belonging to the grain with periodicity s. The system may have a transition state each time this particle goes from one side to the other of a particle of the other grain, *i.e.* for every shift of length ℓ. Since there are n_s such particles in the first grain, there can be a transition for every relative shift δ of the grains equal to $\ell/n_s = s/n_\ell$.

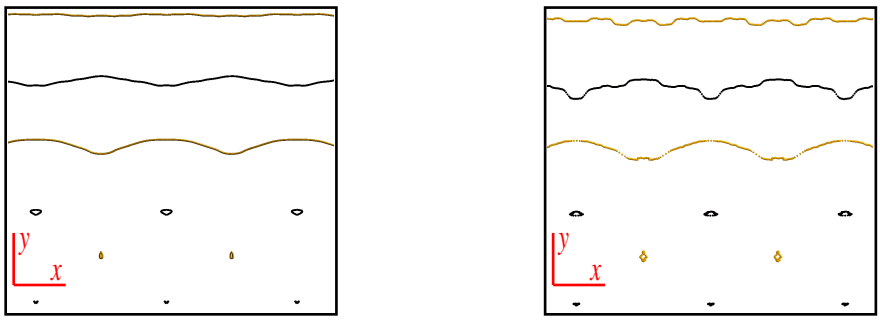

Fig. 6 – Atomic trajectories corresponding to the translation of the upper grain along x with the minimal potential energy (shown on fig. 5): $\sqrt{2}$ gold interface without (*left*) and with $\varepsilon = 3.5$ % strain (*right*).

Because we have also used periodic boundary conditions in the y direction, there are actually two interfaces separating the two gold grains and thus two transitions per shift δ as shown in figure 5.

Fig. 5 demonstrates that on each side of the Aubry transition the $\sqrt{2}$ interface belongs to two different types [22] of incommensurate structures: the unstrained system has continuous phasons (fig. 2) and presents no friction, while the constrained system has no continuous phason (fig. 3) and the grains are pinned to each other. The atomic trajectories corresponding to the minimum energy paths are shown on fig. 6. Soft trajectories characterize the sliding of the grains without friction, whereas stick-flip motions of the gold atoms occur in the pinned interface. In the first case, we obtain the atomic motions corresponding to the Goldstone mode, while in the latter case we get a description of the transition between energy minima.

Conclusions. – We have shown in this work that, if enough atomic positions are known, the analysis in a (x, i)-space of an interface structure can allow us to determine if the interface has an hypo-friction property (no static friction at 0 K) or if the grains are pinned to each other: in the first case the "atomic lines" are continuous while in the second they are discontinuous. The periodicity in the (x, i)-space, which allows this analysis, is fixed by the crystalline structures far from the incommensurate interface. This article shows that an Aubry transition can occur in a real system, namely an Au/Au interface. The atomic trajectories at 0 K have been determined by computing the minimum energy path associated with the grains sliding. The trajectories are continuous and the static friction force, which corresponds to the slope of the energy along the path, is zero for the unstrained $\sqrt{2}$ interface, whereas it has a non vanishing value above the transition.

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