Chemical Physics Letters 664 (2016) 226–232

Contents lists available at ScienceDirect

Chemical Physics Letters

journal homepage: www.elsevier.com/locate/cplett

Research paper

Analytic method for sputtering yield calculation in nanoislands

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ARTICLE INFO

Article history:
Received 15 July 2016
In final form 6 October 2016
Available online 13 October 2016

Keywords:
Sputtering
Nanoislands
Ion bombardment

ABSTRACT

A theoretical approach based on the well-known work of Sigmund to explain the sputtering of surfaces covered with nanoclusters is developed. Predictions of this approach are compared with molecular dynamics results of bombardment of a periodical array of Co nanoislands deposited on a Cu(001) substrate with 1-keV argon ions. Sputtering yield is analyzed distinguishing among particles leaving the target across the nanoisland surface and across the flat substrate surface without nanoparticle above, and the sum of both. Results consider the dependence on two factors: the spacing among nanoislands and the nanoisland height.

1. Introduction

Nowadays, the deposition of size-controlled clusters on metal and carbon-based substrates is of notable technological interest. The (magnetic, catalytic, electrical, biological, etc.) properties of these nano-dimensional islands on a substrate are used in different fields of knowledge to create different nanostructured devices [1]. Additionally, the modification of the characteristics of this type of materials by ion irradiation may be very interesting in the near future. The interaction of isolated nanoparticles [2] or nanostructured surfaces formed by nanoislands (NIs) [3] with irradiation is linked to new processes of energy deposition and exchange. During the irradiation of bulk materials, the collision spikes are cooled due to mechanisms of heat transport. Unlike, in the case of irradiation of NIs or nanoparticles, these mechanisms are concentrated in certain directions (from the NI towards the substrate) or are suppressed, respectively. Phenomena such as desorption (or sputtering of full NIs) [4] are speculated that they are produced by this concentration and marked anisotropy in the flux of energy.

In this work, investigations are focused on the study of sputtering mechanisms produced by the ion irradiation in a material with a high NI density. Two points of view are used, on the one hand, the molecular dynamics (MD) simulation, and on the other hand, the predictions of an analytic model developed from the Sigmund's theory [5]. Results from both methods are compared and consequences are drawn. The system used for the analysis is a binary nanostructured material formed by a periodical array of cobalt clusters on a (001) copper substrate bombarded at 1 keV with argon ions. At this regime, most energy is deposited within the Co NI. This effect produces its slow disintegration by sputtering and mixing. Hence, the height of all the NIs decreases uniformly on the substrate surface during the bombardment [6]. The sputtering yield is the main analyzed parameter, distinguishing the exit point of sputtered atoms: across the NI surface or the substrate surface without cluster above. Previous studies show that the large surface-to-volume ratio of NIs increases the number of sputtered atoms, that is, produces an enhanced sputtering, compared to bulk surfaces [2,7].

2. Computational and analytic methods

The bombardment of a target formed by Co nanoislands on a Cu (001) substrate with 1-keV Ar was studied using MD. Atomic interactions were described by a second-moment tight-binding approximation (TB-SMA) potential [8]. At short distances, the Co-Cu interactions were modelled by a universal repulsive Ziegler-Biersack-Littmark (ZBL) potential [9]. Both potentials were smoothly linked. The Ar-Cu and Ar-Co interactions were also described by the ZBL potential. Equations of motion also included inelastic energy losses for atoms with kinetic energy higher than 10 eV [9].

The bombarded system was perfectly periodical and without defects. To achieve this, the clusters were laid over the substrate instead of being deposited at very low energy, and then the system energy was minimized. The target was formed by 9 NIs symmetrically placed in the shape of a two-dimensional square Bravais lattice. The NIs were separated by a periodical length e (see Fig. 1). In the simulation results, the NIs were created from a Wulff-shaped cluster of 586 atoms. Three adjacent (001) layers

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http://dx.doi.org/10.1016/j.cplett.2016.10.017
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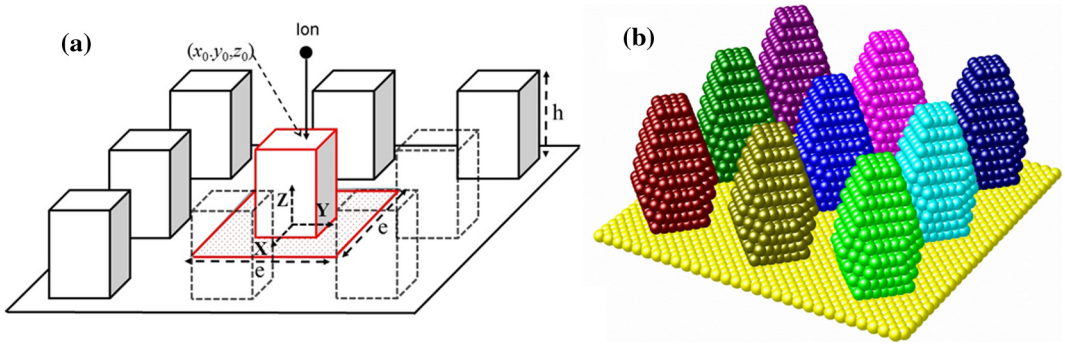

Fig. 1. (a) Periodical parallelepipedic nanoisland array used in the analytic results. (b) Truncated Wulff-type polyhedron array used in the simulation results. From one system to other, only the NI shape is modified, but not the dimensions. The impact point on the target surface (surface S_0) can be any point inside the square of side length e.

of this polyhedron were truncated. Thus, this shape was more like a deposited cluster at very low energy. Three values for the space between nanoislands e were considered: $e = 43.3$ Å (case L), $e = 36.1$ Å (case M) and $e = 28.9$ Å (case S). Three substrate sizes with periodic boundary conditions (PBC) matched their dimensions with these gaps: $36 \times 36 \times 24$, $30 \times 30 \times 24$ and $24 \times 24 \times 24$ unit cells, respectively. For a truncated Wulff-shaped NI, the sputtering effective cross-section σ_{eff} was about 491 Å^2.

Non-periodic boundary conditions were used in the direction normal to the Cu substrate (z-direction), whereas PBC were used in the x- and y-directions, both parallel to the substrate surface. The two bottom layers perpendicular to the z-direction were fixed. Over these, three layers controlled thermally the system using the generalized Langevin equation proposed by Adelman and Doll [10]. These layers absorbed heat after each impact in order to maintain a temperature of 5 K, that is, minimizing the influence of temperature in the process. Nevertheless, in some cases 300 K were also used.

During the bombardment, the NI height decreased with increasing fluence. In fact, this drop in simulation and experimental works [7,11] is approximately exponential. For a uniform surface bombardment, the change in height is the same for each NI. The problem is that the computer simulation of this type of bombardment for obtaining acceptable statistical averages of sputtering yield would mean to use a very large simulation time. Due to this, an alternative procedure was implemented with roughly similar results. In this method, the ion only collided with the central cluster and its surroundings (the target surface was inside a square of side length e: surface S_0, see Fig. 1). When a layer in the central NI was removed, all the layers of the same height of the rest of NIs were also removed. This new system did not differ too much from the state of the system after the removal of all the NI layers placed at the same height during a uniform bombardment. Obviously, the effects induced by the bombardment of lateral NIs are not taken into account. The sequence of elimination of layers parallel to the substrate surface was repeated and finished when all the NIs disappeared completely. The projectiles were Ar ions at 1 keV striking normally the target at a random position inside the surface S_0. The time of monitoring of a collisional process derived from an ion was about 20 ps. The number of simulations, although different for the cases L, M or S, was enough to gather a good statistics.

An analytic expression for the sputtering yield of a periodical NI array was derived from the Sigmund's theory [5]. In this model, sputtering yield is obtained from the damage energy deposited by the incoming ion on the target surface, S. The yield for an ion impacting the surface at $\vec{r}_0(x_0, y_0, z_0)$ is:

$$Y(\vec{r}_0) = \Lambda \int_S dS\, F(\vec{r}, \vec{r}_0) \tag{1}$$

where Λ is a proportionality constant, $F(\vec{r}, \vec{r}_0)$ is the damage-energy distribution function and $\vec{r}(x,y,z)$ is the variable defining the target surface S. The three spatial coordinates are not independent for $\vec{r}$ and $\vec{r}_0$, and therefore z and z_0, respectively, depend on the other two coordinates. In our case, according to the position of the reference frame (the OXY-plane coincides with the flat substrate surface and the OYZ- and OXZ-planes are two symmetry planes of the central NI, see Fig. 1), the parameter z_0 (position where the ion impacts) is equal to h on the NI surface, and 0 on the substrate surface.

The sputtering yield is obtained by averaging the parameter $Y(\vec{r}_0)$ over the different impact points (surface S_0), i.e.:

$$Y = \int_{S_0} dS_0\, Y(\vec{r}_0)/S_0 \tag{2}$$

In this work, a Gaussian approximation for the damage distribution was used. This choice is considered reasonable in previous works [12,13]. The expression for this distribution is:

$$F(\vec{r}, \vec{r}_0) = \frac{E}{(2\pi)^{3/2}\alpha\beta^2} \exp\left(-\frac{1}{2\alpha^2}[z - (z_0 - a)]\right)^2 \times \exp\left(-\frac{1}{2\beta^2}\left([x - x_0]^2 + [y - y_0]^2\right)\right)$$

where $E = 1$ keV is the ion energy (total deposited energy); z_0-a is the z-coordinate of the centre of the damage distribution; and α, β are the standard deviations of this distribution. Two sets of values were used for these parameters to take into account the different characteristics of material. Thus, $a_c = 8.7$ Å, $\alpha_c = \beta_c = 19.0$ Å, if the impact point was on the Co NI; and $a_s = 4.2$ Å, $\alpha_s = \beta_s = 15.9$ Å, if this point was on the Cu substrate. These values were obtained from bombardment simulations of Co/Cu bilayer samples (9 Co layers ~15.8 Å in thickness) and of Cu samples, respectively, in order to avoid the influence of surface roughness and always using a suitable statistics.

The analytic expression for Y was obtained from a periodical distribution of parallelepipedic NIs of width and length d and height h and separated each other a distance e. This shape is a good approximation to the real aspect of these NIs during the bombardment [7]. Both Eqs. (1) and (2) are evaluated in Appendix A, assuming that the width d does not vary during the bombardment. The effective cross-sections of both the parallelepipedic and the Wulff-shaped NIs were taken to be the same, therefore the variable d must be: $d = \sqrt{\sigma_{eff}}$. The ideal situation is to choose some experimental value of sputtering yield Y for the initial system to calculate the parameter Λ in the analytic model. Failing that, any other simulation value could be taken, always with the system without bombarding to have the composition and, therefore, the