Simulations of submicrometer-scale roughening on ion-bombarded solid surfaces

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Roughening of amorphous carbon surfaces bombarded by 5-keV Ar ions is studied by using Monte Carlo simulations. Sputtering-induced erosion is treated in detail by simulating the entire collision cascades originated by the bombarding Ar ions. Surface relaxation due to diffusion is described by a Wolf-Villain-type discrete model. The simulations show that bombarded surfaces have self-affine topography on the submicrometer-scale. For normal incidence the roughness exponent is $\alpha \approx 0.37-0.45$, diminishing to $\alpha \approx 0.25$ when the angle of incidence is increased to 60°. In the studied cases the roughness exponent α is nearly independent of the relaxation of the surface. The dynamic exponent z and growth exponent β show a clear dependence on the relaxation. Without relaxation we found values $\beta \approx 0.3$ and $z \approx 1$, and with relaxation $\beta \approx 0.14-0.20$ and $z \approx 1.6-2.6$. [S0163-1829(96)04444-X]

Recent experiments with roughening on surfaces bombarded by heavy ions have established that on the submicrometer scale the roughening has well defined and nontrivial statistical properties, well described by self-affine topography.[1,2] In the experiments the characteristic value w (e.g., the rms value) of the surface height fluctuations on ion-bombarded surfaces is observed to scale with sample size L according to relation $w \propto L^{\alpha}$, where α is the roughness exponent. Values $\alpha \approx 0.2-0.4$ have been reported for graphite surfaces,[1] and $\alpha = 0.53 \pm 0.02$ for the Fe surface,[2] both bombarded by 5-keV Ar ions. During the initial stage of the roughening $w \propto \Phi^{\alpha/z}$, where Φ is the fluence, and the dynamical scaling is thus characterized by the dynamic exponent z (or by the growth exponent $\beta = \alpha/z$). In the case of the graphite surface the dynamical exponent in the range $z = 1.6-1.8$ was found to be compatible with the data.[1] These values are valid at sufficiently small spatial scales, approximately below 100 nm, and they are not far from the prediction $\alpha \approx 0.38$ and $z \approx 1.58$ of the Kardar-Parisi-Zhang (KPZ) model[3] of nonequilibrium surface growth.

Although experiments indicate the applicability of scaling theory in the description of the statistical and dynamical properties of ion-bombarded surfaces, at present there is no adequate theory to explain the experimental results of the submicrometer-scale roughening. This situation differs from the current state of the understanding of roughening and ripple formation in length scales of micrometers, where models based on noisy Kuramoto-Sivashinsky dynamics offer an explanation for the observed structure formation.[4] These recent advances give only little guidance for the modeling of the submicrometer-scale roughness. There is some indication[5] that the roughening could possibly be modeled in terms of Edwards-Wilkinson-type models,[3] where the shot noise is taken to be proportional to the recoil displacements in collision cascades. However, the role of recoil displacements in the roughening and formation of surface corrugations is not obvious, and a more direct approach based on the sputtering process itself is clearly desirable.

In this Brief Report we present computer simulations of the submicrometer-scale roughening of amorphous graphite surfaces bombarded by 5-keV Ar ions. Sputtered ions emerge from collision cascades which are calculated collision by collision using the Monte Carlo method. The relaxation of the surface is assumed to occur through surface diffusion, which is described by a Wolf-Villain-type model.[3] Simulations are realistic in the sense that erosion of the surface occurs as a result of the sputtering of single ions, and relaxation also occurs on an atomistic level.

The essence of the model is a fast simulation of collision cascades by using a realistic interaction potential for colliding atoms. The simulations were carried out using a computer code which is a modification of COSIPO.[6] This code was chosen because it is known to be adequate for a description of most kinetic impact phenomena, and because it has been successfully used in the simulation of the sputtering from planar and slightly corrugated surfaces bombarded by 5–10-keV heavy ions.[6] The computer code simulates collision cascades in the so called binary collision approximation, and the interaction between atoms is described by a screened Coulomb potential, in practice identical to the Kr-C potential.[6] In addition to other standard features of the code (for details, see Refs. 6) we made some modifications which enable a sufficiently rapid (within the limits of computer resources available to us) updating of the surface structure. The surface area is limited to 100×100 atoms (about 20×20 nm^2), and the target is divided into boxes which contain a single carbon atom. Periodic boundary conditions are used. Atoms, which are moving outwards from the (local) surface, are allowed to sputter when their energy exceeds the surface binding energy $E_b = 7.5$ eV, derived from the heat of sublimation. After leaving the surface the sputtered atoms are not allowed to stick to the surface again, so that the possibility of redeposition is excluded. The surface recession is realized by removing the box from the surface, which is closest to the initial site of the sputtered atom. This practice finds its justification from the notion that most of the sputtered atoms originate from the surface layer. The shadowing effects are taken into account in the simulations by demanding that all collisions occur below the instantaneous surface position at the impact site.

The surface corrugations created by the ion impacts are not stable, and they tend to smooth out by the effect of surface diffusion. On an atomistic scale the diffusion current is generated by atoms that move until the surface free energy

(or chemical potential) is minimized. Simplified discrete models,[7,8] which are believed to contain the essential physics of the surface diffusion, are based on irreversible jumps of atoms guided by the rule to maximize the bonding or number of the nearest neighbors. We have chosen to describe the surface diffusion by a Wolf-Villain-type model,[7] where each atom hops with a given probability p, within a fixed distance l of its initial place, to a site where it attains a maximum number of neighboring atoms; i.e., the bond number is maximized. In practice we use $p=1$ and $l=1$ (to be referred to as model $R1$) or $l=2$ (referred to as model $R2$) to simulate the effect of relaxation. The relaxation process is realized after the completion of a collision cascade originated by each impinging ion. Because each atom completes the diffusion process instantaneously, the relaxation model simulates surface diffusion but without the problems of having competing kinetic rates and a continuous migration of atoms, which would lead to long crossover effects.[7,8]

The values of scaling exponents for the Wolf-Villain model in $(2+1)$ dimensions are $\beta=0.19$ and $\alpha=0.66$ and for the closely related Tamborenea–Das Sarma model[8] $\beta=0.24$ and $\alpha=0.95$.[3,9] In models where surface diffusion is dominant, the dynamic exponent z is related to roughness exponent through $z=2\alpha+d$, where $d=2$ is the dimensionality of the surface.[7,9] The rather large scaling exponents α and z are typical of a roughening process governed by diffusion, since although the topography is smoothed on the atomistic scale, larger-scale terracelike structures tend to be stabilized.[7]

The most straightforward way to examine the statistical properties of the surface is to monitor the mean-square width $w^2(L,\Phi)=L^{-d}\Sigma_{\mathbf{r}}\langle[h(\mathbf{r},\Phi)-h(\Phi)]^2\rangle$ of the fluctuating d-dimensional surface of size L^d, where $h(\mathbf{r},\Phi)$ and $h(\Phi)$ are the instantaneous and average surface positions, respectively, at fluence Φ. If the surfaces is self-affine its width $w(L,\Phi)$ is found to obey the scaling laws[3]

$$w(L,\Phi)\propto\begin{cases}L^{\alpha}, & \Phi\to\infty\\ \overline{h}^{\beta}, & \overline{h}\ll L^{z},\end{cases} \tag{1}$$

where β is the growth exponent, connected to roughness exponent α and dynamic exponent z by the relation $\beta=\alpha/z$.[3] Note that we have chosen to represent the scaling in terms of the average position of the surface instead of the more obvious possibility $w\propto\Phi^{\beta}$. This is done in order to exclude the effect of the steady growth of the mean sputtering yield (and thus the steady increase in the mean recession speed of the surface) during the initial stage of the roughening.

In practice the scaling exponents are determined by exploiting the scaling behavior of the height-height correlation function[3]

$$G(\mathbf{r},\Phi)=L^{-d}\sum_{\mathbf{x}}\langle[h(\mathbf{r}+\mathbf{x},\Phi)-h(\mathbf{x},\Phi)]^2\rangle=r^{2\alpha}g(r/\xi), \tag{2}$$

where the correlation length $\xi\propto\Phi^{1/z}$ in scaling function $g(x)$ corresponds to the wavelength of a typical fluctuation. The correlation length separates the two regions of interest, where $g(x)\simeq x^{-2\alpha}$ and thus $G(r,\Phi)\simeq\Phi^{2\alpha/z}$ and the saturation region, where $g(x)=$const and thus $G(r,\Phi)\sim r^{2\alpha}$. The latter relation is used in the following for the extraction of exponent α. The growth exponent β can be determined from the relation $w^2(L,\Phi)=(L^{-d}/2)\Sigma_{\mathbf{r}}G(\mathbf{r},\Phi)$, leading directly to the scaling laws given in Eq. (1), which are then used to extract the exponent β.

Simulations were performed for amorphous carbon surfaces, which were bombarded by 5-keV Ar ions. The oblique incidence of the beam was varied in the range of 0°–60° and simulations were carried out up to fluences $\Phi\approx3\times10^{17}$ ions/cm^2, corresponding typically to 10^6 cascade histories. Stationarity (or saturation) of surface height fluctuations is typically reached at fluences around 10^{16} ions/cm^2. An example of the topography of the surface, bombarded at normal incidence and without relaxation, is shown in Fig. 1(a). One can see that, in the absence of relaxation, sharp peaks remain on the surface. In reality the gradients of such features are so high that they cannot be stable, and will eventually relax. The effects of relaxation were studied by repeating the calculations with relaxation models $R1$ and $R2$. The resulting

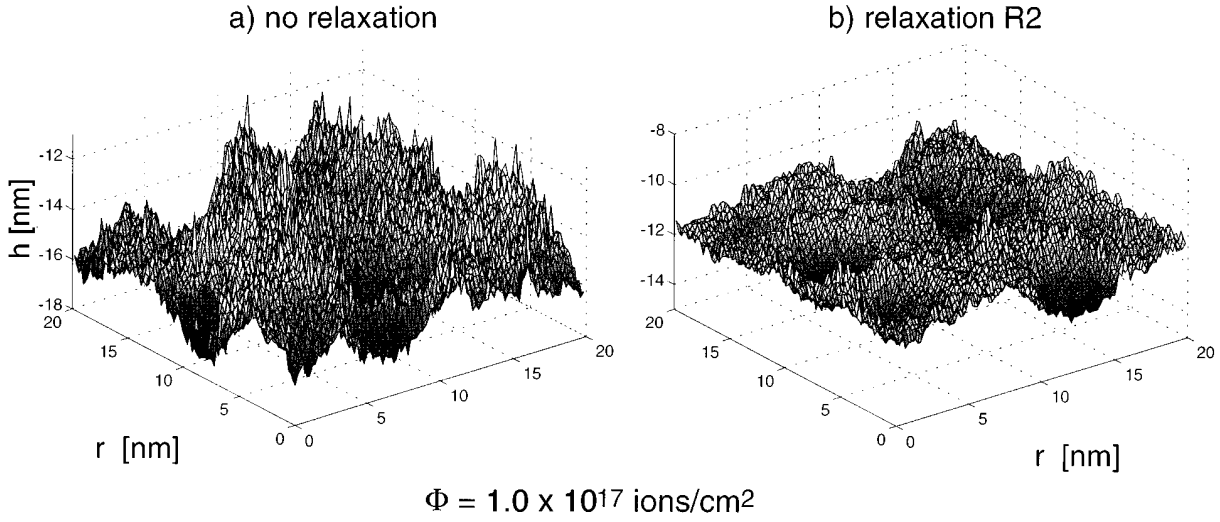

FIG. 1. Erosion of the carbon surface due to 5-keV Ar-ion bombardment at normal incidence, (a) without surface relaxation and (b) with relaxation model $R2$. The fluence at the cases shown is $\Phi=1.0\times10^{17}$ ions/cm^2. An initially flat surface was located at a position $h=0$.

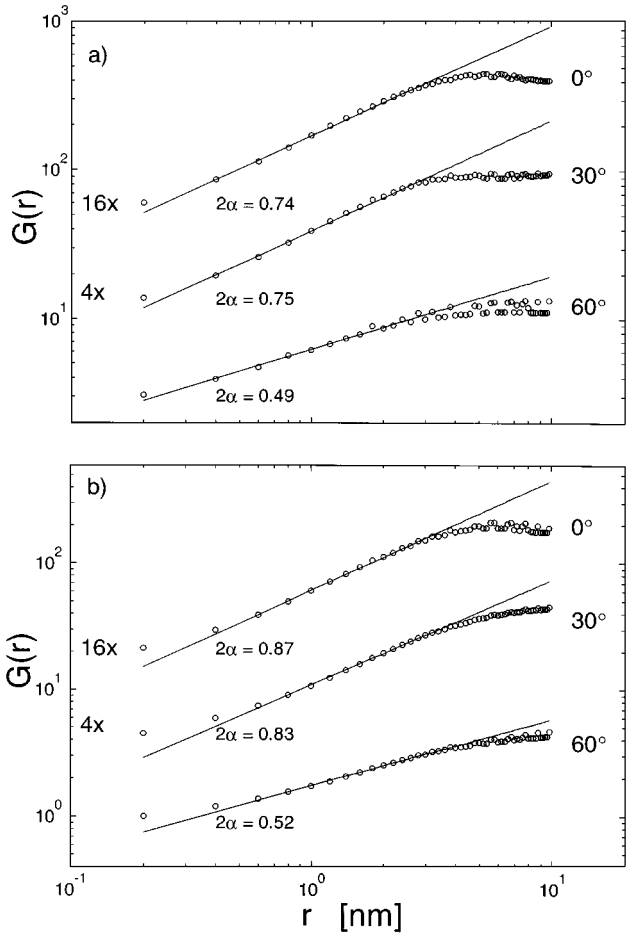

FIG. 2. Height-height correlation function $G(r)$ at saturation for a surface bombarded by 5-keV Ar ions, (a) with no relaxation and (b) with relaxation model $R2$. Results of the bombardment at angles of incidence 0°, 30°, and 60° (from the surface normal) are shown. Slopes of the fits given in the figure correspond to 2α. For better visibility curves are displaced, multiplying them by a given factor 4 or 16.

surface topography for model $R2$ is shown in Fig. 1(b). Comparing the cases shown with results in Fig. 1(a), one can see that relaxation smooths the highest peaks on an atomistic scale. However, the larger-scale structures [in Fig. 1(a) largely masked by sharp peaks] have not changed much, and the overall appearance is a craterlike topography.

The height-height correlation function $G(r)$ (at the saturation) without relaxation is shown in Fig. 2(a), and it is seen to be a power law in the region 0.4–3 nm, i.e., on a scale corresponding to the cascade dimensions. On larger scales the correlation function begins to level off due to the effect of boundary conditions. A fit done on the straight part of the correlation function gives the roughness exponent $\alpha \approx 0.37 \pm 0.02$ for the bombardment at a normal and slightly oblique incidence of 30°. Bombardment at large incidence angles of 60° gives a substantially smoother surface with $\alpha = 0.25 \pm 0.02$. The smoothing of the surface with increasing angle of incidence is to be expected. The corresponding results for relaxation model $R2$ are shown in Fig. 2(b). When the relaxation mechanism is operating, the topography of the bombarded surface on an atomistic scale is smoother and rms fluctuations are substantially reduced, as can be seen by comparing the results in Fig. 2(b) to those in Fig. 2(a). However, the scaling properties of the topography are relatively unaffected, and the height-height correlation function $G(r)$ at the saturation yields now $\alpha \approx 0.42$–0.44 at normal or near-normal incidence, and $\alpha \approx 0.26$ at large incidence angles. The detailed values of the scaling exponents for the relaxation models $R1$ and $R2$ are summarized in Table I. The slight increase in the scaling exponents is probably due to diffusion-driven terrace formation.

TABLE I. Roughness, growth, and dynamic exponents α, β, and $z = \alpha/\beta$, respectively, for carbon surface roughening due to 5-keV Ar-ion bombardment. Exponents $z_G = 2 - \alpha$ and $z_D = 2\alpha + d$ are estimates based on the assumed Galilean invarience (G) or dominance of diffusion (D), respectively. In values of α and β the statistical error of the fits are at most 0.02.

Relax.	Θ	α	β	z	z_G	z_D
none	0°	0.37	0.34	1.1	1.6	2.7
	30°	0.38	0.34	1.1	1.6	2.8
	60°	0.25	0.33	0.8	1.7	2.5
$R1$	0°	0.45	0.23	2.0	1.5	2.9
	30°	0.47	0.22	2.1	1.5	2.9
	60°	0.31	0.20	1.6	1.7	2.6
$R2$	0°	0.44	0.17	2.6	1.6	2.9
	30°	0.42	0.16	2.6	1.6	2.8
	60°	0.26	0.14	1.9	1.7	2.5

The growth exponent β is extracted from the initial values of the rms width $w(L,\Phi)$, shown in Fig. 3(a) for a surface with no relaxation, and in Fig. 3(b) for relaxation model $R2$. Without relaxation the growth exponents are in all cases rather similar, $\beta \approx 0.33$–0.34, (detailed values are reported in Table I). The dynamic exponent $z = \alpha/\beta$ calculated on the basis of values for α and β fall between limits $z = 0.8$ and 1.1, which are lower than values $z = 1.6$–1.8 obtained from relation $z = \alpha - 2$ (i.e., assuming Galilean invariance;[3] see also the analysis of experiments[1]). On the other hand, it is clear that the dynamic exponent has values different from $z = 2\alpha + d$, which holds exactly for models where diffusion processes are dominant. Although the roughness exponents remain relatively unaffected by the relaxation, this is not the case for the growth and dynamic exponents. The growth exponent tends to decrease, while the dynamic exponent correspondingly increases, approaching the value $z = 3.2$ for the Wolf-Villain model. Furthermore, in the presence of diffusion, the exponents are more compatible with relation $z = 2\alpha + d$ than the relation $z = 2 - \alpha$ required by the Galilean invariance.

The results of the present simulations are in accordance, if not in complete agreement, with the experimental findings, supporting the argument that ion-bombarded surfaces are self-affine, and that the self-affine roughness results from the elementary sputtering process. The important point of the present results is the apparent insensitivity of steady-state roughness to diffusion or relaxation in the studied scale. This agrees with experiments,[1] where small-scale scaling is found to be relatively independent of the ambient temperature of the surface even up to 900 K. This insensitivity is partially explained by the extremely good thermal stability and high

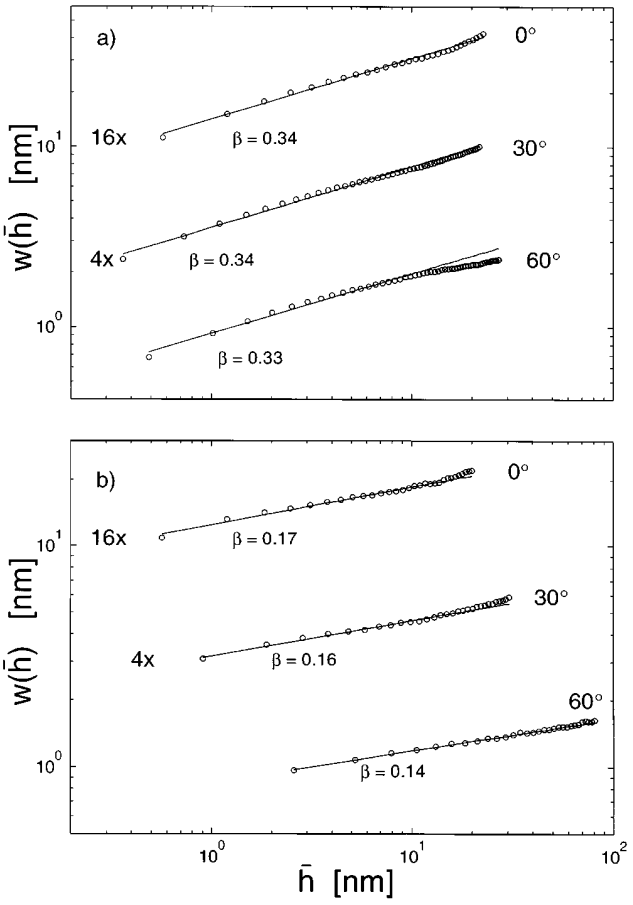

FIG. 3. The rms width $w(\bar{h})$ of surface height fluctuations during the initial stage of the bombardment, when $h \ll L^z$ (now $L=100$), (a) with no relaxation and (b) with relaxation model $R2$. Results of the bombardment at angles of incidence 0°, 30°, and 60° (from the surface normal) are shown. Slopes of the fits given in the figure correspond to β. For better visibility curves are displaced, multiplying them by a given factor 4 or 16.

melting temperature $T_M \approx 3800$ °C of graphite, which enables the surface corrugations to be ''frozen in.'' However, the diffusion certainly affects the scaling properties on larger scales, where structures governed by diffusion begin to emerge.[1]

The scaling exponents found in the present study are not explained by any known nonequilibrium growth model, for example in the KPZ model (2+1 dimensions) $z=1.58$, $\alpha=0.38$, and $\beta=0.24$ (for a summary of α, β, and z in known cases, see Ref. 3). Furthermore, the known continuum models do not allow changes in the dynamic exponent. On the other hand, without the relaxation the agreement with KPZ results is close enough to warrant a closer search for KPZ-like nonlinearity in the erosion.

Within the framework of the existing models, the variability of α indicates either the presence of spatiotemporal correlations or anomalous noise.[3] Our previous model of surface roughening based on recoil displacements[5] allowed for variations in roughness (and growth) exponent, due to inclusion of power-law noise. In order to detect a sign of this type of noise due to sputtering erosion, we monitored the distribution of normalized surface height fluctuations. No sign of the power-law noise was detected, and we were thus able to rule out the explanation of the anomalous exponents in terms of power-law noise originating from the sputtering process. However, sputtering is perhaps not the only process generating surface corrugations, because low-energy impacts are also known to create persistent ''bumps'' in the bombarded surface,[10] and their role in the roughening is yet to be studied. Other aspects warranting closer study are the detailed role of nonlinear effects due to shadowing and redeposition, which are, however, tasks beyond the scope of the present objective of demonstrating the existence of self-affine submicrometer-scale roughness.

In conclusion, simulations suggest that ion bombardment creates self-affine, rough surfaces, and the formation of self-affine topography on the submicrometer scale can be attributed to the elementary sputtering process. The values of roughness exponents in the studied cases are in the range $\alpha \approx 0.25-0.47$, for angles of incidence in range 0°–60°, smoother surfaces corresponding to larger angles. The roughness exponent α was found to be relatively insensitive to the relaxation process on spatial scales below 10 nm. The growth exponent β and dynamic exponent z were found to depend on the relaxation model used, which is according to the expectations. Without relaxation values $\beta \approx 0.3$ are obtained, and no significant dependence on the angle of incidence are found. The corresponding dynamic exponent is $z \approx 1$. When relaxation to sites with the largest number of nearest neighbors within a distance of nearest neighbors, values $\beta \approx 0.20-0.23$ and $z=1.6-2.1$ are obtained. Increasing the jump length to two nearest neighbors, $\beta=0.14-0.17$ and $z \approx 1.9-2.6$ are obtained. Clearly the inclusion diffusion drives the dynamic exponent closer to the values $z \approx 3.2-4$ typical of diffusion-dominated processes. The results are in accordance with, if not similar to, the experimental results for carbon surfaces bombarded by 5-keV Ar ions.

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