Scanning Tunneling Microscopy Observation of Self-Affine Fractal Roughness in Ion-Bombarded Film Surfaces

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We describe a straightforward method for obtaining a precise value for the fractal dimension (self-affine scaling exponent) of a surface, via scanning tunneling microscopy. It is applied to investigate ion-beam erosion of an iron film surface, providing strong support for the applicability of scaling theory to submicron erosion processes. A self-affine surface with scaling exponent $H=0.53\pm0.02$ is observed to develop.

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In deposition processes used to fabricate thin solid films, there is a very sensitive and complex dependence of film microstructure on growth conditions [1]. This microstructure has a profound effect on all physical properties (electrical, mechanical, optical, etc.) of the film, and must be characterized at nanometer length scales in order to gain an understanding of its physical origins. Films deposited under nonequilibrium conditions (as is typically the case) have recently been the subject of much atomic-scale computer simulation and scaling theory [2], and are generally expected to have self-affine fractal surfaces [3,4].

The submicron topology of a surface produced by ion bombardment or erosion has received far less theoretical attention than that of a vapor-deposited film [5]. It is possible, however, that the scaling theories applicable to nonequilibrium film growth may also be applicable to ion bombardment, so long as no eroded material is redeposited onto the surface [6]. If so, the topography of a film deposited onto an initially smooth substrate should be quite similar to that of an initially smooth surface which is subjected to ion bombardment: A self-affine fractal surface is expected to develop, and its root-mean-square (rms) roughness should increase with time [7]. At the tens of microns level, ion-eroded surfaces are generally not observed to be similar to those of vapor-deposited films [8]. Nonetheless, the rms roughness (generally not treated as a fractal or scale-dependent quantity) of such surfaces does usually increase with bombardment time [9,10]. At submicron levels, ion-bombarded surfaces have in fact been reported to be similar to those of vapor-deposited films [11,12], and initially flat surfaces have also been observed to roughen upon exposure to ion bombardment [5,13].

Whether the study involves film growth, ion bombardment, or a myriad of other topics, a knowledge of surface topography and roughness at submicron length scales is frequently essential to understanding the physical origins of that which is under study. We focus here on the use of scanning tunneling microscopy (STM) for the characterization of surface roughness, with an emphasis on self-affine fractal roughness. We report measurements carried out on an epitaxially grown iron film which has been subjected to Ar^+-ion bombardment, motivated by recent observations that the nonequilibrium growth behavior of iron is well described by scaling theory [14]. Previous STM studies of self-affine roughness have been carried out on samples which were held in air [13,15], with the role of surface contamination being uncertain. The measurements which we report here have been carried out in ultrahigh vacuum so as to remove this uncertainty. We observe a self-affine fractal surface to develop on account of the ion bombardment, and obtain a precise value for the scaling exponent, $H=0.53\pm0.02$. Our results provide strong evidence that the applicability of scaling theory to describe submicron erosion processes may extend far beyond that of a previous case study of single-crystal graphite erosion by Ar^+ ion bombardment [13].

A self-affine surface is distinguished from a "genuine," or self-similar, fractal by an asymmetry in the scaling behavior perpendicular to the surface, generally manifested by an absence of surface overhangs [7]. It is therefore generally treated as a single-valued function $h(x,y)$, with average height h and rms roughness (rms "width") $\sigma=\langle[h(x,y)-h]^2\rangle^{1/2}$. The surface is termed self-affine if σ increases with the horizontal length L sampled according to $\sigma\propto L^H$, where the scaling exponent $0<H<1$ is indicative of the texture of the surface [3,4]. It is commonplace in the literature to parametrize a self-affine surface by either the scaling exponent H itself, or a fractal dimension $D=3-H$ associated with that exponent [4,15].

A procedure for determining the value of H from a *single* STM image has been described in detail by Mitchell and Bonnell [15]. Surface profiles recorded in the fast scan direction are Fourier analyzed and the coefficients for the individual profiles are then averaged. If a log-log plot of the integrated power spectrum (a log-log plot of the sum from k to ∞ of the squared Fourier amplitudes versus k, where k is the wave number) is a straight line, then the sample is considered to be self-affine, and the slope of the line will be $-2H$ [15,16]. Commercial instruments can determine H to within about ±0.1 by this method, primarily limited by number of points per scan

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which can be recorded [15] (200–400 is the typical maximum).

Much better precision can be obtained, however, if *multiple* images of varying scan size L are recorded instead of a single image. Combining more than one image is potentially quite problematic since the overall tilt of each image from scan to scan is due to both the macroscopic tilt of the sample and the local slope of the intrinsic surface roughness. We show here how the Fourier analysis approach outlined above can be accomplished in a straightforward manner on multiple images. Since this approach has not been previously utilized, we first illustrate the technique through measurements recorded on a test gold film sample which exhibits self-affine scaling over 3 orders of magnitude, and then present our results for the iron film sample.

The test sample consisted of a gold electrode present on the surface of a commercial rate-monitor crystal [17], whose scaling exponent had been determined by an independent experimental technique (adsorption [18]) to be 0.98 ± 0.1 [19]. Despite the fact that this exponent fell within experimental error of an integer value, STM images recorded on the sample exhibited the visual self-similarity characteristic of a fractal surface (Fig. 1).

The sample was examined in both atmospheric and ultrahigh-vacuum conditions, with two different commercial STM's and with two different tip materials. The measurements in air were carried out with a Digital Instruments, Nanoscope II microscope operating in constant-current mode (1 nA; 25 mV) with a Pt-Ir tip (Fig. 2, upper data set). A variety of scans, each of size L, were recorded at random locations on the surface. The σ values for the rms roughness given by the instrument for the individual scans were then averaged. This procedure was repeated for numerous different scan sizes, generating a set of average σ values vs L. All values of σ were computed *after* the instrument's plane fitting and subtraction procedure had been carried out; thus *each σ value was computed on an image which appeared to be level.* This plane fitting procedure is standard on commercial instruments to account for the fact that virtually all samples are macroscopically tilted with respect to the horizontal scan directions of the microscope. It removes, however, not only the slope due to macroscopic tilting, but also those components of the intrinsic surface roughness whose characteristic wavelengths significantly exceed the scan size. The components of the intrinsic roughness spanning the range in wavelength from 0 to $\approx 2L$, or in wave number from $\approx 1/(2L)$ to ∞, are the primary constituents of σ values obtained in this manner. The function $\sigma^2(k)$ will therefore have scaling properties identical to that of an integrated Fourier power spectrum, and a log-log plot of σ vs scan size L will have slope H.

The value $H = 0.96 \pm 0.02$ determined for the test sample by this procedure is far more precise than that obtained via adsorption or through analysis of a single STM image. It is also quite robust: In the course of data recording for the upper curve, the scan head was changed once and the scan tip was changed three times, with no associated disruptions or discontinuities in the plot. The upper horizontal cutoff to scaling may be just evident by the flattening of the plot which seems to be present for scan sizes exceeding 5×10^4 Å.

The lower set of data points in Fig. 2 were recorded

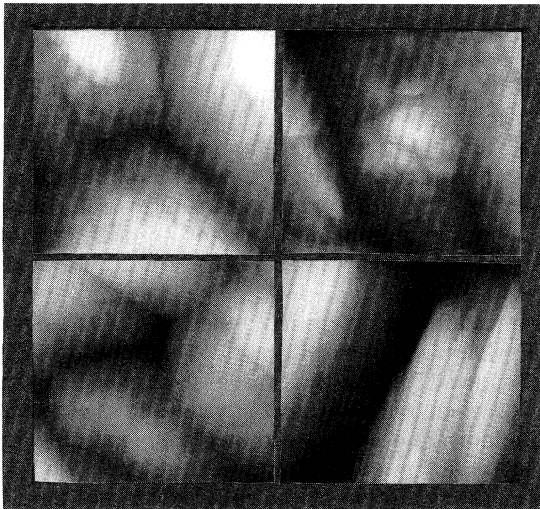

FIG. 1. Top view of STM images recorded on the gold film test sample. The scan sizes, clockwise, starting in the upper left corner are (2000 Å)^2, (5000 Å)^2, (20000 Å)^2, and (500 Å)^2. The corresponding vertical scales (black to white) are 1000 Å, 2000 Å, 4000 Å, and 200 Å.

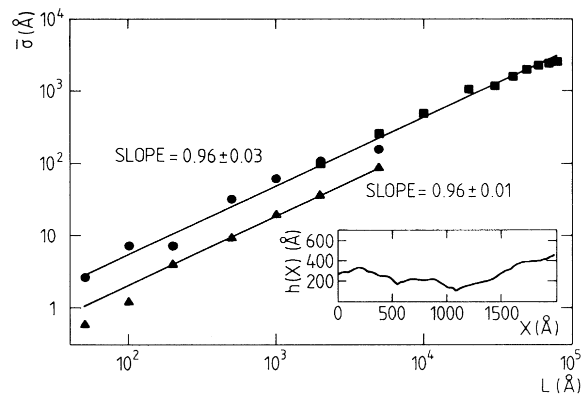

FIG. 2. Average root-mean-square roughness vs scan size for the gold film test sample. Circles: recorded in air with a short-range scan head; squares: recorded in air with a long-range scan head; triangles: recorded in vacuum with a short-range scan head. Each point in the upper (lower) data set represents an average of 5 (10) scans recorded at random locations on the surface. The solid lines depict least-squares fits to the data. The slope of these lines provides a measure of the scaling exponent $H = 0.96 \pm 0.02$. Inset: A vertical profile $h(x)$ typical of this sample.

with a WA Technology (Cambridge, U.K.) microscope on the sample after it was passed through a load-lock into an ultrahigh-vacuum chamber held at 10^{-10} Torr. (We note that no baking, sputtering, or cleaning of the sample occurred between its examination in air and in UHV.) The data were recorded with a tungsten scan tip in constant-current mode (1 nA, 50 mV), and the value of H compared favorably with that determined in air. The σ values recorded on the vacuum STM were, however, 2.5 times smaller than those recorded in air. The presence (or lack) of vacuum conditions, tip radius, tip material, tip bias, and tunneling current level were eliminated as sources for this discrepancy. We attribute the difference to an inconsistency in the z calibration of the two instruments. (Independent scans on single-crystal graphite revealed no discrepancy in the x-y calibrations of the two instruments.)

The ability to change scan heads allows STM characterization of scaling, such as that shown in Fig. 2, to be carried out in principle over 5 orders of magnitude. This is a far greater range than that available through alternate techniques such as adsorption and x-ray scattering [20]. The instrument must, however, be carefully calibrated (for example, via atomic step edges), if an absolute value for σ is required. Observation of the upper horizontal cutoff to scaling is limited only by the range of the available scan heads. The lower cutoff to scaling is sensitive to tip curvature effects, evidenced by the fact that we observed different lower cutoffs to the scaling behavior for data sets recorded with different tips. STM is incapable of detecting any porosity or surface overhangs in the outer surface topology, and is also sensitive to chemical inhomogeneities. These constraints are not, however, considered to be problematic for the system under consideration here: Ion bombardment is not known to produce surface overhangs, and the sample which was studied was carefully guarded in ultrahigh-vacuum conditions.

The iron film sample, originally 2000 Å thick, was deposited by means of molecular beam epitaxy onto MgO(100) held at 150 °C, so as to produce Fe(100) planes parallel to the MgO surface. It was then transferred through air to the UHV chamber containing the WA Technology STM and sputtered so as to remove an oxide layer which had formed due to the air exposure. Heating the sample to ≈ 600 °C annealed and flattened the surface, which was examined *in situ* with Auger spectroscopy to assure no contaminants were present, before commencing the STM studies. STM measurements were carried out *in situ* both before and after bombardment with Ar^+ ions. Measurements could be repeated indefinitely since reannealing the sample returned it to its original morphology.

The lower set of data points in Fig. 3 was recorded (0.1 nA, 500 mV) immediately after the annealing process. The surface is composed of atomically stepped terraces

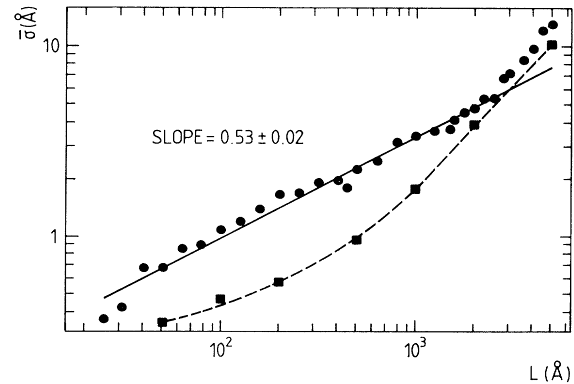

FIG. 3. Average root-mean-square roughness vs scan size for the iron film sample after annealing (squares) and after ion bombardment (circles). The solid line depicts a least-squares fit to the linear portion of the data. Each point of this data set represents an average of 5–15 scans recorded at random locations. Progressively more scans must be averaged as the scan size diminishes, in order to hold the error for all of the points to below 10%. That more scans must be averaged at the smaller sizes reflects the fact that local slope variations are gaining importance with respect to the macroscopic tilt of the sample.

(Fig. 4), with a typical terrace area on the order of $(5\text{–}10)\times 10^4$ Å^2. There is no linear region in the log-log plot of σ vs L, so the annealed sample cannot be considered as self-affine in nature.

Ion bombardment of the sample consisted of 5-keV Ar^+ ions striking the entire sample surface (area $=0.25$ cm^2) at an angle of 25° with respect to the surface normal, with a flux $\approx 5\times 10^{14}$ ions/cm^{2}s. Numerous measurements were carried out, varying the fluence between 10^{17} and 10^{18} ions/cm^2. We observed no effect of fluence on the value of H. A typical scan image for the eroded surface is depicted in Fig. 4, where the line of direction of the ion impact is somewhat in evidence. The upper data set in Fig. 3 is typical of the sample after being subjected

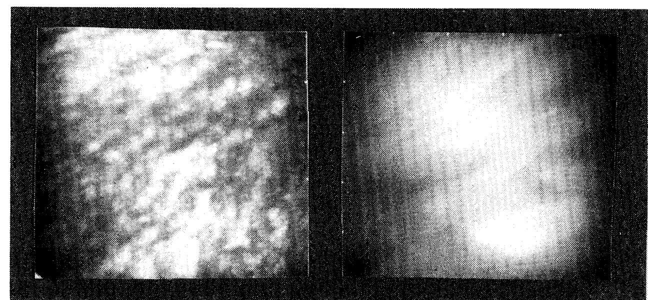

FIG. 4. STM images of the iron film sample after annealing (right) and after Ar^+-ion bombardment (left). The bombardment direction is from the lower left-hand to the upper right-hand side of the latter image. Each image is 2000 Å in edge length. The vertical scale (black to white) is 23 Å.

to ion bombardment. For scan sizes below 2000 Å, the data fall on a straight line with slope $H = 0.53 \pm 0.02$. The straight line fit to the data set intersects with the curve for the annealed surface at $L \approx 2000$ Å. Above this point the power-law scaling is evidently obscured by the preexisting surface roughness.

Our results suggest that the applicability of scaling theory to describe erosion at submicron length scales may extend far beyond the very different previous case study of ion-beam erosion of a graphite surface [13]. In that case, a single-crystal graphite surface was ion bombarded in vacuum and then examined in air. The scaling parameter which we have measured, $H = 0.53 \pm 0.02$, is higher than that of the graphite sample, $H \simeq 0.2$–0.4, but is consistent with $H = 0.5 \pm 0.1$ reported for vapor-deposited silver films [20] and is in reasonable agreement with the value $H \approx 0.4$, implicit in fits reported for the iron film growth data [14]. We know of no scaling theory which explicitly predicts a scaling exponent whose value is $H = 0.5$. This value does, however, fall between the exponents 0.66 and ≈ 0.35, expected [21–23] for deposition processes which, respectively, do or do not allow for surface mobility of particles after striking the surface. We conclude, therefore, that some rearrangement of the iron film surface may be occurring at the atomic level as particles are removed from the surface.

In conclusion, we have described a straightforward method for obtaining a precise value for the scaling exponent H of a self-affine fractal surface, via scanning tunneling microscopy, and have employed this method to study ion-induced erosion of an iron film sample. Our results provide strong support for the applicability of scaling theory to ion-induced surface erosion at submicron levels. A value for the scaling exponent has been obtained, both *in situ* and in well-documented experimental conditions. Further experimental and theoretical studies will be necessary in order to gain an understanding of the precise physical origin of the value $H = 0.53 \pm 0.02$ which has been observed.

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[1] M. Sikkens, I. J. Hodgkinson, F. Horowitz, H. A. Macleod, and J. Wharton, Opt. Eng. **25**, 142 (1986).

[2] For recent reviews, see P. Meakin, Prog. Solid State Chem. **20**, 135 (1990); F. Family, Physica (Amsterdam) **168A**, 561 (1990); J. Krug and H. Spohn, in *Solids Far from Equilibrium: Growth Morphology and Defects*, edited by C. Godriche (Cambridge Univ. Press, Cambridge, 1990).

[3] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1989).

[4] B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, New York, 1982).

[5] G. S. Bales, R. Bruinsma, E. A. Eklund, R. P. Karunasiri, J. Rudnick, and A. Zangwill, Science **249**, 264 (1990).

[6] R. Bruinsma (unpublished).

[7] F. Family and T. Vicsek, J. Appl. Phys. A **18**, L75 (1985).

[8] *Erosion and Growth of Solids Stimulated by Atoms and Ion Beams*, edited by G. Kiriakidis, G. Carter, and J. L. Whitton (Martinus Nijoff, Dordrecht, 1986).

[9] S. Hoffmann, in *Practical Surface Analysis*, edited by D. Briggs and M. P. Seah (Wiley, New York, 1990), 2nd ed.

[10] M. P. Seah and C. Lea, Thin Solid Films **81**, 257 (1981); A. Zalar and S. Hoffmann, Vacuum **37**, 169 (1987).

[11] C. A. Lang, C. F. Quate, and J. Nogami, Appl. Phys. Lett. **59**, 1696 (1991).

[12] T. Michely, K. H. Besocke, and G. Compsa, Surf. Sci. Lett. **230**, L135 (1990).

[13] E. A. Eklund, R. Bruinsma, J. Rudnick, and R. S. Williams, Phys. Rev. Lett. **67**, 1759 (1991).

[14] J. Chevrier, V. Le Thanh, R. Buys, and J. Derrien, Europhys. Lett. **16**, 737 (1991).

[15] M. W. Mitchell and D. A. Bonnell, J. Mater. Res. **5**, 2244 (1990). The fractal dimension in this publication is written as $D = 2 - H$, rather than $D = 3 - H$, since profiles embedded in two (rather than three) spatial dimensions are analyzed.

[16] B. Mandelbrot, D. Passoja, and A. Paullay, Nature (London) **308**, 721 (1986).

[17] Sycon Instruments Inc., East Syracuse, NY, (315)463-5297; 6-MHz quartz-crystal unit for deposition rate monitors.

[18] P. Pfeifer, Y. J. Wu, M. W. Cole, and J. Krim, Phys. Rev. Lett. **62**, 1997 (1989).

[19] V. Panella and J. Krim (unpublished).

[20] R. Chiarello, V. Panella, J. Krim, and C. Thompson, Phys. Rev. Lett. **67**, 3408 (1991).

[21] M. Kardar, G. Parisi, and Y. Zhang, Phys. Rev. Lett. **56**, 889 (1986).

[22] D. E. Wolf and J. Villain, Europhys. Lett. **13**, 389 (1990).

[23] Z. W. Lai and S. Das Sarma, Phys. Rev. Lett. **66**, 2348 (1991).

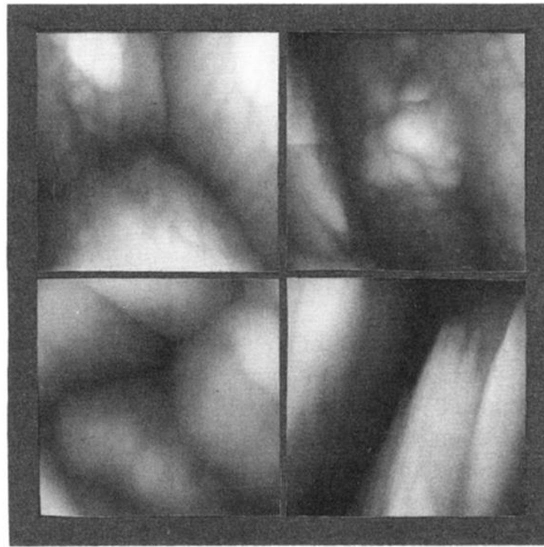
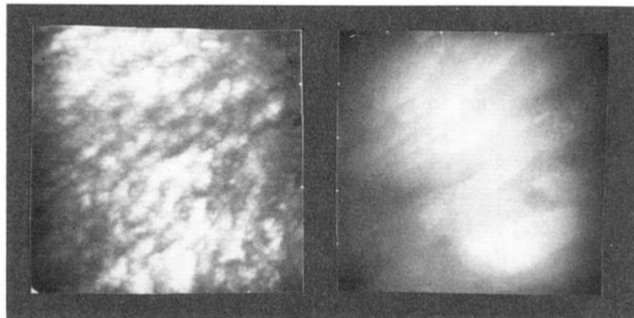


FIG. 1. Top view of STM images recorded on the gold film test sample. The scan sizes, clockwise, starting in the upper left corner are $(2000\ \text{Å})^2$, $(5000\ \text{Å})^2$, $(20\,000\ \text{Å})^2$, and $(500\ \text{Å})^2$. The corresponding vertical scales (black to white) are 1000 Å, 2000 Å, 4000 Å, and 200 Å.



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