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Simple model for etching

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A simple numerical model for the etching of materials is described. Atoms along the surface are removed randomly, with probabilities weighted by both the underlying crystal structure and by the bond strength of atoms to their nearest neighbors. Particular attention is paid to the resulting surface morphology and its possible relationship to a fractal dimensionality. This study is relevant to fundamental questions surrounding the mechanisms of etching, as well as to technologically important issues that arise as the size of etched features extend below 1000 Å.

INTRODUCTION

The shaping of material by chemical etching plays an important role in the fabrication of structures used to study fundamental physics, as well as an important role in technological applications. As long as the structures to be etched are much larger than the atomic scale (usually 10^5–10^7 times larger), simple differential equation models can be used to describe the resulting macroscopic surface.[1–4] However, recent advances in the technology of defining small etched shapes (e.g., by electron beam or x-ray lithography), has permitted the fabrication of structures with 1000 Å or narrower features. These structures contain only 300 atoms along a side. Under such conditions, the actual shape formed during etching will depend, in a critical way upon the details of the *etching chemistry,* as well as on *statistical fluctuations* in the etch rate along the surface. In this regime, many new questions now arise. For example, how flat can a surface be made? Can a shape with a square cross section be maintained during etching? Thus the first aim of this model is to address questions of surface shape and morphology, and in particular, to find a quantitative measure of the surface roughness.

A second aim is to make contact with the chemistry of etching. This is an enormously fertile field, partly due to the complexity of the chemistry and partly due to the difficulty of directly probing the relevant reactions. Because each etch behaves in a unique way on each material (e.g., HF attacks SiO_2, but not Si) a general theory for the chemistry may be unobtainable. Instead, our model concentrates on enumerating the various probabilities for removing a single atom from the surface. As discussed below, these probabilities can be related to temperature and activation energies of certain chemical reactions, although all we intend to provide here is a complete, though abstract, description. In fact, it may be possible to invert the usual process, and start by comparing the computer generated etched surfaces to real surfaces. This allows one to define the range of relative bond probabilities, and thus to limit the range and types of chemical reactions. Details of this approach are found in the Discussion.

The final purpose of this model is to develop a technique capable of describing phenomena that range over vastly different length and time scales. For example, an attempt rate in removing an atom might be determined by a phonon frequency ($\sim 10^{13}$ sec^{-1}), whereas an arrival rate for etch molecules at the surface might be only 10^3 sec^{-1}. This is a range of 10^{10}, a range much too large for conventional techniques, such as molecular dynamics, to span. However, as we shall show these disparate time scales can easily be reconciled within our model.

MODEL

The crystal to be etched consists of a network of "atoms" arranged in a periodic lattice (all of the examples here are for cubic lattices, although more general types are nearly as easy to handle). Only those atoms along the surface are removed by the etch, and only one atom is removed per etch molecule. The algorithm for removing an atom is simple and straightforward: A surface atom is chosen at random and its number of nearest neighbors is calculated. This number is then appropriately weighted to reflect the strength of the individual bonds. Bond strengths, in turn, are determined by a number of factors. First, the strength of the bond might depend on temperature. For example, there might be an activation energy for breaking a bond. Second, the bond strength might depend on which atom is missing. This is a "lock and key" effect, and reflects the fact that some etch molecules have geometrical shapes which are particularly well suited to remove atoms with a selected orientation. Third, the bond strength might depend on history. For example, the etch might easily remove one bond, only to have that surface atom oxidized (and perhaps passivated) by another component of the etch. Thus it is possible to break the first bond more easily than the second. Rather than treating any of these or other mechanisms in detail, we instead enumerate all possible bond configurations and assign a unique probability (ultimately based on the underlying chemistry) of removing that atom given the arrangement of its neighbors. If this number is greater than a random chosen between 0 and 1, the atom is removed.

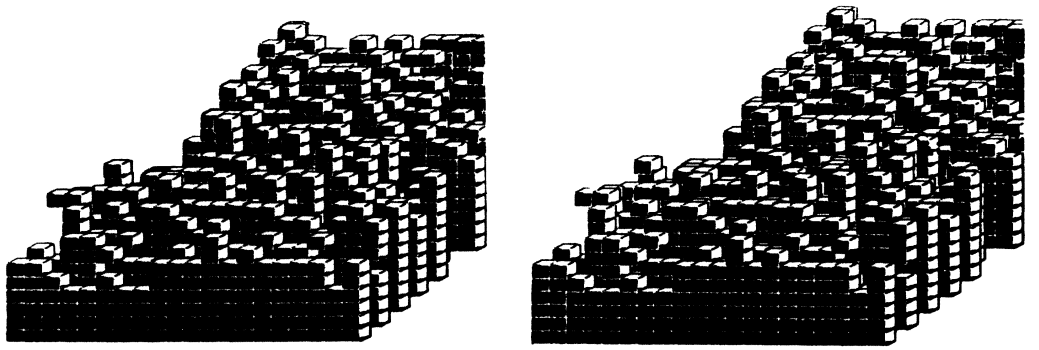
As will be seen in the following sections, this algorithm is sufficiently general to encompass isotropic and anisotropic etches, as well as the etching of alloy systems. With a small change it can also treat the effect of dif-

fusion limiting etch rates. An example of isotropic etching is given in the stereo views of Fig. 1. These two figures arise through two different conditions on the bond strengths; in Fig. 1(a) it is sufficient to break one bond (that is, one missing nearest neighbor) to remove an atom, in Fig. 1(b) two or more nearest neighbors must first be removed. It is immediately apparent that the more bonds which must be broken before an atom is removed, the smoother the resulting surface. Unfortunately, the eye and brain are otherwise poorly adapted to understand random, three-dimensional (3D) images, and much additional qualitative information about the etched surface is obscured. It is for this reason that all of the examples, other than Fig. 1 will deal with etching along two dimensions, rather than 3D surfaces. Roughly speaking, the 2D case is similar to viewing a single plane cut perpendicularly to a 3D etched surface. There is, however, one qualitative difference. In two dimensions, atoms with broken bonds lie entirely along the surface. A 2D slice through a 3D surface, on the other hand, will occasionally have an atom missing in the interior of the crystal. These holes occur because adjacent planes of the crystal may be lower, and thus provide access for the etch to the interior of the imaged plane. No analogous mechanism exists in two dimensions.

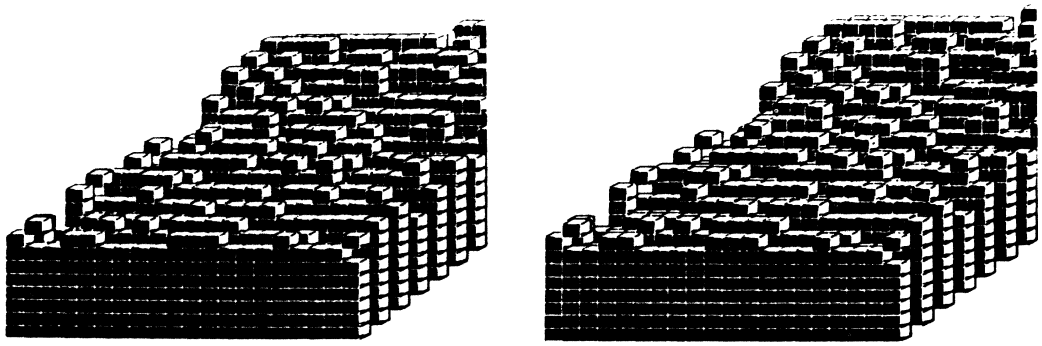
Although there is a fundamental difference between the two dimensions, the features of a 2D surface are sufficiently self-evident to warrant their use in the rest of this work. A detailed comparison with real etched surfaces, however, requires the use of the full 3D simulation. Fortunately, restricting etching to nearest-neighbor interactions means that the 3D case is as simple to implement as the 2D case, although the computation time does increase proportionately.

ISOTROPIC ETCHING

In isotropic etching, each bond has the same strength. For a square lattice, there are fourteen distinct ways to arrange the nearest neighbors along the surface. Adopting the notation U (up, into the crystal and away from the etch), D (down, towards the etch), L (left), R (right) (as in Fig. 2), these are U, D, L, R, UD, DL, LR, UR, LU, RD, UDL, LDR, UDR, LUR. For an anisotropic etch, each of these combinations might present a different specificity to the etch, and thus might have a different probability



(a)



(b)

FIG. 1. Stereo view of a three-dimensional etched surface. Each cube represents a single atom. (a) A strong etch. There is a high probability that a randomly selected surface atom will be removed on the first attempt. (b) A weaker etch. A surface atom is easily removed only if its nearest neighbors are already missing.

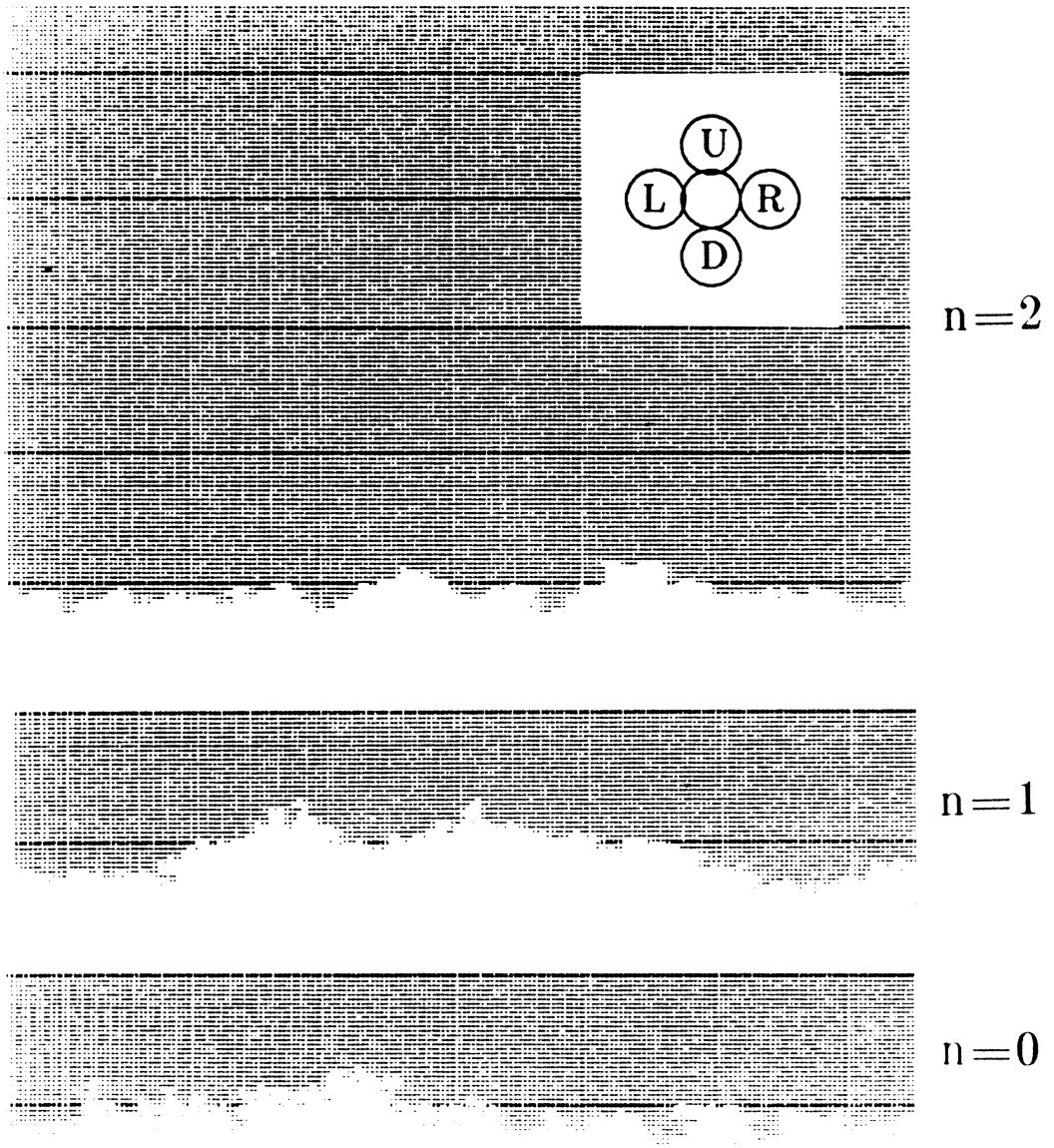


FIG. 2. Etching on a two-dimensional surface, for various values of n. Each point is an atom, so the surface is 200 atoms (approximately 600 Å) wide. The inset illustrates the bond labeling convention. Note the stronger etch ($n=0$) results in a rougher surface than the weaker etch ($n=2$).

for removal. In the isotropic case, however, the probability depends only on the number of bonds, and not on their arrangement. A strong isotropic etch might remove a surface atom on the first attempt (independent of the number of bonds), whereas a weak etch might find it easy to remove an atom only if three out of four bonds were broken. Here we define, for purposes of illustration, the normalized bond strength as $b=(p/4)^n$, where p is the number of broken bonds. This simple numerical expression varies the relative bond strengths with a single parameter n. $n=0$ corresponds to a strong, bond independent etch, while $n=1$ or 2 denotes progressively weaker etches. The effect on the surface can be quite clearly seen in Fig. 2, where there is a general rough to smooth transition. (A similar transition is observed in simple models of crystal growth[5].)

Given these three different surface morphologies, the

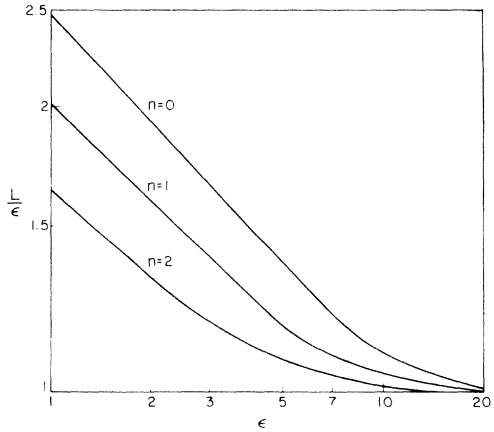


FIG. 3. $L(\epsilon)/\epsilon$ versus ϵ, for various values of n. The slope of the curve is the fractal dimensionality minus 1.

question of classifying the nature of the surface naturally arises. A good classification scheme is important for a number of reasons. First, a classification scheme is valuable in abstracting from the numerical model qualitative features which might otherwise remain hidden. Second, if analytical models for etching become possible, their predictions could easily be compared with these same abstracted quantities. Lastly, if it were possible to describe the surface with a function of only one or two parameters, then one might be able to correlate the observable surface with its underlying physical parameters.

Conventional choices include the number distribution of heights perpendicular to the surface,[6] or the one-dimensional correlation function,[6] or the one-dimensional spatial Fourier transform[7] along the surface. While these methods have proven useful in extracting the rms roughness from experimental measurements, they do not capture the full richness of the surface morphology. In particular, they do not allow the height to be double valued (i.e., no "overhangs"), and they require a large number of parameters to describe each curve. What is needed is a minimal, though complete generating function for the surface.

An obvious choice, given the resemblance of this convoluted surface to a map of the shoreline, is a fractal. After Mandelbrot,[8] we look for a function of the form $L(\epsilon)=\epsilon^d$, where L is the perimeter measured with a "ruler" of length ϵ, and d is the dimensionality. Figure 3 gives the result for $n=0,1,2$. A pure fractal, which is self-similar on all length scales, would yield a straight line on this plot. We find the fractal dimensionality (for low ϵ), is $\frac{4}{3}$ for all values of n, although an n-dependent cutoff of ~ 10 can clearly be seen. To some extent, a fractal description is sufficient, providing this n-dependent cutoff on ϵ is used. The cutoff arises from a simple, physical mechanism. If a fluctuation produces a peninsula shaped region along the surface that is much longer than it is wide, the etch (which is otherwise isotropic) is likely to cut across the peninsula somewhere along its length. Thus, it is not possible to have self-similar roughness on all length scales. However, a fractal with a cutoff does give a moderately complete description of the surface.

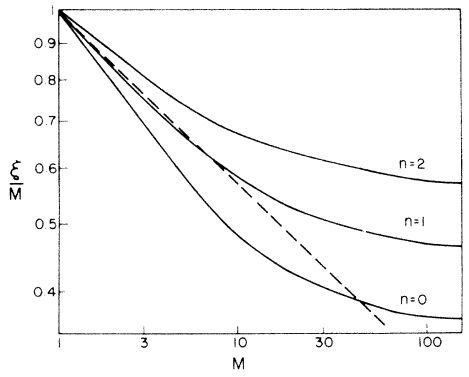


FIG. 4. $\xi(M)/M$ versus M, for various values of n. For small M the slope is related to a self-avoiding random walk, for large M, ξ/M is the ratio of the lattice parameter to the rms surface roughness. The dotted line is the result for a self-avoiding random walk.

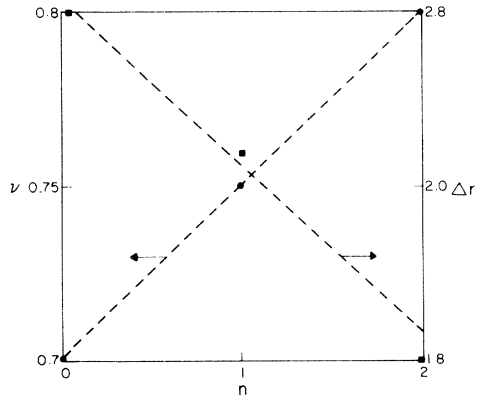


FIG. 5. ν (■) and Δr (●) (measured in units of the lattice parameter), as a function of n. The lines are guides for the eye.

A second choice is motivated by the shape of the perimeter roughly corresponding to that generated by a particle undergoing a random walk on a grid. If we denote ξ as the distance between two atoms on the surface, and M as the actual length of the convoluted perimeter between the two atoms, then one expects[9,10] for a self-avoiding random walk, $\xi \sim M^{\nu}$, where $\nu=\frac{4}{3}$ This quantity (normalized to M) is plotted in Fig. 4 as a function of n. The dotted line is the prediction for a self-avoiding walk in two dimensions.

For large M a constant value of ξ/M is obtained, again due to the fact that the etched surface never departs too far from a flat surface. It can be shown that ξ/M is approximately the inverse of the rms roughness, measured in units of the lattice spacing. (That is, at large M, ξ de-

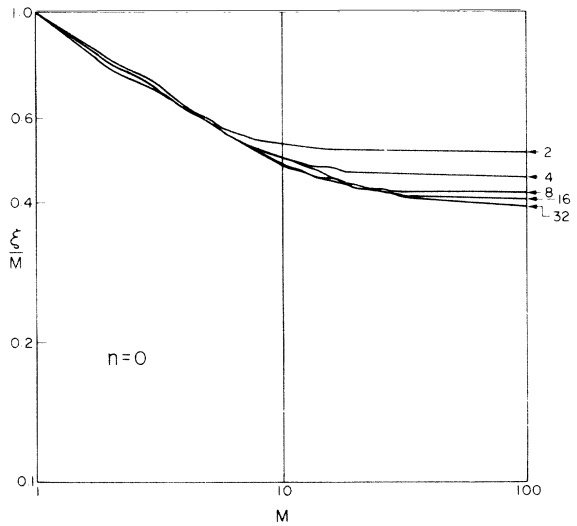


FIG. 6. ξ/M as a function of the distance the etch has penetrated from the surface, for the case $n=0$. By the time ~ 32 monolayers have been removed, the surface is in equilibrium with the etch.

scribes a straight line along the level of the mean surface, with the actual perimeter M varying many times about the mean. $\Delta = M/\xi$ is a measure of the roughness, up to a coefficient of order 1. We avoid using the actual rms roughness since it is appropriate only for a surface with a Gaussian distribution of heights.) Thus, a relatively concise description of the surface can be given by two accessible physical parameters. The first is the slope of ξ for small M, which measures how far the perimeter deviates from a random walk. The second is the rms roughness Δr. With this simplified description of the surface, it is then possible to correlate the underlying bond energies with physically observable parameters. Figure 5 shows the slope $d\xi/dM$, and Δr versus n. Measurement of these two quantities can then be used to determine n.

In the previous analysis, the curves were generated by taking the appropriate averages only after waiting until 500 layers of atoms were etched from the surface. This precaution assures that the etching has reached equilibrium, independent of the initial surface roughness.[11] However, many etched features may only be 500 atoms high, and thus the question of surface roughness as a function of distance from the surface may be of critical importance. Figure 6 plots ξ/M versus M for the case $n=0$. An increased roughening occurs in from the surface as each layer is removed. Equilibrium, starting with a perfectly flat surface, evolves after ~ 32 layers for this strength etch.

DIFFUSION EFFECT

So far, this model assumed there was an adequate supply of etching molecules at the surface, and that geometri-

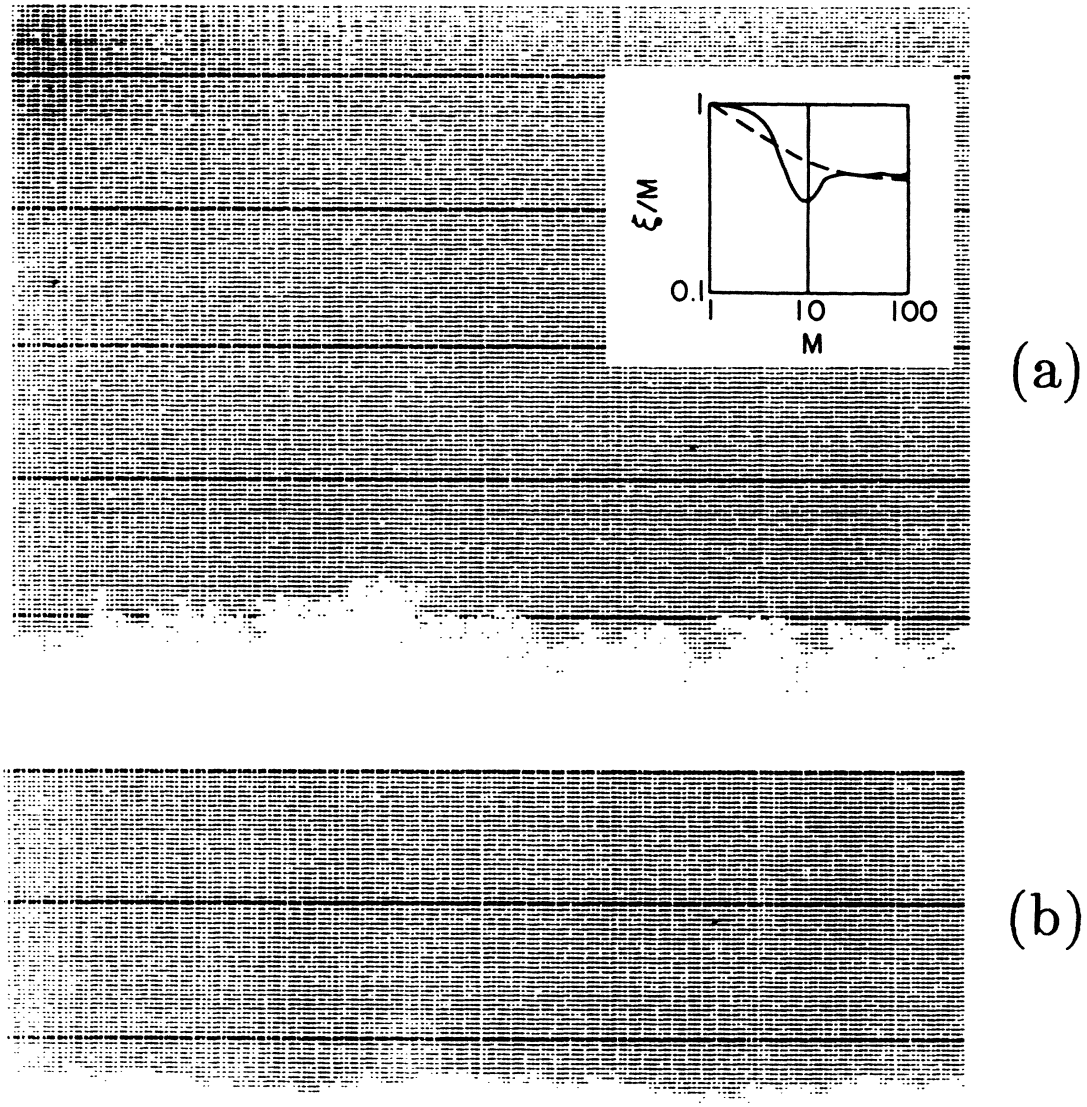


FIG. 7. Two etched surfaces. (a) This is the case with $n=0$, the same isotropic etch illustrated in Fig. 2. (b) This figure includes the effect of the diffusion algorithm ($n=0, \lambda$ of 10), which has produced a smoother surface. The inset compares the smoothing (solid line) effect on ξ to the case without diffusion (dotted line). Note the strong effect around $M=10$.

cal effects (such as shadowing in reactive-ion etching) or etch depletion (due to the blocking effect of etch by-products) are unimportant. Reactive-ion etching, with its complex interplay of chemistry and momentum transfer, is well beyond the scope of this paper, but geometrical effects and diffusion can easily be handled. Diffusion and the subsequent formation of a depletion region are expected to play a role in wet (chemical) and dry (plasma) etching.

Diffusion here is treated in a simple, physically intuitive manner at the atomic level. Two mechanisms can block the etch from reaching the surface. First, during the course of etching some atoms are removed in clumps. These originally were peninsula shaped regions which were then nipped off at the base. Some of these atoms may be seen along the surface in Fig. 2. Clumps can locally reduce the surface etch rate since an etch molecule might attack here, rather than at the surface. A second effect is geometrical, involving the flushing of etch by-products from within the grooves. The narrower the groove, the harder it is for an etch by-product to leave, and for a new etch molecule to enter.

To include these effects which block etch molecules from the surface, we first randomly chose an atom along the surface or along one of the clumps. A "circle" is then drawn around this point and the ratio of empty space to unetched atoms (τ) is calculated. For an atom deep in a crack, τ is near zero, for an atom on the surface $\tau \sim \frac{1}{2}$, and for an atom on the tip of a peninsula, $\tau \sim 1$. The probability of removing the atom then depends on the

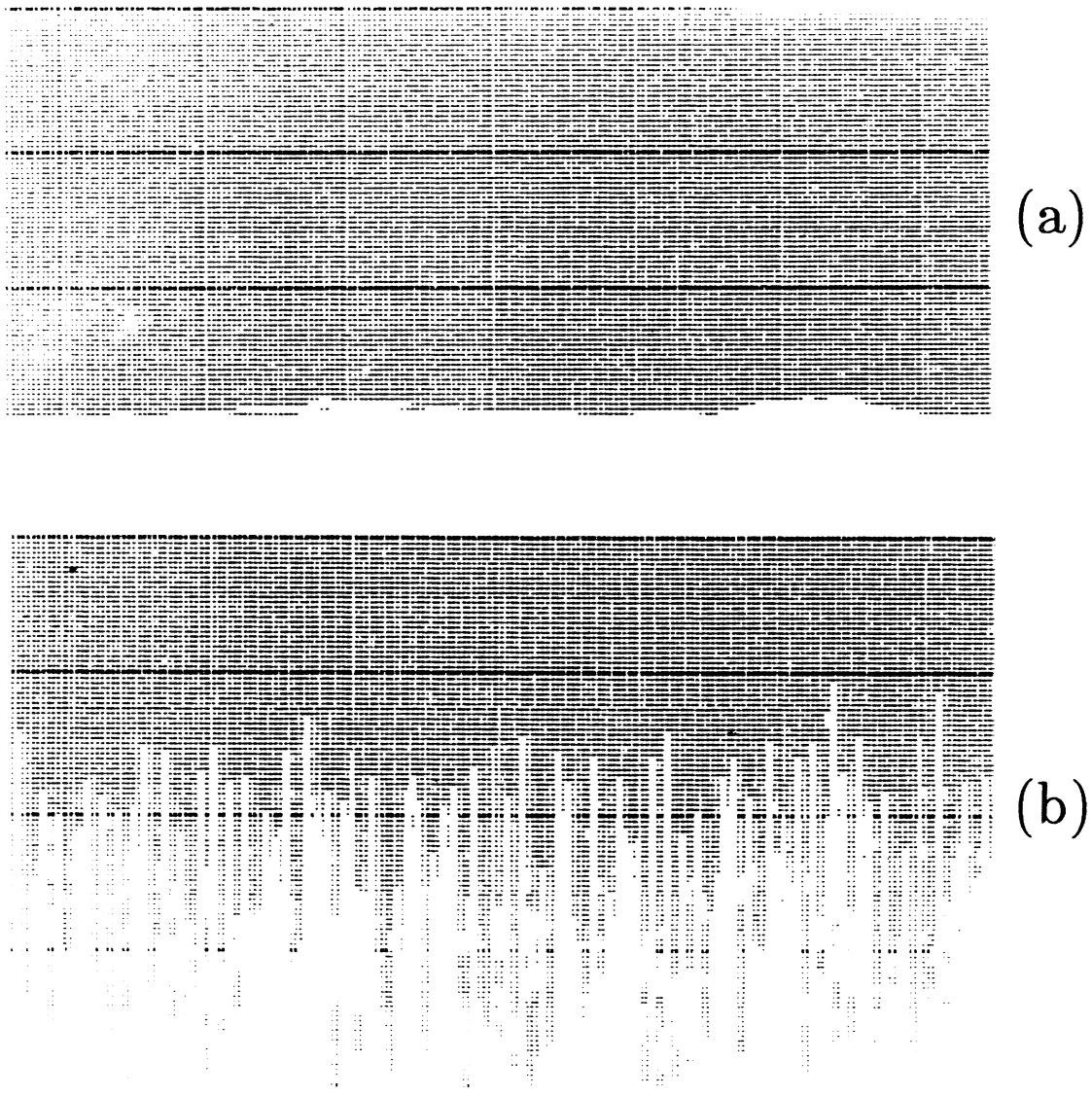


FIG. 8. Anisotropic etching: $p(\mathrm{U})=p(\mathrm{D})=0.001$, $p(\mathrm{L})=p(\mathrm{R})=0.4$, $p(\mathrm{UD})=0.03$, $p(\mathrm{LR})=p(\mathrm{UD})=p(\mathrm{LD})=p(\mathrm{DR})=p(\mathrm{UR})=p(\mathrm{LU})=0.1$, $p(\mathrm{UDL})=p(\mathrm{UDR})=p(\mathrm{DLR})=p(\mathrm{LUR})=0.2$. (a) As can be easily seen, etching is slow in this direction, and is referred to as a "stop" plane. (b) Same crystal, with the surface rotated at 90° compared to (a). The rough surface is characteristic of a "fast" etching plane.

bond probabilities as well as monotonically increasing with τ. The radius of the circle λ plays the role of a diffusion length. As seen in Fig. 7, the effect of a diffusion length is to smooth the surface by removing those spatial frequencies near λ. Specifically, atoms well within one diffusion length all have about the same τ, and so their surface morphology is unchanged from the case without diffusion. Fluctuations on a length scale longer than λ are essentially constant backgrounds in any one averaging circle, and so again do not play a role. However, features with a length scale of λ yield very different values of τ for atoms in the circle, and thus these frequencies are damped out (a particularly strong damping is seen in the anisotropic case, Fig. 9). Physically, a small λ corresponds to an etch where diffusion dominates, or where a small amount of etch by-product can almost completely halt the etching. If a smooth surface is desirable, then a nearly exhausted (i.e., "fully loaded") etch is preferred, although it cannot smooth out all surface roughness.

ANISOTROPIC ETCHING

In anisotropic etching, certain planes of the crystal etch more rapidly than others, an effect which can be used to produce a number of electronic[12,13] devices as well as structures for basic physical measurements.[14] In our model, anisotropy can be produced by appropriate weighting of the bond energies. Figure 8 shows two choices, with the values of U, D, etc. given by the caption. The smooth, almost flat surface of Fig. 8(a) is referred to as a "stop" plane for the etch. Fig. 8(b) is the same crystal, turned 90° to the etch. Etching proceeds quite rapidly down the "fast" plane.

Diffusion has a marked effect on etching along the fast plane. In Fig. 9 the bond probabilities are chosen to produce extreme anisotropy. A diffusion length of 10 atoms, however, effectively suppresses all structure above this length. Another example of an anisotropic etch can be found below.

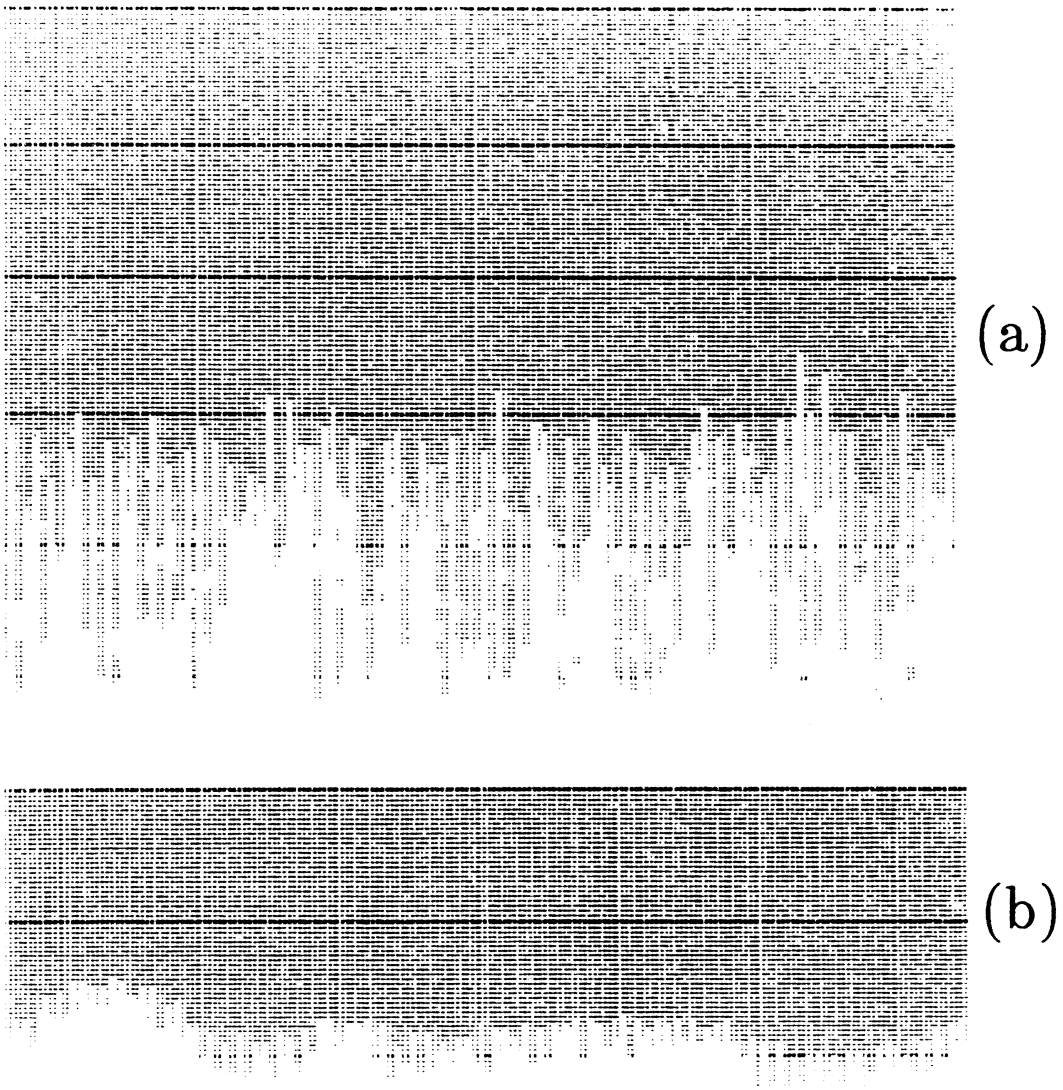


FIG. 9. (a) A very anisotropic etch [same as Fig. 8, with $p(\mathrm{L})=p(\mathrm{R})=0.8$], and (b) the effect of a diffusion length of a diffusion length of $\lambda=10$.

MULTICOMPONENT CRYSTALS

Many interesting materials, such as GaAs, are binary alloys with alternating planes of atom A and B. It is quite possible for an etch to attack A much more strongly than B, thus leading to a characteristic surface shape. Figure 10 exhibits this effect, which for this crystal orientation results in microfaceting. It is also a relatively simple matter to "numerically etch" a crystal with a modulated lattice, or to examine the effect of dislocations on etch rate and anisotropy. Indeed, it is not necessary to begin with a crystal lattice. A glass-like structure with bond energies which depend on bond angles, can, with the proper indexing, be just as easy to handle.

SHAPE ETCHING

Perhaps the most common application for etching, other than in surface cleaning, is in microfabrication; i.e., the etching of small shapes. The smaller the shape, the more important it becomes to control the feature's final dimensions. For example, if a small square protruding above a plane is placed in an etch, will the shape round? How much thinning can the stub take before etching away?

Figure 11 illustrates this case, with an isotropic etch ($n=1$). External angles round up to the rms roughness for that etch and then stop. Internal angles broaden as new material is exposed, widening the base of the stub. The sides of the square, away from the corners, remain basically flat and parallel.

Diffusion has the effect of rounding the corners, since an atom on the corner has a $\tau \sim \frac{3}{4}$, compared to $\frac{1}{2}$ for the flat, or $\frac{1}{4}$ for an interior angle. In other words, corner atoms are exposed to a larger volume of etch, and thus etch faster. This rounding is limited to a radius of $\sim\lambda$.

In Fig. 12 a square object is exposed to an etch on all four sides. The four faces of the square are stop planes (in the familiar case of silicon, the square can be thought of as a slice through a 3D crystal, with the paper perpendicular to the $\langle 110 \rangle$, and the faces of the cube slightly skewed $\langle 111 \rangle$ planes).

In the isotropic case the surface first roughens and then the square shrinks, maintaining its shape and orientation until it has nearly disappeared. In the anisotropic case, the stop planes are hardly attacked. The etch moves most rapidly at the corners, where other planes are exposed. Eventually, all the etching occurs from the corner along the fast direction, 45°, off the original faces. The shape eventually becomes a cube rotated at 45°, with its faces exhibiting facets characteristic of the slower etching planes.

DISCUSSION

The etching algorithm described in this paper uses a statistical, rather than a dynamical approach. An individual atom is not followed along some trajectory through time, as it might be in diffusion limited aggregation[15] or in some models of crystal growth.[16] No particular dynamical laws are invoked to control the motion of the particles; rather the statistics are presumed to contain all

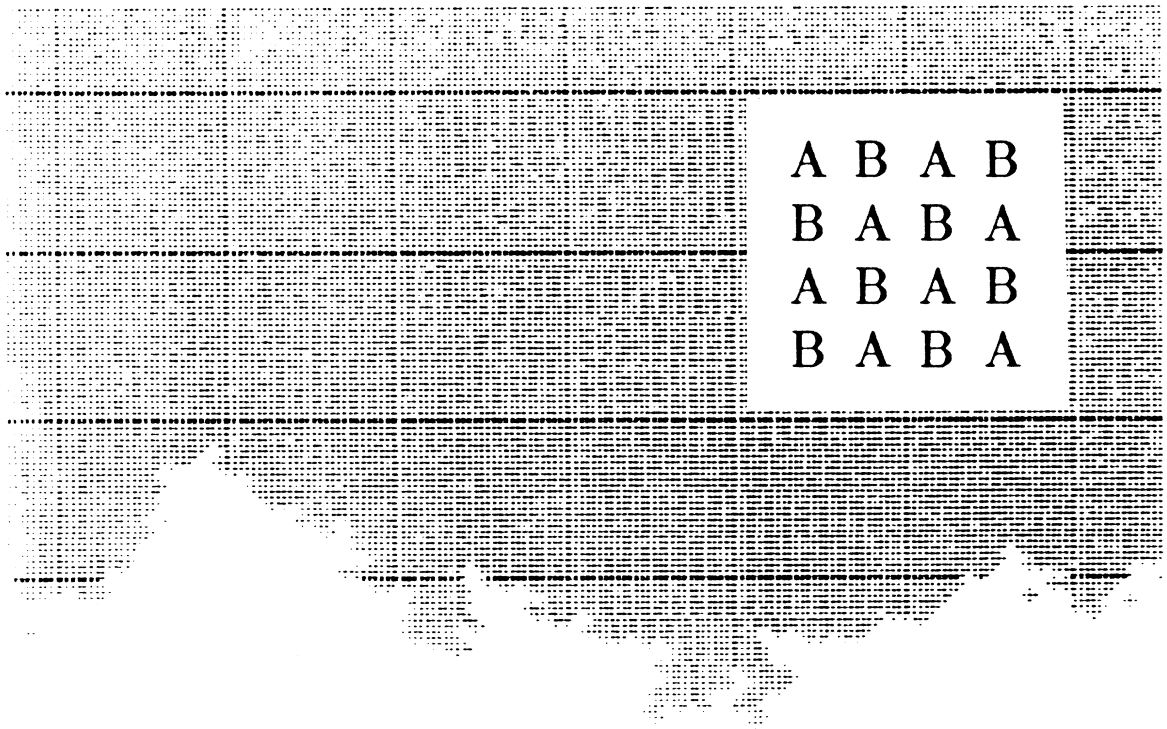


FIG. 10. Alloy system. Orientation of A-B crystal is shown in the inset, and the bonds have etching probabilities given by $n=1$, and $p(\mathrm{A})=0.01p(\mathrm{B})$.

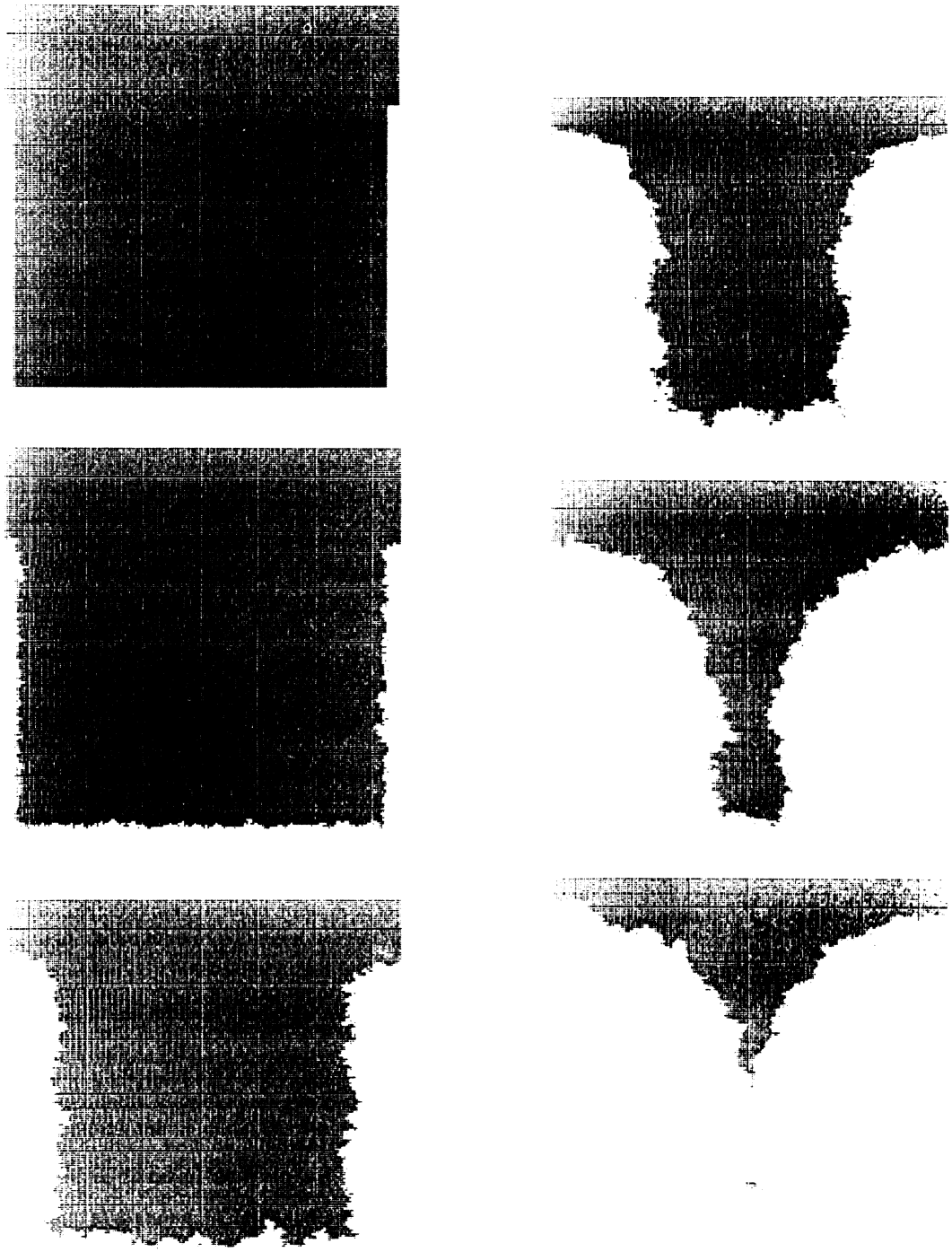


FIG. 11. Etching of a square stub attached to a plane, as a function of time. This is an isotropic etch with $n=1$. The stub is 200 atoms wide.

the information. There are two reasons for adopting this singular approach. The first is simplicity, from both a computational and from a pedagogical standpoint. The second reason is to place the emphasis on *surface* and *statistical* effects, both of which may be expected to control the etching behavior of a small number of atoms when the surface to volume ratio is high. This is very much like trying to determine the ideal gas law by examining the behavior of particles in a box. On the one hand, we can follow a particle in the bulk of the gas as it experiences many scattering events, then follow the scattering off the wall, and then calculate the momentum transfer, or we can examine only those particles about to hit the wall, weighting their numbers and velocities with appropriate statistics. The first approach is most general, but the second approach may be just as accurate and could supply

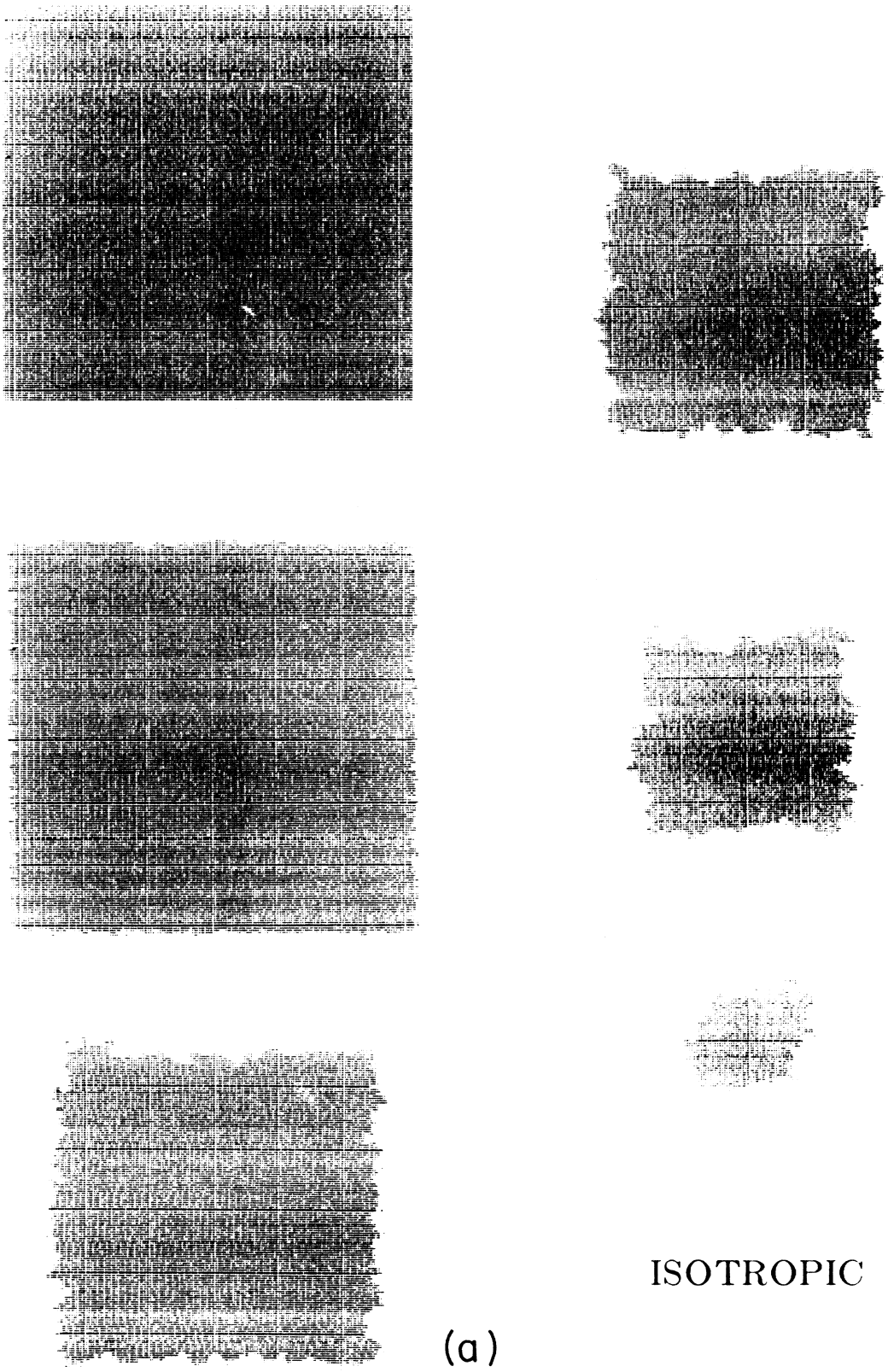


FIG. 12. A square surrounded on all four sides by etch. (a) Isotropic etch ($n=1$) and (b) anisotropic etch with $p(\mathrm{U})=p(\mathrm{D})=p(\mathrm{R})=p(\mathrm{L})=0.001$, $p(\mathrm{LR})=p(\mathrm{UD})=0.03$, $p(\mathrm{LD})=p(\mathrm{DR})=p(\mathrm{UR})=p(\mathrm{LU})=0.2$, $p(\mathrm{UDL})=p(\mathrm{UDR})=p(\mathrm{DLR})=p(\mathrm{LUR})=0.66$. The crystal is oriented such that the faces are stop planes.

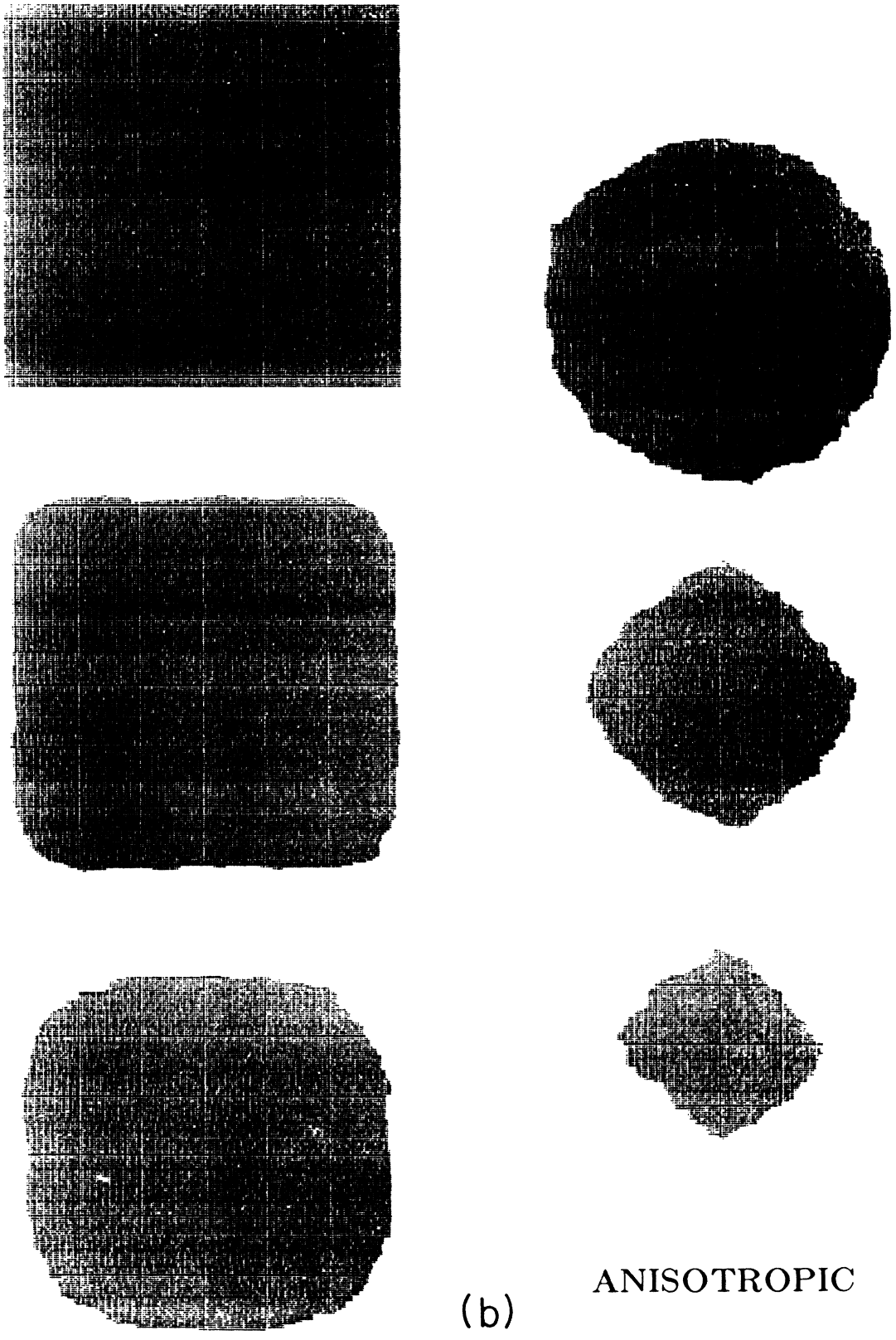


FIG. 12. (*Continued*).

a more transparent explanation.

This model makes a number of specific, qualitative predictions. These include the preservation of exterior angles during etching, the rounding of interior angles, and the dependence of surface roughness on etch strength. In addition, anisotropic etching of crystalline materials and isotropic etching of binary alloys are shown to exhibit characteristic shapes.

It is perhaps now feasible with present technology to check these predictions. Surface probes, such as transmission electron microscopy or scanning tunneling microscopy can image individual atoms, albeit only on specially prepared surfaces. These techniques will someday allow a direct measure of surface morphology. At slightly lower resolution (~ 100Å) it should be possible to check these predictions with more conventional imaging techniques, such as scattering electron microscopy or adsorbed gas measurements. In either case, the ultimate purpose is to make contact with the underlying chemistry.

A first step along these lines is to use the etching algorithm as a broad filter with which to examine possible etching mechanisms. For example, if anisotropic bond lengths are thought to be the sole origin of anisotropic etching, one can use them to predict bond probabilities, and thus the surface shape. If the shape does not agree with experiment, then a bond length mechanism can be ruled out. Since to some extent shape etching acts as an "amplifier" of the etching process, that is, it produces a larger than atomic scale effect from atomic scale processes, this model is most easily checked by comparison with shape etching. Shape etching includes lithographically patterned structures, as well as naturally occurring etch pits. We are presently extending our model to larger three-dimensional surfaces, in order to begin this comparative process.

Independent of this model's ability to describe etching phenomena is the strength of its underlying "statistical" framework. This framework can be checked and enhanced in a number of ways. First, molecular dynamics simulations, where applicable, can be used to put limits on the range of bond probabilities, and to check to see if a statistical description deviates from dynamically based models. Second, the statistical approach can be tried in an entirely different model system, in order to test its generality. For example, we have used a similar model to describe vapor phase crystal growth. The predictions of this new model are now being tested, and will be reported on in future publications.

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[11]Real etched surfaces take longer to reach equilibrium than the model would indicate. Polished surfaces, no matter how smooth, contain hidden disorder from the polishing grit which is exposed preferentially by the etch. Surfaces produced by cleaving, or by alternate anodizing-oxide etch cycles, would be closer to the ideal.

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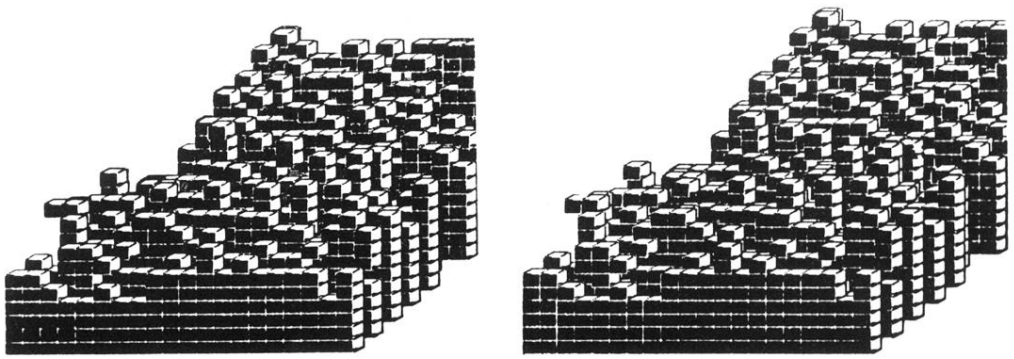
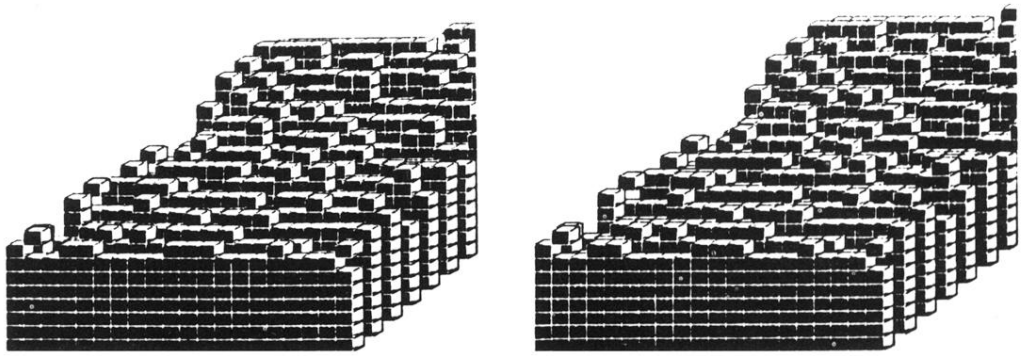


(a)



(b)

FIG. 1. Stereo view of a three-dimensional etched surface. Each cube represents a single atom. (a) A strong etch. There is a high probability that a randomly selected surface atom will be removed on the first attempt. (b) A weaker etch. A surface atom is easily removed only if its nearest neighbors are already missing.

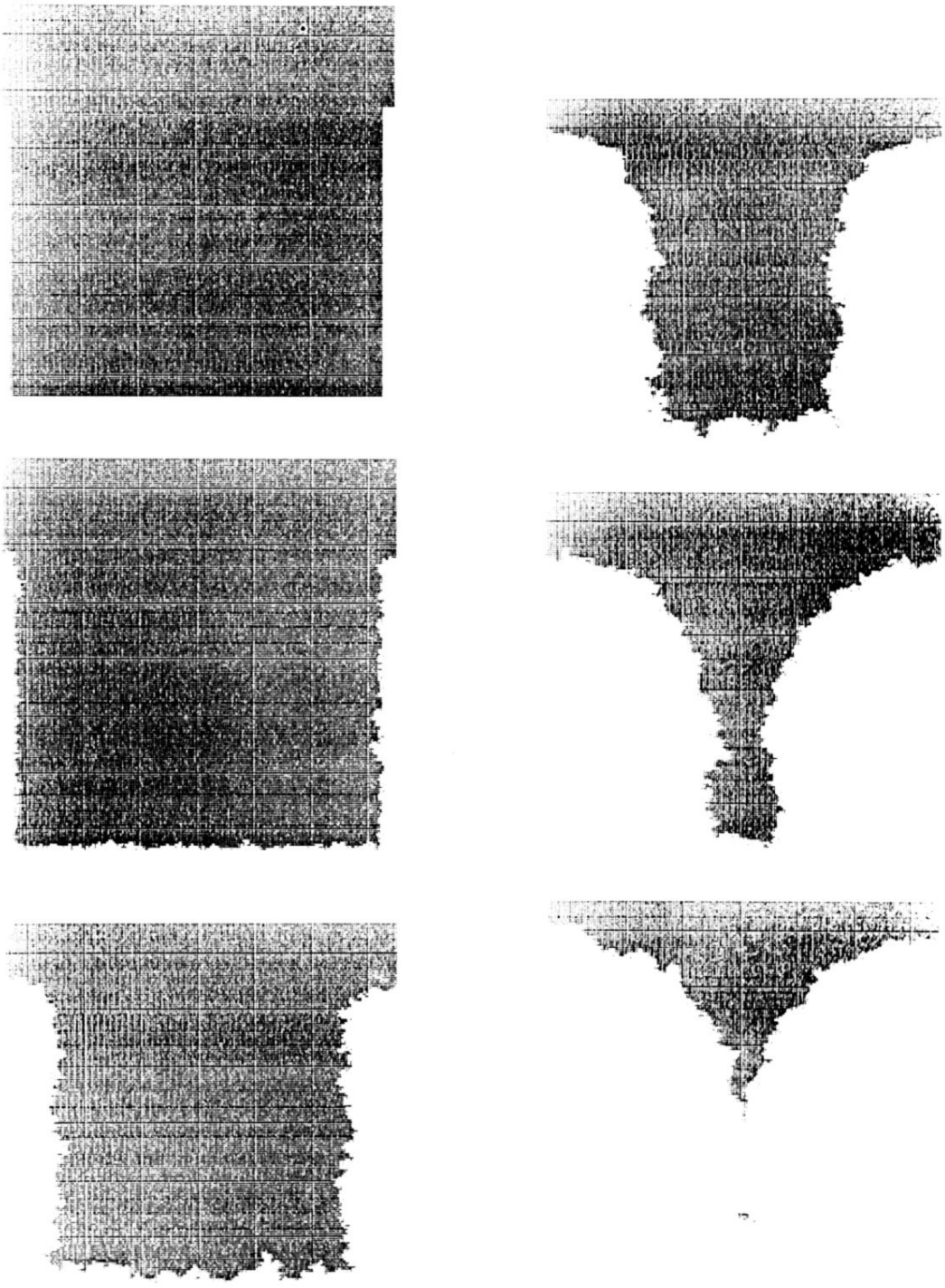


FIG. 11. Etching of a square stub attached to a plane, as a function of time. This is an isotropic etch with $n = 1$. The stub is 200 atoms wide.

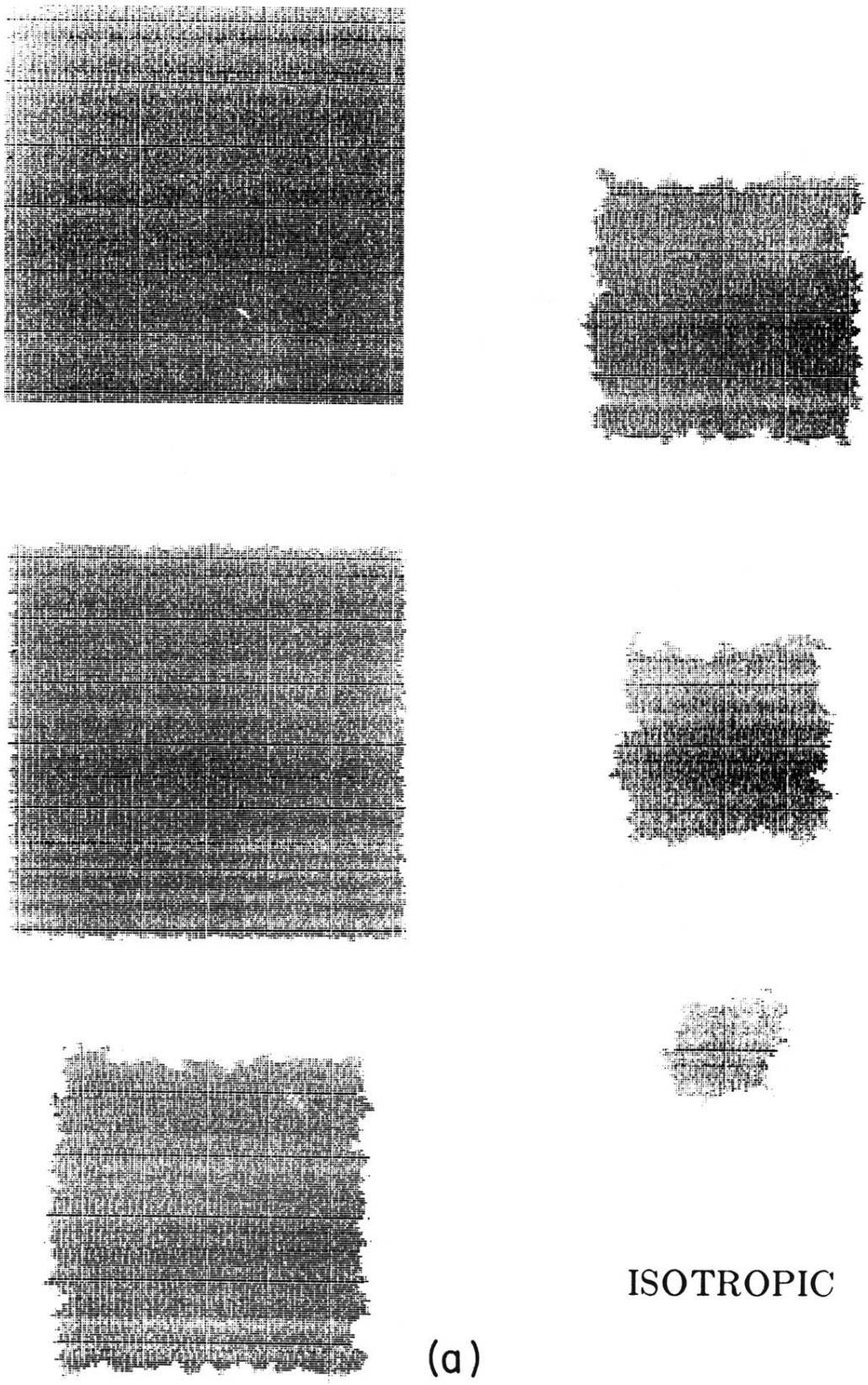


FIG. 12. A square surrounded on all four sides by etch. (a) Isotropic etch ($n=1$) and (b) anisotropic etch with $p(\mathrm{U})=p(\mathrm{D})=p(\mathrm{R})=p(\mathrm{L})=0.001$, $p(\mathrm{LR})=p(\mathrm{UD})=0.03$, $p(\mathrm{LD})=p(\mathrm{DR})=p(\mathrm{UR})=p(\mathrm{LU})=0.2$, $p(\mathrm{UDL})=p(\mathrm{UDR})=p(\mathrm{DLR})=p(\mathrm{LUR})=0.66$. The crystal is oriented such that the faces are stop planes.

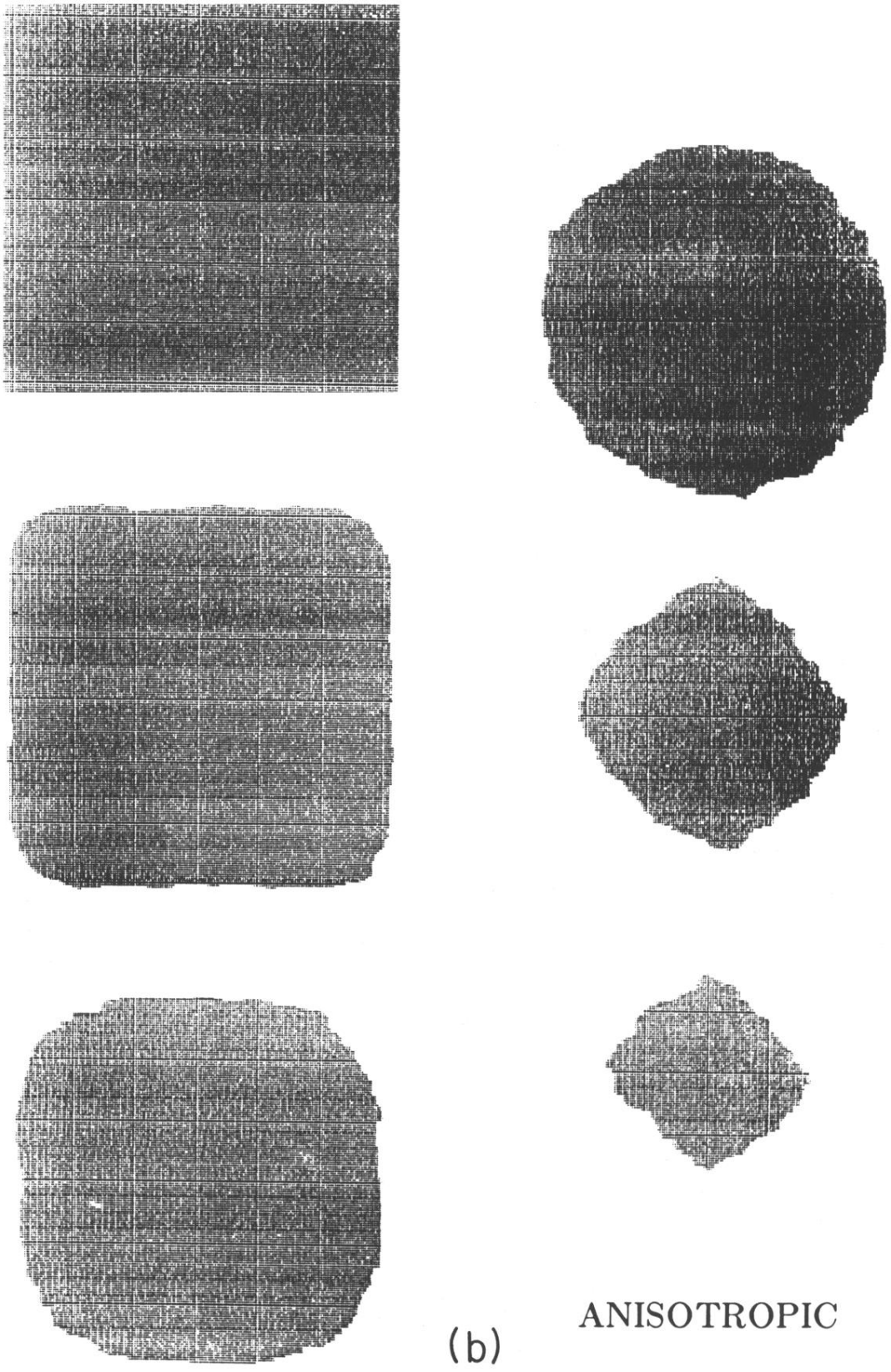


FIG. 12. (*Continued*).