Dynamics of pattern formation during low-energy ion bombardment of Si(001)

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Abstract

Sputtering of surfaces by collimated, low-energy ion beams results in spontaneous pattern formation in many systems. In order to explore the mechanisms that control the pattern formation, we have used in situ light scattering to measure the evolution of sputtered Si(001) surfaces. The results are interpreted within a linear instability model originally proposed by R.M. Bradley and J.M.E. Harper [J. Vac. Sci. Technol. A 6 (1988) 2390] that includes the dependence of the sputter yield on the local surface morphology. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

When collimated beams of low-energy ions are used to sputter surfaces, the surfaces can develop a distinct periodicity. An example is shown in Fig. 1, reproduced from [2]. In this case, a Si(001) surface has been bombarded with 750 eV Ar ions at an angle of 67.5° from the surface normal at a temperature of 555°C. The initially flat surface has developed a distinct ripple-like morphology with a period of approximately 400 nm.

This effect has been observed in a wide range of materials including semiconductors [2–10], oxide glasses [11–13] and metals [14–19]. Depending on the substrate and the ion beam parameters, the periodicity can range from tens to thousands of nanometers and the structure can be organized in 1-d ripples [3] or 2-d pits [19] or hillocks [10]. The structures form spontaneously, with no template required, and can uniformly cover a wide area of the surface.

At the core of this pattern formation is the interaction of the energy deposited by the incoming ion with the substrate surface. By depositing energy below the surface, removal of atoms by sputtering is fundamentally different than deposition of atoms on the surface. In this paper, we will review recent studies of the dynamics of pattern formation on sputtered surfaces of Si(001), focusing on the fundamental physical mechanisms that cause pattern formation and the modification of surface kinetic processes by ion bombardment.

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2. Measurements of ion-induced pattern formation

Many techniques for measuring surface morphology have been applied to the study of ion-induced pattern formation. Initial observations were performed using scanning electron microscopy (SEM) [3], but because of the poor sensitivity to depth of SEM, only ripples with large amplitudes could be studied. More recently, scanning probe microscopies (scanning tunneling microscopy (STM) and atomic force microscopy (AFM)) have been applied to this problem. In this case, the excellent depth sensitivity and spatial resolution of these techniques have made it possible to study the surface morphology, periodicity and correlation over a large range of conditions [4,6,14,15].

Although they provide detailed insight into the structure of the surface, these techniques require sputtering to stop in order to measure the surface. This makes it difficult to study the kinetics of surface evolution since multiple samples must be grown independently and compared. In order to understand the dynamics of pattern formation, in situ real-time probes are advantageous. Optical probes, both X-ray and visible, do not interfere with the sputtering process and have been used in several modes for studying pattern formation. X-ray reflectivity has been used to measure the RMS surface roughness [5,12] evolution during sputtering of SiO$_2$ and Ge surfaces. Grazing incidence X-ray scattering [19] has been used to study the amplitude and wavelength of ion-induced patterns, but the technique requires a synchrotron source.

Recently, we have developed an optical technique called light scattering spectroscopy (LiSSp) that makes it possible to measure the surface during low-energy ion sputtering. The technique, described briefly here and more fully in [20], measures the power spectral density (PSD) of the surface height as a function of surface wavevector, $K$. As calculated by Elson [21], under the conditions used in our work the PSD is proportional to the scattered intensity at the scattering vector, $K$, which is equal to the difference between the incident and reflected wavevector.

The PSD is determined by measuring the dependence of the scattered intensity on $K$, which depends on the wavelength of the light and the angles of incidence and reflection. In conventional light scattering, a fixed wavelength of light is used and the scattering vector is changed by modifying the scattering angles. However, when used as an in situ probe, it is not possible to access the range of scattering angles needed to determine the PSD. For studying sputter evolution, we have therefore developed a variation of this technique using spectroscopic detection. The scattering pattern is determined by using an incident white light source (Xe arc lamp) and measuring the spectrum of the scattered radiation with a solid-state spectrometer. In this way, the PSD can be measured at fixed angle, without having to move the sample or the detector. By measuring spectra at several different fixed scattering angles, a range of surface spatial periodicities from 300 to 10,000 nm can be measured simultaneously.

An example of a spectrum obtained from a Si(0 0 1) surface is shown by the solid line in Fig. 2. The inset in the figure shows the real space image of the surface from which the spectrum was measured. The squares indicate the equivalent PSD obtained by Fourier transforming the real space image. Note the excellent agreement between the peak position and peak shape; only the amplitude scale has been adjusted arbitrarily. Subsequent correlation of the amplitude of the light scattering measurement with AFM images demonstrates excellent linearity between the two techniques.
3. Results of Si(001) sputtering

The LiSSp technique has been used to study the evolution of ion-induced ripples on Si(001) surfaces. A sequence of LiSSp spectra taken during sputtering is shown in Fig. 3. The sputtering was done with 750 eV Ar ions at an angle of 67.5° from the surface normal along the [110] azimuth. The sample temperature was 560°C and the ion flux was 0.7 mA/cm². The wavevector corresponding to the peak in the LiSSp spectrum is equal to $2\pi/\lambda$, where $\lambda$ is the wavelength of the surface ripple. Note that the position of the peak does not change as its amplitude increases indicating that the ripple wavelength is fixed.

Multiple series of LiSSp spectra were measured to determine the dependence of ripple wavelength on sample temperatures and ion flux. The temperature dependence of the ripple wavelength is shown in Fig. 4, made over a temperature range of 460–600°C. The wavelength at the lowest temperature was outside the range of LiSSp and so was obtained by AFM measurements. The ion flux for all the temperatures in this figure was 0.7 mA/cm². The dependence of the ripple wavelength on ion flux ($f$) is shown in Fig. 5. In the following sections, we will discuss the physical significance of these measurements, focusing on understanding the flux dependence of the ripple wavelength and its
implications for the steady-state concentration of ion-induced defects.

4. Instability theory

The physical basis for the formation of ion-induced patterns is described by an instability theory originally proposed by Bradley and Harper [1]. This theory is valid for small amplitudes only so that non-linear terms are neglected. Consideration of non-linear effects have been addressed by others [22,23]. The height of the surface is defined as \( h(r, t) \) at any position, \( r = (x, y) \), and time \( t \). The evolution of \( h(r, t) \) is determined by a balance between roughening and smoothing processes that occur during sputtering, as shown schematically in Fig. 6. The roughening is due to removal of atoms from the surface by sputtering while the smoothing is accomplished by surface diffusion or other surface transport.

Using a theory developed by Sigmund [24,25], the effect of sputtering on \( h(r, t) \) can be described quantitatively. The key issue here is that the energy of the ion is deposited below the surface, as shown schematically in Fig. 6. The elliptical contours around the ion path represent contours of constant energy deposition. The probability of sputtering at a particular point on the surface is proportional to the amount of energy deposited there, and it turns out that the local sputter yield is therefore proportional to the curvature of the surface,

\[
\frac{\partial h}{\partial t} = S_e \frac{\partial^2 h}{\partial x^2} + S_s \frac{\partial^2 h}{\partial y^2}.
\]  

(1)

The sputter roughening parameters, \( S_e \) and \( S_s \), are linearly proportional to the ion flux and depend in a complex way on the incidence angle and ion beam parameters describing the range, the amount of energy deposited, and the longitudinal and transverse width of the energy deposition; the calculation is explained fully in the original paper by Bradley and Harper. These parameters can be obtained from simulations such as TRIM [26] that describe the ion–solid interaction within the binary collision approximation.

Counterbalancing the ion-induced roughening is the effect of surface diffusion, as described by Herring [27] and Mullins [28]. Diffusion, driven by surface energy minimization, occurs to smooth the surface morphology and is proportional to the divergence of the surface curvature,

\[
\frac{\partial h}{\partial t} = -B \nabla^4 h.
\]  

(2a)

The constant of proportionality, \( B \), is given by

\[
B = \frac{D \gamma}{n^2 kT},
\]  

(2b)

where \( \gamma \) is the surface energy, \( D \) is the diffusivity, \( C_m \) is the concentration of the mobile species on
the surface, \( n \) is the atomic density, \( k \) is Boltzmann’s constant and \( T \) is the surface temperature.

In addition, the arrival of the individual ions is a stochastic process that is not perfectly spatially uniform. Without the presence of the curvature-dependent sputter yield, the surface would still develop a Poisson roughness due to the random nature of the removal process. The roughening rate is described by a Poisson roughening term, \( \Gamma'(r,t) \). When these effects are put into a single rate equation, the evolution of the surface height is given by

\[
\frac{\partial h}{\partial t} = S_x \frac{\partial^2 h}{\partial x^2} + S_y \frac{\partial^2 h}{\partial y^2} - B\nabla^4 h + \Gamma'(r,t).
\] (3)

To simplify the analysis of this equation, consider the evolution of the individual Fourier components of the surface defined by

\[
h_q = h(r,t)e^{-iqr}dr.
\] (4)

Since this equation is linear, the evolution of the entire surface can be determined by superimposing the individual Fourier components. The expression for the evolution of \( h_q \) is

\[
\frac{\partial h_q}{\partial t} = R_q h_q + \Gamma_q,
\] (5a)

where

\[
R_q = S_x q_x^2 + S_y q_y^2 - Bq^4.
\] (5b)

\( R_q \) defines the rate at which each Fourier component grows or decays. At values of \( q \) larger than \( q_c = (B/S)^{1/2} \), surface diffusivity dominates and \( R_q \) is negative. Features with higher frequency than \( q_c \) will not grow. At values of \( q \) less than \( q_c \) (large wavelengths), the curvature-dependent sputtering dominates and \( R_q \) is positive. By finding the critical values of \( R_q \), the fastest growing wavevector, \( q^* \) is found. The corresponding real space wavelength is \( \lambda^* = 2\pi/q^* = (S/2B^{1/2}) \) and the maximum rate of growth is \( R^* = S^2/4B \). The resulting surface morphology is dominated by the evolution of the fastest growing Fourier component so that a preferred periodicity with wavelength \( \lambda^* \) develops.

5. Discussion

The theory described above enables us to interpret the measured surface periodicity in terms of the parameters \( S \) and \( B \). These parameters are directly related to the processes of sputter roughening and surface diffusivity, respectively. In this section, we discuss how measuring the temperature and flux dependence of these parameters is used to determine the fundamental surface kinetic processes controlling ripple formation.

The sputtering roughening terms, \( S_x \) and \( S_y \), depend on the collision cascade and are linearly proportional to the ion flux. These are expected to be relatively independent of temperature. The surface diffusivity parameter, \( B \), depends on both the diffusivity and concentration of the mobile defects on the surface. In the presence of the ion beam, the defect concentration may be modified by ion-induced defects so that the surface transport is different during ion bombardment than during thermal treatments. Separating out just the flux and temperature dependence, the dominant period of the surface morphology, \( \lambda^* \), is given by

\[
\lambda^* \propto f^{-1/2} \left[ \frac{D_s(T)C_{ss}(f,T)}{kT} \right]^{1/2},
\] (6)

where \( f \) is the ion flux and \( C_{ss}(f,T) \) is the steady-state concentration of mobile species on the surface which depends on both the ion flux and the temperature. We make the assumption that a steady-state concentration is achieved since a changing surface concentration would prevent a single surface periodicity from developing. In addition, we assume that the diffusivity is dependent on \( T \) but independent of flux.

The measured dependence of \( \lambda^* \) on temperature is consistent with an Arrhenius dependence of \( D_s(T)C_{ss}(f,T) \propto \exp(E/kT) \). The solid line in Fig. 4 corresponds to this form with a value for the activation energy of \( E = 1.2 \text{ eV} \). This result is similar to the activation energy for mobility of surface dimers on Si(001) as measured by STM [29,30], suggesting that the concentration of mobile species is only weakly dependent on temperature.

Understanding the processes controlling the concentration of mobile surface defects is essential
to understanding the surface transport controlling the ripple formation. Since the ion beam creates mobile surface defects, their steady-state concentration is determined by a balance between defect creation and annihilation mechanisms. Although we do not know the dominant annihilation mechanism a priori, different mechanisms imply a dependence of the steady-state surface concentration on ion flux. As expressed in Eq. (6), the surface spatial periodicity depends on \((C_{ss})^{1/2}\), so measurements of the ripple wavelength versus flux can be used to determine which mechanism is predominant.

Several different possible mechanisms for defect creation and annihilation are shown in Fig. 7 and considered below. In each case, the rate of defect creation (Fig. 7(a)) is linearly proportional to the flux, \(f\), and a defect yield, \(Y\).

### 5.1. Trap-dependent annihilation

In this case, the rate of defect annihilation is determined by diffusion to traps such as step edges or voids. The annihilation rate is proportional to the defect concentration and inversely proportional to a trapping time, \(\tau_{\text{trap}}\).

\[
\frac{dC}{dt} = fY - \frac{C}{\tau_{\text{trap}}}. \tag{7}
\]

The trapping time, in turn, is proportional to \(L^2/D\) where \(L\) is the mean spacing between traps, and \(D\) is the defect diffusivity. Solving for the steady-state condition gives the result that \(C_{ss} \sim f/D\) so that the ripple wavelength \(\lambda^*\) is independent of flux.

### 5.2. Islanding-dependent annihilation

In this case, defect annihilation occurs by the agglomeration of individual defects into clusters or islands. The rate of cluster formation is proportional to the square of the defect concentration so that

\[
\frac{dC}{dt} = fY - \frac{C^2}{\tau_{\text{island}}} \tag{8}
\]

and \(\tau_{\text{island}} \sim 1/D\). In this case, the steady-state concentration is given by \(C_{ss} \sim (f/D)^{1/2}\) and the ripple wavelength \(\lambda^*\) is proportional to \(f^{-1/4}\).

### 5.3. Ion-induced defect annihilation

In this mechanism, the rate of defect annihilation depends on the incoming ion so that the rate equation becomes

\[
\frac{dC}{dt} = fY - f\sigma, \tag{9}
\]

where \(\sigma\) is a cross-section for an ion-induced process. The steady-state concentration in this case is independent of the ion flux so that the ripple wavelength depends on \(f^{-1/2}\).

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Fig. 7. Schematic of possible mechanisms controlling creation and annihilation of mobile defects. (a) Creation of defects by incident ion. (b) Annihilation of defects at fixed traps. (c) Annihilation of defects by clustering. (d) Annihilation of defects by ion-induced mechanism. The flux dependence of each mechanism is discussed in the text.
The measured dependence of the surface periodicity on flux is shown in Fig. 5. The lines on the figure represent a power-law flux dependence, $\lambda = f^{-n}$, corresponding to the indicated exponents of 0, 1/4 and 1/2. The data clearly shows a flux dependence with an exponent of 1/2. This agrees with the mechanism of ion-induced defect annihilation, indicating that the steady-state defect concentration is independent of the ion flux. This result is not necessarily universal for other surfaces, or even for the Si(001) surface under different conditions. Further studies are planned to test the range of validity for this result.

6. Conclusion

We have used in situ light scattering to measure the dynamical formation of patterns on ion-bombarded Si(001) surfaces. This technique makes it possible to efficiently measure the temperature and flux dependence of the evolving spatial periodicity. The results are consistent with an instability model of ripple formation, and indicate that the steady-state concentration of mobile defects on the surface during ion bombardment is independent of the ion flux.

Acknowledgements

We gratefully acknowledge technical assistance and useful discussions with Tom Mayer and John Hunter. Portions of this work were performed at Sandia National Laboratories and supported by the United States Department of Energy under contract DE-AC04-94AL8500. Work at Harvard was supported by the US Department of Energy under contract DE-FG08-89ER45401.

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