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SPUTTER ROUGHENING INSTABILITY ON THE Ge(001) SURFACE: ENERGY AND FLUX DEPENDENCE

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ABSTRACT

We have measured surface roughening kinetics during low energy Xe ion sputtering of Ge (001) surfaces. The results are interpreted in terms of an instability theory developed by Bradley and Harper [1]. Although the calculated magnitude of the roughening rate does not agree with the measured value, the variation of the rate with ion flux and energy is in agreement with the theory.

INTRODUCTION

Sputtering by low energy ions is common in semiconductor processing and characterization. Ions are used, for example, in surface cleaning, reactive ion etching processes, and sputter depth profiling. Control of surface morphology during processing is becoming increasingly important as the feature size of advanced devices continues to decrease. Although it has been known for a long time that ion sputtering can lead to macroscopic roughening of surfaces [2], the atomistic origins of the roughening process have not been well understood.

We have used X-ray reflectivity (XRR) to measure the kinetics of surface roughness evolution during low energy sputtering [3,4]. In this work, we focus on the dependence of the sputter roughening rate on ion energy and ion flux. We interpret these results in terms of a microscopic model developed by Bradley and Harper [1] that explains the roughening phenomena in terms of a linear instability.

SPUTTER ROUGHENING INSTABILITY

The instability theory developed by Bradley and Harper relies on a theory of sputtering originally proposed by Sigmund [5]. While previous theoretical studies of sputtering have assumed that the sputtering yield is independent of the surface curvature [2], Sigmund's theory explicitly includes a curvature dependence. The consequence of the curvature dependence is that for periodic surface features, the troughs sputter faster than the crests so that sputtering increases the amplitude of the perturbation and leads to an instability. We briefly describe this theory below; for details, the reader is referred to the original papers [1,5].

In order to quantitatively describe the evolution of the surface morphology, we define the surface height profile, $h(r,t)$, as the height at point r relative to the average surface height. The effects of the various kinetic processes are most easily understood by considering their effect on a single Fourier component of the height profile,

$$h(q,t) = \int h(r,t) e^{-iq \cdot r} dr. \quad (1)$$

The evolution of an arbitrary surface can be understood as a superposition of the Fourier components.

The following linear equation describes the evolution of $|h(q,t)|^2$:

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$$d|h(q,t)|^2/dt = R_q |h(q,t)|^2 + \alpha \quad (2a)$$

$$\text{where} \quad R_q = -Fq + Sq^2 - Dq^4 \quad (2b)$$

The constant term, α , which is independent of spatial frequency describes the stochastic process of random atom removal. The positive Sq^2 term in R_q is derived from the Sigmund theory and indicates the dependence of the sputter yield on the curvature. It has the following dependence on the sputtering parameters [1]:

$$S = -fa/\rho Y_0(\theta) (\Gamma_1 \cos^2(\varphi) + \Gamma_2 \sin^2(\varphi)) \quad (3)$$

where f is the ion flux, a is the range of the ion in the solid, ρ is the atom density, θ is the angle of incidence and $Y_0(\theta)$ is the sputter yield on a flat surface. φ is the azimuthal angle between the incident ion direction and the surface wavevector, q . Γ_1 and Γ_2 are dimensionless coefficients that describe the curvature dependence of the sputtering rate and depend on the range and lateral extent of energy deposition and on the angle of incidence.

The negative terms in R_q lead to smoothing of the surface by the processes of viscous flow (F) and surface diffusion (D). The rates of smoothing, driven by surface energy minimization, have been calculated by Mullins [6] and Herring [7]:

$$F = \gamma/\eta \quad (4)$$

where γ is the surface energy and η is the coefficient of viscosity, and

$$D = 2D_s\gamma\Omega^2v/kT \quad (5)$$

where D_s is the surface diffusion coefficient, Ω is the atomic volume, and v is the number of molecules per unit area of surface.

Integrating this equation yields the time dependent behavior of $h(q,t)$:

$$|h(q,t)|^2 = h_0(q)^2 \exp(R_q t) + (\alpha/R_q) (\exp(R_q t) - 1) \quad (6)$$

where $h_0(q)$ is the initial roughness spectrum.

The sign of R_q in eq. 6 determines whether a particular spatial frequency will grow or decay during sputtering. Because the surface remains crystalline during sputtering at the temperatures investigated in this work, we can ignore viscous flow smoothing and set $F=0$. In this case, R_q is positive for all $q < S/D$, with a maximum value of $R^* = S^2/4D$ when $q^* = S/2D$. The existence of a critical wavevector (q^*) that grow faster than all others is consistent with the development of a pronounced ripple morphology on the surface which has been observed experimentally [2,3,4,8].

EXPERIMENT

We use X-ray reflectivity to measure the evolution of the surface roughness during sputtering. This technique allows us to measure the roughness in real time during the sputter bombardment with a resolution of 0.05 nm; for details of the measurement technique, see ref 9 and 10. All the results reported in this work are for Ge (001) surfaces and Xe ions. The sputtering is performed in a UHV vacuum chamber mounted on the X-ray system. The sputter

source is a Kaufmann style gun and the chamber is backfilled during sputtering to a pressure of 10^{-5} torr. The ions are incident on the sample at an angle of 55° from normal and along a $\langle 110 \rangle$ azimuth.

The surface roughness was obtained by fitting the XRR spectra using an optical multilayer theory [9]. The roughness was assumed to have a gaussian form. The measured mean square roughness, σ^2 , is related to the spatial frequency spectrum by

$$\sigma^2(t) = \int |h(q,t)|^2 d^2q / L^2 \quad (7)$$

where L is the lateral length scale of the measurement (typically $0.5 - 1 \mu\text{m}$ for the XRR technique).

ENERGY AND FLUX DEPENDENCE OF ROUGHENING RATE

The evolution of the surface roughness with time is shown in figure 1 for ion energies of 300, 500 and 1000 eV as indicated on the figure. Sputtering was performed at a temperature of 350°C . The dependence of roughening on ion flux is shown in figure 2 for sputtering by 1000 eV Xe ions at 300°C . The ion flux corresponding to each data set is indicated on the figure.

The solid lines in figure 1 represent a fit to the form given by eq. 6. In order to obtain this fit, a value for the surface diffusivity (D_s) of $4 \times 10^{-13} \text{ cm}^2/\text{s}$ was used (consistent with values obtained from previous measurements[4,11]). Fitting the data to this form allows us to determine the dependence of S on ion energy and flux. For the data in this work, the value of S can be obtained equally well by measuring the exponential roughening rate and equating this to the critical roughening rate, $R^* = S^2/4D$.

Since in the Bradley-Harper theory the critical roughening rate depends on S^2 , the measured exponential roughening is predicted to depend on the square of the flux. The measured roughening rate vs. the square of the ion flux is shown in figure 3. The linearity of the data indicates that the roughening rate does indeed depend on the square of the ion flux.

The dependence of S/f (where S has been normalized by the ion flux) on the ion energy is shown in figure 4. The solid line represents a power law fit, indicating that S scales like $E^{3/2}$ over the energy range measured.

In figure 5, we compare the results of our measurements of S/f with calculations using values for a and $Y_0(\theta)$ obtained from TRIM90 calculations [12]. As shown in the figure, the energy dependence of S is well explained by the TRIM calculations (a straight line indicates perfect correlation). As indicated by eq. 3, the energy dependence of the parameter S depends primarily on the sputter yield and the projected range. The other parameters, Γ_1 and Γ_2 , depend only weakly on ion energy.

The large discrepancy in the magnitude of S between the calculations and the measurements (a factor of roughly 550) is surprising since good agreement was obtained between the calculated values for SiO_2 roughening and the experimental values [3]. However, the same level of disagreement was observed when we measured the temperature dependence of Ge sputter roughening [4]. Uncertainty in the value of D could account for some of the disagreement between the theory and the measurement. However, it is unlikely that D is incorrect by 4 orders of magnitude as would be required to obtain agreement. Also, measurement of the sputter ripple frequency on Ge surfaces [4] require that D has roughly the value that we use here. An alternative explanation is that the theory was developed for amorphous surfaces, not the crystalline Ge(001) surface in this work. In fact we obtain good quantitative agreement with

Figure 3. Dependence of sputter roughening kinetics on square of ion flux. Roughening rate is determined from rate of exponential increase in surface roughness of data in figure 2. Linearity of data indicates roughening rate depends on square of flux as predicted by theory ($R^* = S^2/4D$).

Figure 4. Energy dependence of sputter roughening parameter S from data in figure 1. S is normalized by ion flux.

Figure 5. Comparison of measured value of normalized sputtering parameter (S/f) with calculations.

Figure 1. Evolution of Ge(001) surface roughness during Xe sputtering at energies of 300, 500 and 1000 eV. Surface temperature was 350°C. Solid lines represent fits to data using kinetics described in eq. 6.

Figure 2. Evolution of Ge(001) surface roughness during 1000 eV Xe sputtering for fluxes indicated in figure. Surface temperature was 300°C.

DISCLAIMER

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results for the sputter roughening of amorphous SiO₂ [3]. Finally, we know that during sputtering a significantly larger number of defects than the sputter yield are created transiently on the surface [13]. If the diffusion of these defects is biased by the surface morphology, then the rate of roughening could be significantly larger than that calculated by the Bradley Harper theory. More extensive AFM measurements of the development of the surface ripple structure at different energies and flux would be useful in resolving some of these issues.

CONCLUSION

We have measured the roughening kinetics of Ge (001) surfaces during sputtering with Xe ions. The energy and flux dependence of the roughening rate is consistent with the predictions of the Bradley-Harper instability theory. Because the roughening is due to an instability, the roughening rate depends on the square of the ion flux, not linearly on the ion flux. Although the trends in the data agree with calculations of the roughening rates from the theory using parameters obtained from TRIM simulations, there is a large discrepancy between the magnitude of the calculated roughening rate and the measured rate. Although we do not know the origin of this discrepancy, possibilities include the effect of surface crystallinity, surface diffusivity or transient ion-induced defects.

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