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ENERGY DEPENDENCE OF THE MOLECULAR EFFECT IN SPUTTERING

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ABSTRACT: We report measurements of sputtering yields of Au for Xe^+ and Xe_2^+ impact in the energy range 1-50 keV. It was found that non-linear effects exist well outside the range predicted in a recent thermal spike model.

Many sputtering experiments have been analyzed using the linear collision cascade theory, however, several workers have reported evidence that points out that in certain conditions the validity range of the linear cascade approximation is exceeded, for example, when the energy density in the cascade region is high enough as not to allow any longer to assume as negligible the probability of collision between moving atoms.

This condition is easily reached using heavy molecular ion beams, and results using these projectiles have been reported[1-4]. To have some further insight in the energy dependence of these non-linear effects, we have measured the sputtering yield of both atomic and diatomic Xe incident on Au at energies between 1 and 50 keV for Xe and 2.5 and 12 keV/atom for Xe_2, using a quartz crystal oscillator technique. The gold was evaporated on the face of the quartz crystal at a pressure of 8×10^{-9} Torr or less. The crystals used were AT cut and similar to those used by EerNisse[5], and the sputtering yield was calculated as:

$$S = \frac{k\, N_{av}\, \Delta f}{M_2 N_1} + \frac{M_1}{M_2}$$

where k is the calibration constant of the crystal, measured to be 1.78×10^{-8} g cm^{-2}Hz^{-1}, Δf is the frequency change and N_{av} is Avogadro's number. M_1 and M_2 are the masses of incident and target atoms, respectively, and N_1 is the number of incident atoms per unit area. The term M_1/M_2

takes into account the mass of the trapped projectiles[1] .

N_1 was determined by integrating the incident current, while the monitoring of the secondary electron yield provided a check on the state of cleanliness of the target surface.

The frequency change was measured with a digital frequency meter with an accuracy of 0.1 Hz. The base pressure in the high vacuum chamber was 5×10^{-10} Torr and climbed to 2×10^{-9} Torr during irradiation. The experimental equipment and techniques will be described in further detail elsewhere[6] .

To minimize the influence of dose effects on the sputtering yield the reported data were all taken at low fluences, i.e. before the measured fluence dependent increase in the sputtering yield was larger than the experimental uncertainty (i.e. $< 5 \times 10^{14}$ atoms/cm^2).

The measured sputtering yields are plotted in Fig.1, together with measurements by other workers[3,7-11] for Xe on Au. The measured diatomic yields are plotted at the corresponding yield and energy per incident atom. It is found that differences with previous data lie within the variations introduced by the different measurement fluences[3,5].

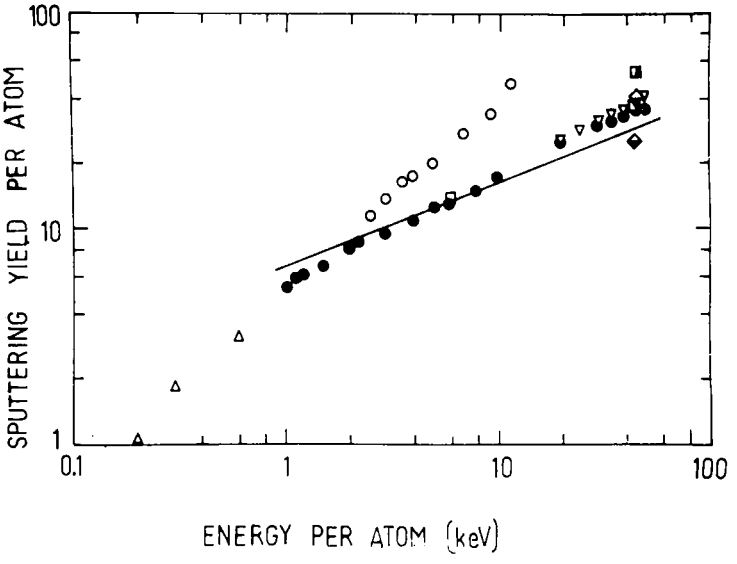

Figure 1 Sputtering yields per atom for Au bombarded with Xe and Xe_2. ▼ Almen and Bruce[7]; △ Rosemberg and Wehner[8]; □ ◨ Andersen and Bay[3]; ▽ Nenadovic et al[9]; ◇ ◆ EerNisse[10]; ○ Szymonski and de Vries[11]; ● Xe and ○ Xe_2, this work; ——— Sigmund's theoretical prediction.

As a theoretical estimate of the sputtering yield in the linear collision cascade approximation, we choose the one given by Sigmund's theory of sputtering[12], which is:

$$S(E) = \frac{\lambda \, S_n(E) \, \alpha}{U_o}$$

where λ is a calculable material constant, U_o is the surface binding energy, taken as 3.8 eV for gold[13], $S_n(E)$ is the nuclear stopping power, which was calculated by Lindhard et al[14], and α is plotted in ref.12. The calculated sputtering yields are also shown in Fig.1, and when compared with the experimental data, it is seen that the agreement is within the stated accuracy of the theory, although there is a difference in the energy dependence similar to what was found in other cases (see, for example, figs.17 and 18 of ref.12).

There is no previously published data for Xe_2 on Au. However, we may compare our observed enhancement in the yields for molecular ions with reported data on ions of similar mass, Te[3] and Sb[4], incident also on Au. The ratio of molecular to atomic yields are compared in Fig.2 as a function of the incident energy per atom in reduced units[14]. In this presentation, there seems to be a definite energy dependence, characterized by a range $\varepsilon < 0.01$, where nonlinear effects are larger the higher the energy, and by a region between $\varepsilon = 0.02$ and the highest energy where data exists, which presents a broad maximun.

A thermal spike model has been developed to predict the energy range where the nonlinear effects should affect the sputtering yield[15]. This model has been applied for Xe incident on Au, and extended for Xe_2 on Au by assuming that the energy per unit volume in the cascade is twice that of the atomic case, which corresponds to assuming a 100% overlap in the cascades of the individual projectiles, and thus represents an upper bound to the nonlinear effects expected from this model.

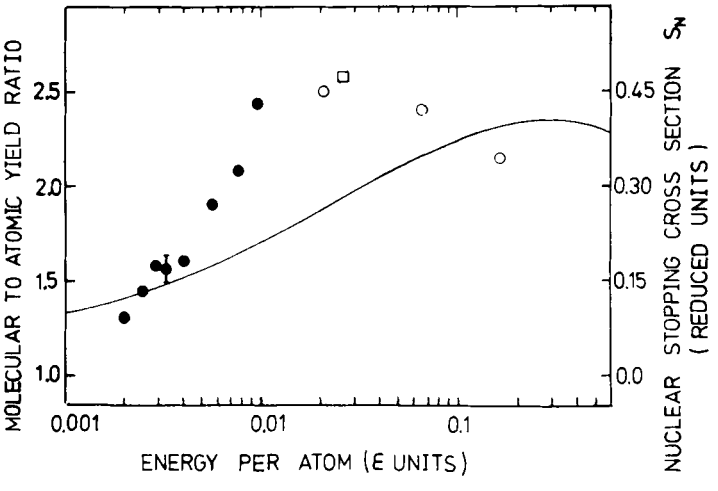

Figure 2 The molecular-to atomic yield ratios of Au as a function of the reduced energy[14], for: ● Xe, this work; O Te, Andersen and Bay[3]; □ Sb, Thompson and Johar[4].

In this case, the heat conduction equation supplies us with the scaling factor $\tau_{mol}=\sqrt{2}\ \tau_{atom}$ for the time constant of the cascade, while the slowing down time of the projectile, τ_o, and recoil atoms, τ', are taken to be the same for atomic and molecular projectiles.

The results of these calculations are plotted in Fig.3, where we can see that the mean energy per atom in the cascade exceeds U_o for energies less than 92 KeV for Xe and 175 KeV/atom for Xe_2 (ε=0.074 and 0.14 respectively) and the time constant of the cascade exceeds both τ_o and τ' for energies higher than 16 and 13 KeV/atom (ε = 0.13 and 0.010) for Xe and Xe_2 respectively. Thus,according to this model, we should expect a significant contribution of nonlinear effects to the sputtering yield in the range between 13 and 175 KeV/atom for Xe_2, and 16 and 92 KeV for Xe. For atomic Xe, there is no clear increase or deviation on the energy dependence that enables us to define a threshold for the appearance of nonlinear effects, and furthermore, as was mentioned before, there is good agreement in the absolute values of our measured data and calculated yields.

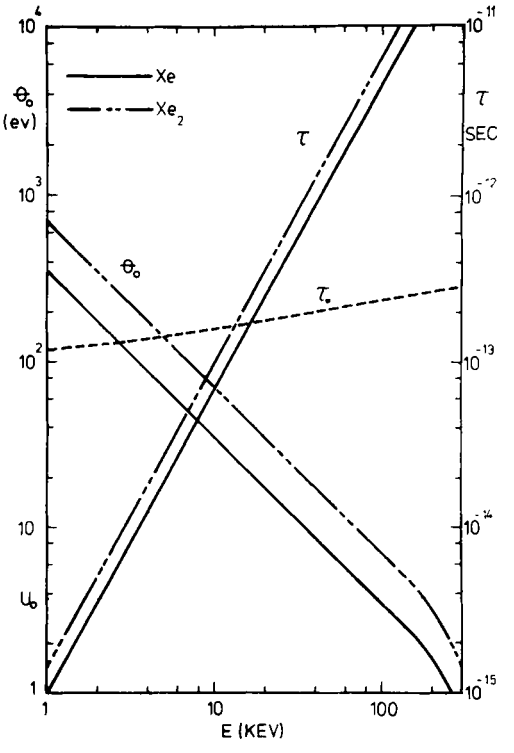

Figure 3 Energy density and time constant in the thermal spike[15] vs incident atom energy for Au bombarded with Xe^+ and Xe_2^+. τ_o is the slowing down time of the projectile and U_o the surface binding energy.

Examining Fig.2 it is evident that for molecular projectiles nonlinear effects take place at energies which are well outside the range predicted by the thermal spike model. We attribute this failure of the model to the no consideration of dense subcascades[16], which are connected with fluctuations in the deposition of energy, whose magnitude at the surface, according to a recent calculation[17], would be of the same

order as the mean value of the deposited energy, as well as to the small number of atoms involved in the cascade at low energies, which makes the applicability of the thermal approach questionable.

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