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Surface tension enhancement of TRIM sputtering yields for liquid metal targets

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Abstract

A critical part of all models of physical sputtering is the magnitude of the attractive interaction of a surface atom with the bulk, the surface binding energy, which is often equated to the heat of sublimation, independent of temperature and regardless of the state of aggregation of the target. Sputtering occurs when target particles which are accelerated by atomic collisions caused by an incident energetic projectile, reach the surface with enough energy to overcome the surface binding energy. This surface binding energy influences sputtering in two ways: (1) it acts as a barrier which must be overcome by the escaping atoms and so affects the sputtering yield, and (2) its directional properties influence the directions taken by the ejected target atoms. Lower surface binding energies increase the sputtering yield, while higher surface binding energies reduce the sputtering yield. The thermodynamics of liquid surfaces is used to determine the temperature dependence of surface tension and thus the binding energy of atoms in the liquid surface. Liquid lithium and gallium have surface tensions that follow the same universal function of T/T_c observed for most simple atomic liquids. The decrease of surface binding energy with increasing target temperature is calculated, and the TRIM code is then used to determine the resulting increase in sputtering yield with temperature as functions of the incident angle for these targets under bombardment by low energy deuteron projectiles. © 2001 Published by Elsevier Science B.V.

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1. Introduction

The interaction between the plasma and the materials used for limiters, divertors and vacuum vessels of fusion machines is characterized by extremely high heat fluxes and high fluxes of deuterium, tritium and various impurities. A solid state material exposed to this environment will melt and crack unless its temperature can be kept below about a third of the melting temperature in Kelvin [1]. One solution to this problem may be the creation of a cool dense gaseous radiating layer between the hot plasma and the material surface, however this may not protect these surfaces during extreme operating events such as disruptions. Other solutions involve making the surfaces movable in order to decrease local flux loads and provide opportunity for surface replacement. Of the various proposals for accomplishing this goal, the use of liquid metal flow, as first proposed in the UWMAK design [2] is believed to be one of the most promising. In that design a moving liquid lithium sheet was proposed for the moving metal cooling surface. Lithium has the advantage of being low $Z(=3)$, so the plasma can tolerate a relatively high concentration of it, but it has the disadvantages of burning when in contact with water and having a small difference between melting $T_m = 180°\mathrm{C}$) and boiling point temperatures ($T_b = 1324°\mathrm{C}$). An alternative candidate material, liquid gallium does not have these disadvantages ($T_m = 30°\mathrm{C}$, $T_b = 2247°\mathrm{C}$), though it is higher in $Z(=31)$. Both lithium and gallium have thermal conductivities that are good for cooling reactor walls (80 W/K for lithium at 100°C, 57 W/K for gallium at 250°C), although only liquid gallium has a thermal conductivity that increases with temperature. Experiments

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are now in progress to test these materials for use as limiter and divertor plates in tokamak fusion devices [3,4]. As part of this effort, measurements are being made of the sputtering [3], yields of these liquid metals when exposed to plasma. A pronounced temperature dependence is observed in the sputtering yield [3], and it is the objective of this paper to provide computer simulated data concerning this target temperature dependence of the sputtering and self-sputtering yields of liquid lithium and gallium as functions of the incident energy and angle.

Experimentally there is a considerable body of evidence which suggests that the surface state of a target under low-energy bombardment is the main factor in determining sputtering yield. It was recognized early on by Wehner that the heat of sublimation, atomic concentration at weaker atomic bonds, surface roughness, as well as structure, atom type and orientation were important parameters in the determination of whether an atom is sputtered [5]. An extensive theoretical treatment by Sigmund [6] of the sputtering yield for amorphous polycrystalline targets assumes a random slowing down of the projectiles in the infinite target media indicates that the majority of sputtered atoms originate from a very thin surface layer on the order of 5 Å thick, and the generalized yield formula from this treatment depends on only one property of the material – the surface binding energy. Sigmund also concludes that yield measurements on liquid targets are of special interest in understanding the applicability of this generalized theory to the case of low energy incident ions on monatomic amorphous targets. In a liquid, surface tensions are positive and diffusion rates are five orders of magnitude larger than the corresponding rate in a solid at the same temperature, so liquid structures should recover rapidly from any incident ion induced surface damage so as to present a surface of constant smoothness at the atomic level. The surface tension in solids is strongly negative, and with the slower diffusion cannot be atomically smooth. With negative surface tension, the tangential forces push the surface together causing buckling or wrinkling at the outermost atomic planes. Moreover liquids do not have pores or channels, and no long-range order, although structurally similar to the corresponding solid on a nearest neighbor basis, so channeling and focused collisions are complications eliminated from sputtering of liquid state targets. In either solid or liquid, the magnitude of the surface binding energies should not be hugely different because the latent heat of fusion is usually only a few percent of the heat of vaporization. In this paper we calculate the change in surface binding energy with temperature for the liquid metals lithium and gallium and use this energy in the TRIM code to determine the corresponding change in sputtering yield, as the incident energy and angle are varied.

2. Model

In the László and Eckstein [7] TRIM.SP [8,9] model of light ion sputtering from lithium and gallium the surface binding energies were taken to 1.68 and 2.97 eV, respectively, which are close to the heats of sublimation of the solid phase of these elements. In the liquid phase, the atomic density profile must vary continuously across the interface from the bulk to the outside surface, which is usually only a few atomic distances when far from the critical point. This implies a net attraction to the liquid phase for an atom in the surface transition region, which means that work must be done to move an atom from the bulk to the surface – the surface free energy. It also implies the existence of a tangential pressure (force per unit area transmitted perpendicularly) which is a function of position at the surface – the surface tension. The relation between these quantities – surface free energy and surface tension is a result from the classical theory of the capillarity (March [10], Orowan [11]). For simple atomic and molecular liquids, many observations of the surface tension, as summarized by Buff and Lovett [12], indicate that the surface tension is practically a universal function of T/T_c, a result first deduced by Van der Waals using the principle of corresponding states. Empirically, the surface tension and thus the surface binding energy of liquids is found to be usefully fitted by the relation [13]:

$$\gamma^{LV} = \gamma_0^{LV}(1 - T/T_c)^{1+r}, \tag{1}$$

where Guggenheim [14] predicted $r = 2/9$. This surface tension decreases nearly linearly as temperature increases, when far from the critical temperature. At the critical temperature the distinction between liquid and gas vanishes and so does the surface tension. This behavior of the surface tension with temperature has been observed for many liquid metals, including Gallium for both plasma liquid and gas liquid metal interfaces (see [15]). For Li, the surface binding energy at the melting temperature of $T = 453$ K is 1.6 eV, with a critical temperature of 3223 K, and a plot of the surface binding energy as a function of temperature, T is shown by the heavy solid line with negative slope of Fig. 1. The surface binding energy of Ga at its melting temperature $T = 302$ K was taken by 2.82 eV, and the critical temperature is $T_c = 5410$ K, and a plot of the surface binding energy versus T is shown in Fig. 2 (heavy solid line). Two versions of the TRIM code were used to calculate the sputtering yield as the surface binding energy was varied according to Eq. (1). These are SRIM2000 [16] and the TRIM95.SP [8] code. Both are Monte Carlo simulations based on the binary collision approximation and include the contributions of weak collisions to the complete cascade and target atoms are randomly distributed. The TRIM95.SP uses the

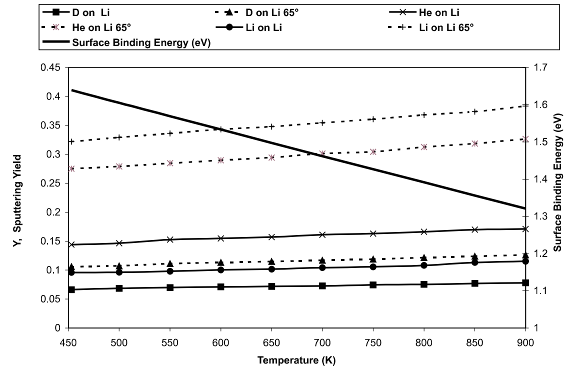

Fig. 1. Lithium surface binding energy versus target temperature from Eq. (1) and resulting yield calculated from SRIM2000 for D, He and Li projectiles at 75 eV normal incidence (solid lines) and 65° oblique incidence (dashed lines) on Li target.

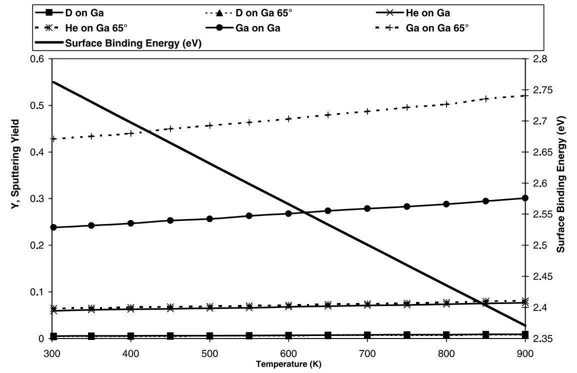

Fig. 2. Gallium surface binding energy versus target temperature from Eq. (1) and resulting yield calculated from SRIM2000 for D, He and Ga projectiles at 75 eV normal incidence (solid lines) and 65° oblique incidence (dashed lines) on Ga target.

Krypton–Carbon (Kr–C) interaction potential, and an electronic energy loss characterized by 50–50 Lindhard–Scharff and Oen–Robinson. The SRIM2000 code uses the Ziegler–Biersack–Littmark (ZBL) interaction potential, which may not be as good for large Z atoms due to its non-relativistic origin. For a given Z the two types of interaction potential are close but deviate most for large distances of closest approach and low energies. Both codes give comparable but not identical results for the sputtering yields. Statistics are similar, both codes are run for 99,999 incident projectiles. Both sputtering and the self-sputtering yields were calculated using each of the codes, the self-sputtering yields are normally very high because of the maximum energy transfer efficiency when mass of both projectile and target are the same. This tends to give better statistics for the sputtering yield, and fewer statistical lumps in the sputtering yield curves, but is a potential plasma catastrophe because the process becomes self-supporting as the yields increase above unity giving rise to runaway sputtering.

3. Model results

In Figs. 1 and 2 are plotted the sputtering yields for incident D, He, and Li projectiles on Li and D, He, and Ga projectiles on Ga targets with surface binding energies as functions of temperature determined by Eq. (1) at both normal (0°) and at a tangential incidence (65°), the latter chosen because in some fusion machines this is the mean angle of incidence of the impinging projectiles gyrating due to magnetic and electric fields [17]. As the temperature increases surface binding energy decreases almost linearly and sputtering yields increase almost linearly. The increase in sputtering yield with temperature is relatively modest, as expected from the analytical sputtering theory of Sigmund [6]. The smallest sputtering yields are found for the light deuterium on each of the targets, at normal incidence and the largest yields are self-sputtering at 65° incidence. SRIM2000 tends to predict a significantly larger helium sputtering yield than TRIM95.SP, while yields for other species tend to be more comparable between these two codes.

It is known from both experiment and theory that there is a strong dependence of the sputtering yield on the angle of incidence, especially for light ion bombardment.

In the case of solids, the angle of incidence is the most important parameter in determining how surface topography changes during sputtering, and is regarded as a more sensitive test of theories of the sputtering process than just the variation of the yield with energy. For the case of liquids we seek to establish the relative importance with respect to sputtering of the change in surface binding energy with temperature as expressed by Eq. (1) and the variation in the angle of incidence. In Figs. 3–6 are shown the variation of the sputtering yield with angle of incidence at each of the three different energies 75, 125 and 200 eV for D projectiles on Li liquid and solid targets using SRIM2000 (Fig. 3), D projectiles on Ga liquid and solid targets using TRIM95.SP (Fig. 4) Li projectiles on Li liquid and solid targets (self-sputtering) using SRIM2000 (Fig. 5), Ga projectiles on Ga liquid and solid targets (self-sputtering) using TRIM95.SP (Fig. 6). The liquid state uses the binding energy determined by Eq. (1) at 900 K, while the solid state target uses the heat of sublimation near the melting temperature as the surface binding energy. The dashed lines are the sputtering yields for the solid targets. For energies in the keV range, Bay and Bohdansky [18] observed for light ion sputtering on solid targets, the peak of the maximum of the yield becomes higher in angle the lighter the projectile and increases with increasing projectile energy and increasing surface binding energy. The

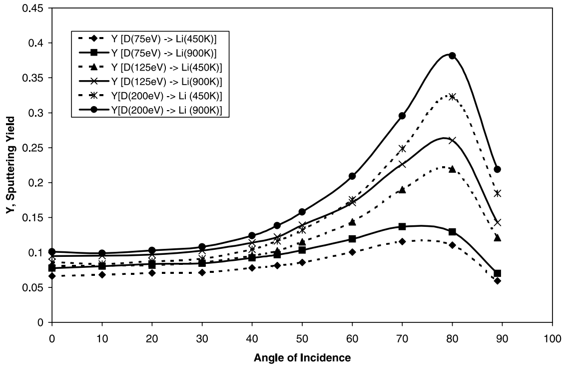

Fig. 3. Sputtering yield determined from SRIM2000 versus angle of incidence, for D projectiles at 75, 125 and 200 eV incident on solid lithium target near the melting temperature (heat of sublimation is the surface binding energy) (dashed lines) and on liquid lithium target at 900 K with surface binding energy determined from Eq. (1) (solid lines).

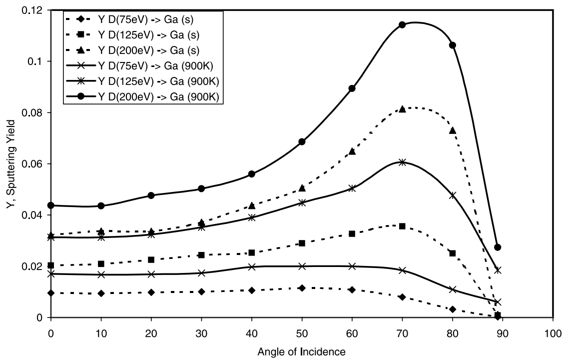

Fig. 4. Sputtering yield determined from TRIM95.SP versus angle of incidence, for D projectiles at 75, 125 and 200 eV incident on solid gallium target near the melting temperature (heat of sublimation is the surface binding energy) (dashed lines) and on liquid gallium target at 900 K with surface binding energy determined from Eq. (1) (solid lines).

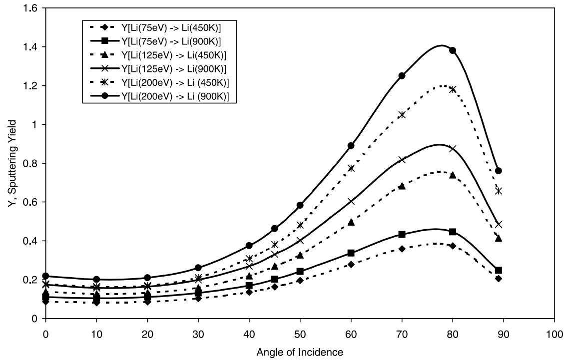

Fig. 5. Self-sputtering yield determined from SRIM2000 versus angle of incidence, for Li projectiles at 75, 125 and 200 eV incident on solid lithium target near the melting temperature (heat of sublimation is the surface binding energy) (dashed lines) and on liquid lithium target at 900 K with surface binding energy determined from Eq. (1) (solid lines).

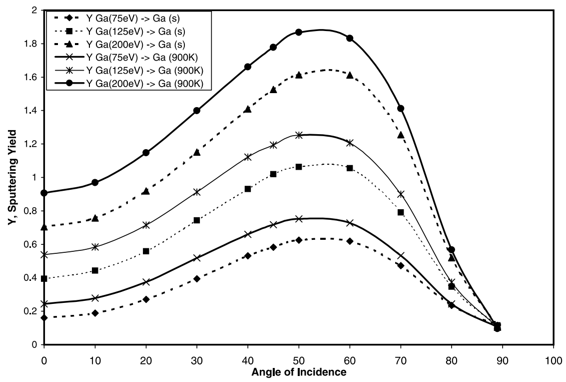

Fig. 6. Self-sputtering yield determined from TRIM95.SP versus angle of incidence, for Ga projectiles at 75, 125 and 200 eV incident on solid gallium target near the melting temperature (heat of sublimation is the surface binding energy) (dashed lines) and on liquid gallium target at 900 K (surface binding energy determined from Eq. (1) (solid lines).

first two of these effects are observed even in these plots at much lower energies. Compare the sputtering figures for D (Figs. 3 and 4) with the corresponding self-sputtering figures for Li and Ga (Figs. 5 and 6). The peak is higher in incident angle for the case of D sputtering than for the self-sputtering. Even in this limited range of energies, from 75 to 200 eV there is a clear sign of a shift in the peak to larger incident angles as the projectile energy increases. However, comparing the peak angle for sputtering from the solid and lower liquid surface binding energy does not show much change (compare dashed and solid line peaks). The theories of Yamamura [19,20], which extends the theory of Sigmund [6] to light ion low energy sputtering accounts for some of these observations, but at the lowest energies the incident angular dependence is stronger in the computer simulations than that obtained from the analytical theory. This has been observed in previous studies with solids at the heat sublimation surface binding energy [21], and remains the case for liquid metal targets [22].

4. Summary

The simulation results show that there is a weak dependence of the sputtering yield on the temperature of the target, which enters through the surface binding energy.

This dependence on temperature is more pronounced at the peak of the sputtering yield curve as a function of the angle of incidence than it is at normal incidence. Increasing the temperature of the liquid raises the sputtering dependence due to the universal relation for surface tension and surface binding energy on temperature.

The extensive self-sputtering yields due to light particle bombardment are the most severe restrictions on the maximum density allowed at the plasma edge. This becomes more pronounced for liquid metals due to the temperature dependence of the surface binding energy especially at some of the oblique angles where the sputtering yield peak occurs which may be close to that expected for incident particles in magnetic fusion devices.

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