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REMARKS ON THE THEORY OF FAST NEUTRON SPUTTERING*

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Abstract

Theoretical estimates of neutron sputtering yields are in serious disagreement with experiment, unlike the situation with ion sputtering. Possible reasons for the discrepancy are sought without success. It is shown that chunk ejection by neutrons is not due to single neutron events nor to the dynamic interference of cascades. The need for more complete experimental data to guide development of the theory is emphasized.

Introduction

Predictions of the rates of erosion of solids by fast neutron irradiation (1, 2) have been based on the sputtering theory of Sigmund (2). These estimates are not in agreement with experimental observations. It is the purpose of this paper to explore a few of the possible reasons for these rather puzzling discrepancies. Theoretical estimates of the neutron sputtering yields of four metals are listed in Table 1. These

TABLE 1

Neutron Sputtering Yields Calculated from the Sigmund Theory

Target	U_0 (eV)	Yield (atoms/ 10^5 neutrons)	
		^{235}U Fission	D-T Fusion
Al	3.33	2.4	4.2
Cu	3.50	1.9	6.0
Nb	7.56	0.89	2.7
Au	3.80	1.1	4.0

are based on the Sigmund theory and apply to spherical targets immersed in an isotropic flux of neutrons. Published calculations (3) of the energy deposited by fast neutrons were used. The primary recoil energies were corrected for inelastic atomic scattering by the method of Lindhard (4). Leachman's (5) form of the ^{235}U fission spectrum was

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used. The heats of sublimation were taken from Gschneider (6). As far as they exist, the experimental neutron sputtering data are assembled in Table 2. The observations are

TABLE 2
Experimental Neutron Sputtering Yields

Target	Neutron Source	Yield (atoms/ 10^5 neutrons)	Reference
Au	D-T	300	Garber (9)
		< 60	Keller (11)
	^{235}U fission	10	Anno (12)
		0.2	Verghese (14)
		500	Blewitt (8)
	Pu-Be	5×10^4	Keller (10)
Fe	^{235}U fission	600	Anno (13)
Nb	D-T	$(2.5 \pm 1) \times 10^4$	Kaminsky (7)

very scattered, so much so that some doubt must be expressed that even the magnitude of the yield is known. Nevertheless, it is likely that the predicted yields are much too low. While some of the discrepancy can be assigned to geometrical effects, since the experimental targets were not spherical and were not usually irradiated isotropically, most of it must be due to inadequacies in the theory.

Sigmund's theory (2) is based on the concept of a displacement cascade which underlies all radiation damage theory. He calculates the energy deposited in a thin region near the target surface as the displacement cascade initiated by the primary recoil spreads out, excluding any energy lost by electron excitation. Aside from geometrical factors, the sputtering yield is just the ratio of the deposited energy to the binding energy of surface atoms, this being taken as the heat of sublimation of the target. Sigmund compared his theory with the existing data on reflection sputtering (the ordinary kind) of polycrystalline metals by ions with energies from 10^2 to 10^6 eV. Further tests of the theory have been made by Andersen and Bay (15, 16). The agreement between theory and experiment is generally excellent. It is observed, however, that the yields measured for heavy ions, in the energy region where these are maximal, are often considerably greater than predicted. This is exemplified by the case of 10^5 eV Xe^+ ions on Cu, where the discrepancy reaches about 30%. To test the possibility that these discrepancies are due to nonlinear effects occurring in high density cascades, Andersen and Bay (16) compared the yields observed with monatomic and diatomic ions of the same velocity. The yields per incident atom were found to be significantly larger in the latter case. For example, the yield from 414 keV Te_2^+ ions on polycrystalline Ag was 5/3 the

yield from 207 keV Te^+ . The experiment with the molecular ions represents an extreme example of overlapping cascades and shows the importance of cascade nonlinearities. The nature of the nonlinearity is not known. It could involve, for instance, a cooperative overcoming of the surface binding forces when many adjacent atoms are set into motion at about the same time. A possibly related topic is the observation that sputtered ions often appear in rather large clusters (17), containing as many as ten or twelve atoms. Attempts to produce sputtered molecular species in computer simulations have so far proved unsuccessful (18).

Another class of experiment which seems not to be comprehended by the Sigmund theory is concerned with the temperature dependence of sputtering yields. Although the yields from polycrystalline metals are usually considered temperature independent, it has been observed (19, 20) that as the target temperature approaches the melting point, the yield may rise as much as a factor 10. This occurs over a range of 100 K or more. Thompson and Nelson (19) ascribe the additional sputtering to the evaporation of atoms by thermal spikes. Experiments with 25 to 45 keV Ne^+ , Ar^+ , and Xe^+ ions on several metals (20) lead to spike radii near 100 Å, lifetimes of a few picoseconds, and spike temperature rises of a few hundred degrees. Such a thermal spike is also a cascade nonlinearity and may well be very similar to that mentioned above. The description in thermal terms is not unique and should not be taken too literally, especially since the spike lifetimes are so short that the concept of temperature is not really applicable. Nonetheless, the thermal spike approach provides a method for dealing, however clumsily, with cooperative ejection of particles from a solid surface.

Dynamic Interference Between Cascades

Kaminsky's original experiment (7) on the 14 MeV neutron sputtering of Nb was characterized by the observation that the ejected material appeared in large chunks. Simple arguments on energetics make it plain that single neutron hits cannot be responsible for the ejection of such chunks. A typical chunk may be regarded as a sphere of radius 1 μm . Such a sphere contains about 2.3×10^{11} atoms altogether, of which about 1.8×10^8 are in the surface. The minimum energy for producing such an object is probably its surface energy. That is, we imagine the chunk to be quarried out of the irradiated foil, the only cost being creation of the associated new surface. The free surface energy of Nb is about 0.18 eV per atom (21). Thus, at least 32 MeV would be required to eject such a chunk, an amount far in excess of that available from 14 MeV neutron hits on Nb, where the maximum recoil energy is about 600 keV. Thus, single neutron effects cannot be responsible for the ejection of such chunks.

The possibility that two or more cascades may interfere and thus produce nonlinear effects is easily calculated: Assign a characteristic dimension L to each displacement cascade, such that sputtering occurs only if the cascade is within L of a surface. The rate at which such cascades occur, per unit area of the target surface, is $n\sigma\phi L$, where n is the atomic density of the target, σ is its neutron cross-section, and ϕ is the flux density of the incident neutrons. The neutron hits are distributed in space and time according to Poisson statistics. Thus, the probability that one or more neutrons will make collisions in a volume V during a time t is

$$1 - \exp(-n\sigma\phi Vt) \approx n\sigma\phi Vt$$

as long as the collision probability is small. For two cascades to interfere, V must be approximately L^3 and t must be the lifetime of a cascade. Thus, the rate at which interferences occur is

$$B = (n\sigma\phi)^2 L^4 \tau$$

where τ is the cascade lifetime.

The quantities L and τ can be estimated from simple considerations. The former may be regarded as the total pathlength traversed by the primary recoil. Fig. 1 shows estimates of L for Nb recoils in Nb. These are taken from the work of Schiøtt (22) who

FIG. 1

Total Path Length Calculated for Nb Recoils in Nb.

used the well-known LSS theory (23) and from the hard-core approximation calculation of Holmes and Leibfried (24), using an exponentially screened Coulomb potential to describe the atomic scattering. The two LSS curves are for no inelastic energy loss ($k_L = 0$) and for an amount of inelastic loss based on Lindhard's theory of electronic stopping (4, 23). The LSS estimates are the more reliable, but the hard core results set a strong upper limit. Fig. 2 shows similar estimates of the time necessary for the primary to slow down from its initial energy to various final energies, E_f . This time is approximately the time necessary to produce the entire cascade. The Holmes-Leibfried treatment of L is easily modified to estimate the slowing down time (25). Another estimate can be obtained

FIG. 2

Slowing Down Times Calculated for Nb Recoils in Nb.

by integrating the nuclear stopping power using Lindhard's treatment (26) of an r^{-2} potential. Again the hard core results set a strong upper limit.

From Figs. 1 and 2, we take for 600 keV Nb recoils the values $L = 0.45 \mu\text{m}$ and $\tau = 3.6 \text{ psec}$. With $\sigma = 3.91 \text{ barns}$ and $\phi = 2.4 \times 10^{10} \text{ n/cm}^2 \text{ sec}$, the interference rate is $B = 3.8 \times 10^{-10} \text{ cm}^{-2} \text{ sec}^{-1}$. In 54 hours, the number of interferences is only $7 \times 10^{-5} \text{ cm}^{-2}$, to be compared with the observed (7) density of chunks, around 10^3 cm^{-2} . Thus,

dynamic interferences between cascades cannot account for the ejection of chunks. It must be concluded that the ejection of chunks in neutron sputtering is primarily associated with aspects of the target which are not considered in displacement cascade production theories, such as details of microstructure.

Thermal Spikes and Neutron Sputtering

Although the thermal spike concept has not been a very fruitful one in radiation damage, the success of Thompson and Nelson (19, 20) in applying it to the temperature dependence of ion sputtering makes it worth examining in the context of neutron sputtering. We imagine a thermal spike to be a sphere of radius L which persists for a time τ . Within the sphere and during its life, the atoms are heated to a uniform temperature T_s . If one of these spheres should intercept the target surface, atoms would evaporate from the surface. The flux density of atoms evaporating from a plane surface at a temperature T is

$$f(T) = 4n (kT/2\pi m)^{1/2} \exp(-U_0/kT) \quad (1)$$

where k is Boltzmann's constant, m is the mass of the evaporating atoms, and U_0 is the binding energy of atoms in the target surface. This result is obtained by integration of the evaporation energy spectrum given by Thompson and Nelson (19). The spike temperature can be associated with the energy of an atom recoiling from a neutron interaction by

$$E = 2\pi L^3 n k(T_s - T_0) \quad (2)$$

where T_0 is the temperature of the target. Assuming for simplicity that the neutrons are monoenergetic and that the recoiling atoms are uniformly distributed in energy, the neutron sputtering yield can be written as

$$S = n\sigma \int_0^{E_m} f(T_s) (dE/E_m) \tau \int_0^L \pi(L^2 - x^2) dx \quad (3)$$

where T_s is defined by Eq. (2), E_m is the maximum possible recoil energy and x is the distance of a thermal spike below the target surface. If the target temperature is neglected and if the spike parameters L and τ are taken as constants, Eq. (3) can be written as

$$S = \frac{8n\sigma E_m \tau}{3(2\pi m U_0)^{1/2}} \left[y^4 \int_y^\infty e^{-u^2} \frac{du}{u^4} \right], \quad (4)$$

where

$$y = [2\pi L^3 n U_0 / E_m]^{1/2}. \quad (5)$$

The function of y in Eq. (4) can be evaluated in terms of tabulated functions. We note here only that it has a maximum value of about 0.1 when y is about 0.5.

To compare Eq. (4) with the measurements of Blewitt, et al. (8) on Au, assume that 2 MeV neutrons typify the ^{235}U fission spectrum. Then, with $E_m = 40$ keV and $\sigma = 6.4$ barns, we must have $S < 6 \times 10^{-5} \tau$, where τ is measured in psec. Since the lifetime of a cascade will probably not exceed about 5 psec, yields in excess of about 3×10^{-4} would not be expected. At the same time, to obtain a value of y near 0.5 requires L to be about 20 Å. From Schiott's tables (22), the range of Au recoils in Au is about 90 Å at 40 keV and about 60 Å at 20 keV. Thus, the thermal spike model used here requires spikes that are smaller (and therefore hotter) than expected. This could merely reflect the assumption that the spikes are spherical. The model could be modified to consider spikes which were cylinders or prolate ellipsoids with major dimension corresponding to the primary recoil range. While this calculation has not been carried through, it could give reasonable cascade dimensions, although it would probably require lifetimes greater than is plausible. Finally, application of Eq. (4) to 14 MeV neutron sputtering of Nb gives $S < 5 \times 10^{-4} \tau$ (psec) and L about 40 Å, the latter to be compared with the much larger figures in Fig. 1. Here the neglect of inelastic energy losses in cascade development must be significant. It also seems possible that subcascade formation at the high energies of these Nb recoils might need to be considered.

Discussion

While the thermal spike treatment has some possibility of accounting for neutron sputtering yields greater than those given by the Sigmund theory, it should be clear that no real theoretical understanding of neutron sputtering yet exists. This depressing state of affairs cannot easily be rectified by tinkering with the essentials of cascade production theory, of defect migration theory, or the like, since changes in these areas would have important implications in the theory of ion sputtering, where agreement between theory and experiment is satisfactory. However, it must be said that the experimental ion sputtering data are generally in accord among different workers, something which Table 2 shows not to be the case in neutron sputtering. Before much useful theory can be done in the latter field, it will be necessary to improve greatly the experimental situation, to the extent that one can say with reasonable certainty what the yields are in particular cases.

The remarks made here have certain implications for research on fusion devices. The general lack of theoretical understanding of neutron sputtering is so complete that one must consider that it may be strongly influenced by almost any other accompanying influence on the CTR first wall. Thus, alteration of the microstructure of the wall by

the formation of blisters, voids, or gas bubbles, roughening of the surface by ion sputtering, and thermal effects must all be considered of potential significance in neutron erosion phenomena. Thus, many complicated experiments involving these variables must be contemplated, at least until some reasonable understanding has been achieved.

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