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ON THE USE OF THE PHENOMENON OF AMORPHIZATION OF CRYSTALS FOR CLARIFYING THE MECHANISM OF CATHODE SPUTTERING

(Presented by Academician L. A. Artsimovich on 25 II 1969)

1. In recent years it has been established that irradiation of crystals by fast particles under certain conditions can lead to the accumulation of radiation defects and, as a consequence, to amorphization of the crystals ($^{1-3}$). It has also been established that observation of the process of amorphization of crystals under ion bombardment and of the reverse process—the restoration of the crystalline structure—is one of the few methods that make it possible to study the kinetics of formation and annealing of radiation defects in a dynamic regime ($^{4-6}$). The results presented in the present work show that the phenomenon of amorphization can also be successfully used to clarify the mechanism of destruction of solids by ion bombardment (cathode sputtering).
2. Fundamental in theories of cathode sputtering is the question of the mechanism by which the energy released in the collision of the bombarding ion with an atom of the target is carried to the surface of the target.

In **cascade** theories it is assumed that the displaced target atom formed as a result of the impact of the primary ion (the recoil atom) produces a cascade of collisions possessing the following properties (see (^{7})). The cascade is a sequence of independent binary collisions of target atoms. The arrangement of target atoms is considered random. An atom can leave the place it occupies and begin to move freely in the target if the energy it receives from another atom exceeds a certain threshold value. These properties of cascades are identical in all, without exception, theories of radiation effects in solids. Other properties (for example, the form of the energy-transfer cross section) may differ somewhat in different theories (^{7}). Thus, strictly speaking, cascade theories are applicable only to amorphous bodies, since in them all effects that may be due to the ordered arrangement of the atoms of the target are disregarded *a priori*. According to cascade theories, sputtering occurs when part of the displaced atoms formed in the cascade reaches the surface of the target and leaves the target.

Modeling of the dynamics of radiation defects in crystals has shown that the ordered arrangement of atoms has a large influence on radiation effects. In particular, it was found that correlation of collisions of crystal atoms plays an essential role. Owing to collision correlation, both energy and matter can propagate (if the conditions ([8]) are satisfied) along close-packed crystallographic directions over distances considerably exceeding those over which they can propagate in a disordered medium. The mechanism of energy transfer by means of focusing correlated collisions (focusons) forms the basis of focuson theories of sputtering ([9,10]).

In **focuson** theories it is assumed that the energy released in the collision of the incident ion with a target atom is expended

exclusively for the formation of focusons, and focusons of only one type—those that propagate along the closest-packed axes of the crystal. If the energy delivered by such focusons to the target surface exceeds the binding energy of a surface atom, that atom is detached from the target and sputtering occurs. The focuson mechanism naturally explained the experimentally observed preferential yield of sputtered material along the crystallographic axes of the target, which was regarded as the main argument in favor of focuson theories. Recently, however, it has been shown ([11,12]) that such an explanation of the preferential yield is not the only possible one.

It should be noted that both cascade and focuson theories contain parameters that are not known with complete accuracy. Therefore, the authors of these theories considered it a great success that the sputtering coefficients they obtained differed from the experimental values by no more than a factor of two ([13])—this, of course, does not apply to semiempirical theories.

3. From the above enumeration of the initial assumptions on which the cascade and focuson theories of sputtering are based, it follows that, in view of the extreme idealization of these assumptions, in any experimental test of the existing theories of sputtering the issue can by no means be the choice of one of the two sputtering mechanisms, but, at best, only an estimate of the relative role of these mechanisms in each particular case of sputtering. One possible way of carrying out this estimate consists in comparing the sputtering coefficients of substances that are similar in other properties, in one of which both mechanisms of energy transport may exist, while in the other only one of them may exist ([11]). If it turns out that the sputtering coefficients of such a pair of substances are close, it is natural to suppose that the mechanism of energy transport that is impossible in one of the substances is not significant*. If, however, the sputtering coefficients differ noticeably, then, in order to estimate the relative role of the mechanisms, some other property of the pair of substances being compared should be

invoked, for example the electron-emission coefficients (see also (14)).

As a pair of substances whose sputtering coefficients it is expedient to compare, the most natural choice is the amorphous and crystalline phases of one and the same element. The phenomenon of amorphization under ion bombardment provides a unique opportunity, by a simple change in the temperature of the target, to transfer the target from the crystalline state to the amorphous one, and vice versa. Thus, the phenomenon of amorphization makes it possible to obtain a comparable pair that is closest in all other properties except structure.

4. A germanium crystal was chosen as the target. The choice of target was due to the following reasons. First, upon bombardment of a germanium crystal by ions of both low (13) and high (15) energies, a preferential yield of sputtered material along the crystal axes is observed, and this is considered evidence for the possible existence of focusons in this crystal. Second, it has been reliably established by electron-diffraction methods (2,3) that, upon bombardment of a germanium crystal with germanium ions, it passes specifically into the amorphous state. Third, annealing of defects occurs at a sufficiently high temperature, which facilitates carrying out the experiment. Irradiation of the (111) face was performed with argon ions of energy 30 keV and a current density of 0.5 mA/cm^2. The method for determining the sputtering coefficient S and the coefficient of electron the emission coefficient γ was conventional (14). The dependences of S and γ on the angle of incidence φ of ions on the target were measured at different target temperatures. The target was rotated about the [110] axis and in the direction toward the [001] axis.

The experimental results are shown in Fig. 1. It can be seen that at a target temperature of 440° the curves have the usual form characteristic

Fig. 1. Dependences of the sputtering coefficient S and the secondary-electron emission coefficient γ on the angle of incidence φ of ions on the target. 1—at 220°, 2—at 440°. On the right—the dependences of S and γ on target temperature at different angles of incidence of ions on the target.

for ordered structures. At a target temperature of $\sim 200°$, the angular dependences of both S and γ are smooth. The transition from one type of dependence to the other is reversible and occurs in a rather narrow temperature interval (in the figure on the right).

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* An attempt of this kind was undertaken in (16). On the basis of the closeness of the sputtering coefficients of Mo, W, and the alloy W-Mo, and assuming that focusons are impossible in the alloy, the authors concluded that in the case studied the focuson mechanism is insignificant. In our opinion, however, without a careful metallographic analysis of the alloy lattice, the interpretation of the experimental results is not entirely correct.

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Figure 1: Fig. 1. Dependences of the sputtering coefficient S and the secondary-electron emission coefficient γ on the angle of incidence φ of ions on the target. 1—at 220°, 2—at 440°. On the right—the dependences of S and γ on target temperature at different angles of incidence of ions on the target.

It can be seen that the general character of the change in the sputtering and emission coefficients as functions of target temperature at different angles of incidence of ions on the target is not the same. In those angular regions where maxima and especially minima occur, both S and γ change considerably with a change in target temperature. Such changes are naturally associated with a change in the probability of collision of an ion with a target atom. Of greater interest to us, however, is that at those angles of incidence where there are neither maxima nor minima, the sputtering and emission coefficients of amorphous and crystalline germanium are close. Thus, amorphization of germanium and, as a consequence, the impossibility of a focusing mechanism for energy transport do not lead to a decrease in the sputtering coefficient. On this basis it appears possible to conclude that, in the sputtering of germanium, the mechanism of focused collisions does not make a noticeable contribution to sputtering.

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