DIRECTED EMISSION OF PARTICLES FROM A COPPER SINGLE CRYSTAL SPUTTERED BY BOMBARDMENT WITH IONS UP TO 50 kev ENERGY

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Sputtering of the (100) plane of a copper single crystal was studied for various energies and angles of incidence of argon and hydrogen ions. It is shown that the particles of the sputtered matter retain a favored direction of emission along some crystallographic directions ([100], [110], etc.) when the energy of the bombarding ions is raised up to 50 kev. Thus a deposit in the form of separate patches is formed on a screen parallel to the (100) face of the copper. The pattern of the deposit changes with increasing ion energy, but is practically independent of the angle of incidence of the particles. The density distribution law of the patches corresponding to the [110] and [100] directions has been studied. The spot material is distributed according to a cosine law. The relief of the (100) copper plane produced through sputtering by A^+ and H_2^+ ions with energies up to 50 kev was also studied. The data obtained cannot be reconciled with existing theoretical papers on cathode sputtering.

INTRODUCTION

THE sputtering of a substance by ion bombardment is a complex process, usually accompanied by many supplementary phenomena which make its investigation difficult. Therefore, in spite of the extensive material already accumulated on cathode sputtering,[1,2] the mechanism by which the ions interact with the sputtered substance is not yet clearly understood.[3-9]

We note that until recently only sputtering of polycrystalline samples in gas-discharge plasma was studied. This yielded averaged values for the measured quantities and made the interpretation of the results difficult. Many phenomena connected with the crystalline structure of the sputtered substance dropped out of view in this method. Consequently, the dependence of the sputtering coefficient and sputtering threshold of single crystals on the crystallographic direction was not formulated with sufficient rigor and was not investigated. Not until 1955 did Wehner[10] show that a metallic single crystal is sputtered most intensely in the close-packed crystallographic directions, if the bombarding-ion energy does not exceed 200 or 300 ev. It was proposed in reference 10 and other papers[7,8] that the preferred sputtering of a single crystal in particular directions would disappear if faster ions were used. However, in later experiments[11] the energy of the bombarding ions was raised to 5 kev and copper single crystals nevertheless were observed to sputter preferably in the close-packed [110] directions. In addition, it was noted that when the ion energy exceeded 1 kev, copper single crystals were intensely sputtered also in the [100] directions, which follow the [110] directions in the order of close packing.

It was deemed interesting to ascertain whether the sputtering coefficient of a metallic single crystal depended on the crystallographic direction at higher bombarding-ion energies (up to several times ten kev). We therefore investigated the sputtering of copper single crystals by beams of argon and hydrogen ions with energies up to 50 kev.

The use of an ion beam[12-15] offers many advantages over sputtering in gas-discharge plasma, where the number, energy, and angle of incidence of the ions cannot be determined accurately. The use of an ion beam has allowed us in the present investigation to study the sputtering of a single crystal copper by ions incident at various angles on the investigated (100) face.

EXPERIMENTAL SETUP

The experimental setup is illustrated in Fig. 1. The ion beam was produced by an ion gun, the principal element of which was an Ardenne-type[16] ion source with double contacting of the plasma, improved by several investigators.[17]

The ion gun produced a well focused beam (6 — 12 mm in diameter) of argon and hydrogen ions,

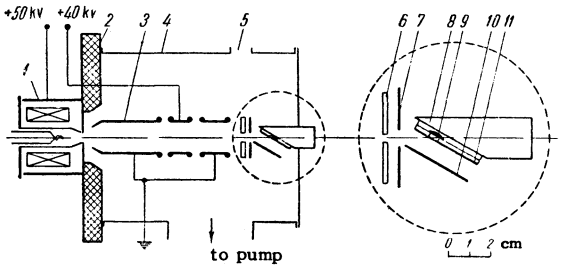

FIG. 1. Diagram of experimental setup: 1 – ion source, 2 – insulator, 3 – single electrostatic lens, 4 – container, 5 – viewing window, 6 – quartz screen, 7 and 10 – glass collectors, 8 – copper holder, 9 – copper single crystal, 10 – mica diaphragm.

the currents being 3 and 20 ma respectively at an accelerating voltage of 50 kv.

The ion beam, accelerated and focused by the single electrostatic lens, passed through an opening (4 – 8 mm in diameter) in a quartz screen and struck the specimen. The sputtered particles were deposited on a mica or glass collector, placed in front of the specimen parallel to its working surface. The collector used at normal ion incidence ($\alpha = 0°$), was a glass disk (7, Fig. 1) 80 mm in diameter with an opening 5 – 10 mm in radius for the ion beam. The collector for inclined beams ($\alpha = 45$ and 60°) was a rectangular glass plate measuring 30 × 40 or 40 × 60 mm. The distance between the sputtered plane and the collector ranged from 16 to 20 mm.

The object was a single crystal of copper 15 × 15 × 5 mm attached with low-melting point cadmium solder to a heavy water-cooled copper holder, so that the sample temperature did not exceed 100 – 200° C during the experiments. Before starting a series of experiments, the (100) face of the single crystal intended for the sputtering was ground and washed with alcohol. The dimensions of the sputtered surface were limited by a mica diaphragm with round hole of 5 to 6 mm diameter, placed over the working surface of the specimen.

The vacuum maintained in the container during the time of the experiments was $1 - 2 \times 10^{-5}$ mm Hg. The sputtering time and the ion-current density were adjusted experimentally until sufficiently sharp and contrasting sputtered patches were obtained, suitable for observation in transmitted light.

EXPERIMENTAL RESULTS

Beamed emission of particles at different energies, angles of incidence, and masses of the bombarding ions. We first investigated the sputtered substance deposited on the collector at normal incidence of argon ions on the target ($\alpha = 0$). Figure 2a shows the pattern produced on the collector when the (100) plane of single-crystal copper is bombarded with 8-kev ions. The photographs show that sputtering takes place essentially in four close-packed [110] directions from the (100) face, so that four symmetrical patches are produced on the collector (the distance between centers of the diagonal spots is twice the distance from the sample to the screen). The sputtered patches were elliptical, since the conical beams of sputtered material struck the collector at 45°. Figure 2a does not show the patch corresponding to the [100] direction since the atoms emitted from the target normal to the sputtered surface pass through the hole in the collector and fall on the quartz screen. Pictures similar to Fig. 2a were obtained also with 12, 20, 30, and 50 kev argon ions.

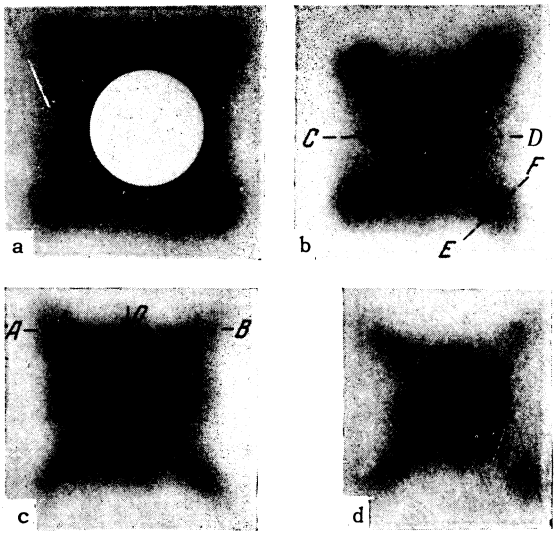

FIG. 2. Pictures of sputtered patches formed on the collectors under the following bombarding conditions:
a) A^+, E = 8 kev, $\alpha = 0°$, j = 0.02 ma/cm², τ = 55 min;
b) A^+, E = 10 kev, $\alpha = 60°$, j = 0.5 ma/cm², τ = 15 min;
c) A^+, E = 40 kev, $\alpha = 60°$, j = 0.6 ma/cm², τ = 20 min;
d) H_2^+, E = 40 kev, $\alpha = 60°$, j = 0.8 ma/cm², τ = 50 min.

The presence of a hole in the center of the collector for the passage of the ion beam did not permit us to investigate the central portion of the deposit at normal ion incidence. This became possible, however, when the investigated crystal plane and the collector plate parallel to it (on which the sputtered substance was deposited) were placed at a certain angle to the ion beam so that the latter passed beyond the collector and struck the specimen at an angle α. The most favorable angle for this purpose was 60° (the distance from the specimen to the collector being 16 mm).

Experiments have shown that when single-crystal copper was sputtered with an inclined beam of argon ions ($\alpha = 60$ and 45°), beamed emission of sputtered particles also takes place. The patterns

of the deposits on the collector are analogous to those obtained at normal incidence ($\alpha = 0$) on the target. In addition, it becomes possible in this case to observe the central maximum corresponding to the [100] direction (Figs. 2b and 2c). The intensity of sputtering increases greatly with increasing angle α. Thus, to obtain a deposit of equal intensity the specimen had to be sputtered by normal-incidence ions several times longer than at $\alpha = 60°$ (other conditions being equal).

The overall view of the deposit changes with increasing ion energy. Starting with an argon-ion energy of ~10 kev ($\alpha = 60°$) the central patch increases sharply, possibly because of the appearance of four new spots, due to sputtering of substance near the [113] and [112] directions. Figure 3 shows curves obtained by measuring the density

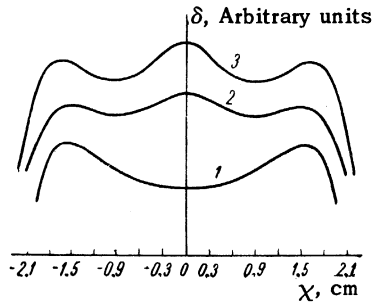

FIG. 3. Curves obtained by photometry of the sputtered deposit along the line AB of Fig. 2c, at the following ion energies: 1 – 1.5 kev, 2 – 10 kev, 3 – 40 kev.

of the deposits in the direction AB (see Fig. 2c) at various ion energies. At a bombarding-ion energy of only 1.5 or 2 kev (curve 1), no maximum was observed in the intensity of single-crystal sputtering near the directions [113] and [112]. On the other hand, sputtering ions with energies 10 kev or more (curves 2 and 3) produced a clearly pronounced maximum in the deposit density, corresponding to directions close to [113] and [112]. This phenomenon can be observed most clearly when the argon ion energy reaches 30–40 kev (see Fig. 2c). At small ion energies (near 10 kev) the increase in sputtering intensity near the [112] and [113] directions could be detected only by photometry of the deposit picture.

We compared the density of the sputtered copper in the individual patches by suitable photometer measurements. The result was that the maximum density P_c in the central patch ([100] direction) relative to the maximum density P_s in the side patches ([110] direction) increases with increasing energy of the bombarding ions. Thus, as the energy is changed from 1.5 to 50 kev, the ratio $p = P_c/P_s$ more than doubles, from 1.5 to 3.75 (Fig. 4). The patches obtained by sputtering single-crystal copper at argon ion energies of 20 and 40 kev and $\alpha = 45°$, and at 6, 10, 20, 30, 38,

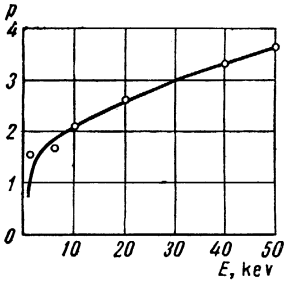

FIG. 4. Ratio of maximum density of deposit P_c in the central patch (corresponding to the [100] direction) to the maximum density of the positive P_s in the side patches (corresponding to the [110] directions) as a function of the energy of bombarding ions, E.

45, and 50 kev and $\alpha = 60°$ were analogous to those shown in Fig. 2.

The preferred emission of particles from the (100) plane of copper in the close-packing directions was observed not only in sputtering with argon ions, when the best ratio of gas-ion mass to metal-atom mass is obtained,[4] but also in sputtering by the much lighter hydrogen ions (Fig. 2d).

Distribution of deposit density in individual patches. It is seen from Fig. 2 that the density in an individual patch of the deposit increases from the center towards the periphery. By photometry of the deposit we can establish the law of distribution of the density of the sputtered substance inside an individual patch. For this purpose we plotted, with a recording microphotometer MF-4, curves of the intensity of light transmitted by the investigated deposit. Examples of such curves (for the central and side patches) are shown in Fig. 5. The horizontal axis represents the ratio $r = x/d$, where x is the distance from the center of the patch to the investigated point, measured in the photometry direction, and d is the distance between the sample and the collector. The vertical axis represents the quantity

$$J(r) = [S_0 - S(r)] / [S_0 - S_{min}]. \quad (1)$$

Here $S(r)$, S_0, and S_{min} are the intensities of light passing through the collector plate at the investigated point, in a section free of deposit, and in the center of the patch (densest section) respectively.

Let us assume now that the density of the central spot ([100] direction) obeys the "cosine law,"[18] i.e., the amount of matter per unit solid angle, emitted from the sputtered surface at an angle φ to the normal from the plane of the sample is proportional to $\cos\varphi$.

Let a plane surface of radius R be sputtered. It can be shown that in this case the distribution of the density of matter deposited on a plane collector parallel to the sputtered surface is given by the relation

$$P(r) = P_0 \frac{1 + r_1^2}{2r_1^2} \left\{ 1 - \frac{1 - r_1^2 + r^2}{\sqrt{1 + 2(r^2 + r_1^2) + (r^2 - r_1^2)^2}} \right\}, \quad (2)$$

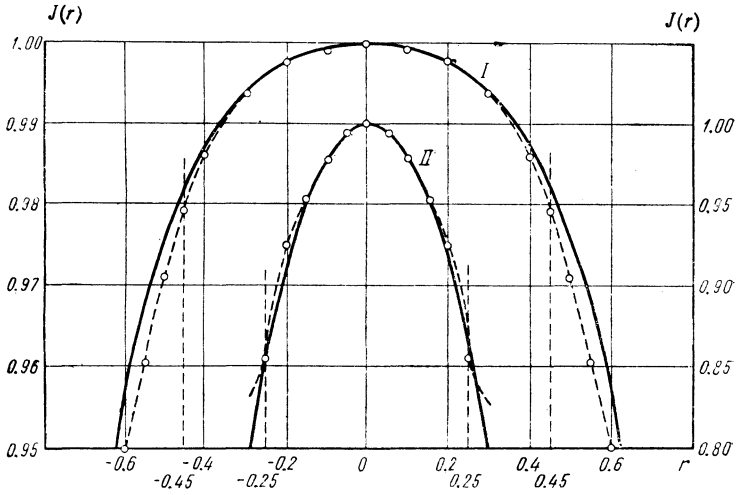

FIG. 5. Comparison of the experimental curves obtained by photometry (solid curves) with the theoretical ones, obtained from the "cosine law" (dotted): I – for the central patch [100] (photometry along the line CD, Fig. 2b), II – for the side patch [110] (photometry along the line EF, Fig. 2b).

where $r_1 = R/d$ and P_0 is the density of the deposit at the center of the patch. However, Eq. (2) is unwieldy, and it is more convenient to use in the calculations the approximate expression

$$P(r) = P_0/(1 + r^2)^2, \qquad (3)$$

which gives results that agree within 2.5% with those obtained with (1), provided $r_1 = 0.2$ and r ranges from 0 to 0.45; these conditions are satisfied in our experiment.

Inasmuch as the density in this section of the deposit is connected with the intensity of light transmitted through it by the logarithmic law

$$\lg(S_0/S(r)) = kP(r) \qquad (4)$$

(k is a proportionality factor), we can readily obtain from (1), (3), and (4) the following relation

$$J(r) = \{1 - 10^{-kP_0(1+r^2)^{-2}}\}/\{1 - 10^{-kP_0}\}, \qquad (5)$$

which should be compared with the experimental curves to verify whether the "cosine law" is satisfied for the density distribution of the sputtered matter near the [100] direction (within the limits of a central patch of $x_c = 7$ mm corresponding to $\varphi \approx 24°$). The quantity kP_0 can be determined graphically if one point of a plot of (5) is made to coincide with the experimental curve.

The points computed from Eq. (5) are plotted in Fig. 5 (curve 1) and coincide within 0.15% with the experimental curve (within a single patch, $r = x_c/d = 0.45$). This means that the density distribution of the sputtered matter in the space near the preferred direction [100] obeys the cosine law with accuracy to 1.5%. The photometer error and the error due to the use of an approximate formula amount to 3%. Equally good agreement was obtained for the central patch ([100] direction) in sputtering of the (100) plane of copper by ions of various energies: 1.5, 40, and 50 kev.

The distribution of the deposit in the side patch in the [110] direction was plotted with a photometer along the line EF of Fig. 2b. In this case the cosine law $P = P_0 \cos\psi$ (where ψ is the angle of deviation from the [110] direction), for comparison with the experimental curves, is expressed approximately by a formula analogous to (5). The comparison of the experimental and computed curves for the side patch ([100] direction) is shown in Fig. 5 (curve II). In this case the cosine law also holds accurate to 1 – 2% within the confines of a single spot ($r = x_s/d = 0.25$), the radius of which is $x_s = 4$ mm, corresponding to $\psi \approx 10°$.

We can thus state that the density of a discrete patch on the picture is distributed in space in accordance with the cosine law.

Microrelief of the surface after sputtering. The anisotropy in the sputtering of a single crystal relative to the different crystallographic directions, at ion energies up to 50 kev, indicates that the mechanism of evaporation of matter from local molten sections of the surface is no longer applicable even for high ion energies. This is also corroborated by the absence of molten sections on the surface of a sample sputtered by fast ions.

Figure 6 shows a relief of the (100) face of copper, formed by sputtering with argon and hydrogen ion beams at 40 kev and an angle of incidence of 60°. We see the tetrahedral pyramidal recesses, characteristic of this plane, similar to those produced by sputtering with slow ions.[11] Oblique incidence of the ions on the (100) plane causes, in addition, the formation of less regular figures, stretched out in the direction of ion incidence (Fig. 6b). The outlines of these figures, as well as of the more detailed relief, are sufficiently clear, without visible traces of melting.

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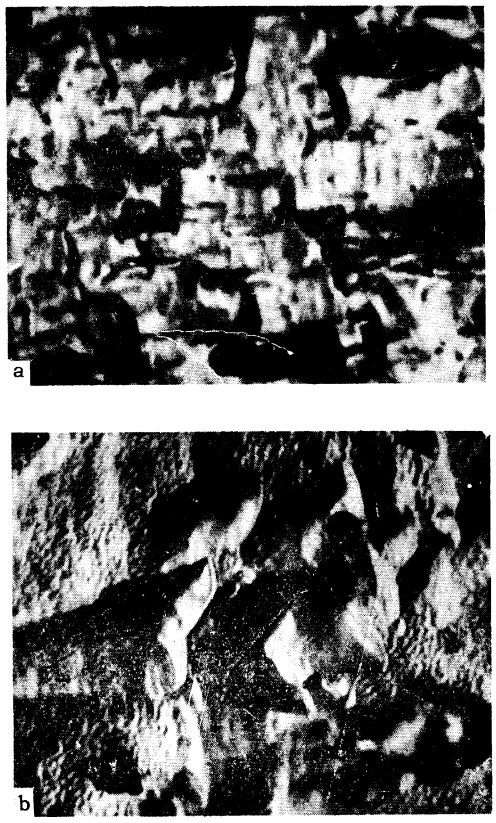

FIG. 6. Relief of the surface of the (100) face of copper after sputtering first with an inclined beam ($\alpha = 60°$) of argon ions at E = 40 kev, j = 0.4 ma/cm², τ = 60 min, and then with hydrogen ions at E = 40 kev, j = 0.8 ma/cm², and τ = 75 min. a – (×1000), b – (×3000).

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190