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The Investigation of Cathode Sputtering in the Near Threshold Region. I

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It is proposed to make use of the method of tracer atoms for the investigation of cathode sputtering in the near threshold region, and for the determination of the magnitude of the threshold energy of sputtering. Results are presented of such an experimental investigation of the sputtering of cobalt by the ions of mercury and argon, traced with its radioactive isotope Co^{60}.

THE investigation of cathode sputtering in the near threshold region, i. e., at small energies V_p of the sputtering ions, bordering on threshold, has always been associated with great experimental difficulties. The problem consists in the determination of both the threshold of sputtering, i. e., the mimimum energy V_0 which the bombarding ion must have for the actual occurence of this process, as well as the dependence of the coefficient of sputtering μ on V_p, for $V_p \geq V_0$[1]. Only a very small part of this problem, consisting in the determination of the quantity V_0 for thermoelectric cathodes covered with active adsorbed films (for example, thoriated tungsten), could be in some measure resolved, thanks to the fact that for a fixed origin of sputtering of this film, the precise method of thermoelectric emission could be used. The determination of the relation $\mu = f(V_p)$ for this case could be realized by an approximate, indirect, and rather unwieldy method[2]. But as regards the vast majority of all other important and interesting materials, and in the first place metals, the straightforward solution of the indicated questions would be practically hopeless in view of the fact that for $V_p \geq V_0$, the magnitude of μ is so small that its immediate determination, for instance by weighing the sputtered deposit, by measuring its transparency, etc., is unrealizable practically. Therefore, in the past, the following method has generally been used: the determination of the relation $\mu = f(V_p)$ was performed in the region of medium and large energies. Then, using the fact that in these regions this relation, according to the experimental data, is generally more or less linear, i. e., $\mu = B(V_p - V_0)$, a linear extrapolation was performed to $\mu = 0$. By such means, untrustworthy in principle, it is of course possible to obtain rough, inaccurate values of the quantity V_0. This deprives us of the opportunity to seek for a correlation between this quantity and other physical characteristics of the metals investigated, i. e., to clarify the nature of the phenomenon of cathode sputtering. Recently, a communication [3] appeared, in which it is suggested that the above mentioned investigations be performed by using the change in the contact potential of a probe in the gaseous discharge, i. e., the shift of the probe characteristics under the influence of a layer of sputtered material deposited on it. This method is essentially a variation of another, already used in the past for this same purpose, in an application to oxidized cathodes[4]. It is readily seen, however, that the probe method is far from universal, because its sensitivity will depend on the difference in the work functions of the electrons in the material being sputtered and in the probe. For the majority of metals, this difference of work functions among them is not great, and therefore there is no basis to expect great success in applying this method. Even the values presented in reference 3 for the quantity V_0 appear so improbably large (~50-70 v.) that one does not feel obliged generally to take them into account.

Thus we see that although a large number of works have been devoted to the investigation of the cathode sputtering of solid bodies[1], still, at the present time, the laws governing this phenomenon inthe near threshold region remain an open question. Meanwhile, at the present time we already have at our disposal a powerful method not previously accessible in the near threshold region – the method of radioactive tracer atoms. We have

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[1] N. Morgulis, Usp. fiz nauk 28, 202 (1946)

[2] N. Morgulis, "Rozporoshuvannia metalichnoi poverkhni pri udarakh pozitivnikh ioniv", Izd-vo AN Ukrainian SSR, 1936, p. 46.

[3] G. Wehner and G. Medicus, Phys. Rev. 89, 339 (1953); G. Wehner, Phys. Rev. 93, 633, 653 (1954); J. Appl. Phys. 25, 270 (1954).

[4] N. Morgulis and Ia. Liubarskii, Fiz. zap. Akad. Nauk USSR 9, 233 (1941); Sb. statei potekhn. fizike, 1947, P. 261.

made use of this method for the solution of the above-stated problem. Some preliminary results obtained by us in this direction are presented below.

The appartus used consisted of a tube with the usual gasotron discharge by an incandescent tungsten cathode in the following atmospheres: 1) mercury vapor, and 2) argon. A small (diam. ~ 3 mm) flat target-probe made of the investigated metal mixed with a radioactive isotope, with a suitable protective ring of common nickel, was introduced into the plasma of the discharge. All leads to the probe and its rear surface were carefully insulated. Placing the corresponding negative potential V_p on the probe with respect to the potential at that point in the plasma, we bombarded it with ions with the current I_p and energy V_p (neglecting the extremely small magnitude of the initial energy of the ions). For the accurate determination of the quantity V_p, the probe characteristic was obtained at each measured point of the curve $\mu = f(V_p)$ (see below). In this way, we determined the potential of the plasma at the point where the target-probe was located. An example of such a probe characteristic in mercury vapor for the important case of a small value of $V_p = (V_z - V_\pi) = (25\text{-}14) = 11$v, is given in Fig. 1, in which is seen its normal character. The correction to the quantity I_p, associated with the secondary emission of mercury ions (in the first case) from the target, is insignificant.

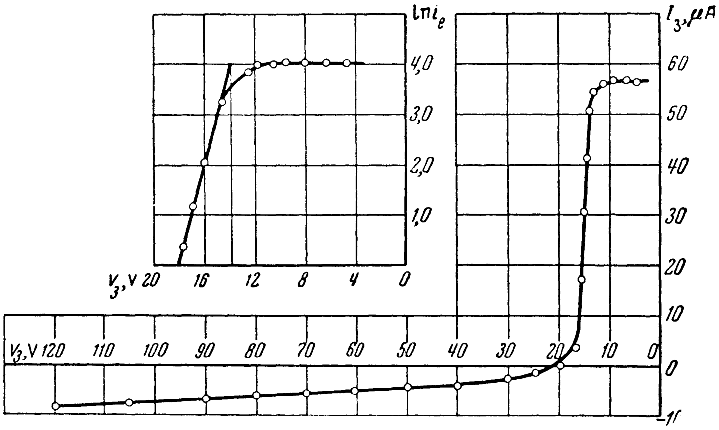

FIG. 1

At a small distance $d \sim 2$ mm in front of the probe, a collector was placed, on which atoms of the metal under investigation, sputtered from the target-probe, were deposited. In order to reduce the reflection of the atoms under investigation[5] from the collector to a minimum, the collector was fabricated either from the same metal, or from a metal with very similar properties (Ni), carefully purified and degreased. Moreover, immediately before the actual experiment, the surface of the collector was cleaned additionally with bombarding ions, i. e., by some sputtering. While the experiment was being carried out, the collector was maintained at a "floating" potential, i. e., was disconnected from the circuit. However, sputtering from the collector of the deposit from the target being studied could be completely ignored because its negative potential with respect to the plasma $V_{\pi\lambda} = (kT_e/e)\ln(u_p/u_e)$ was extremely small since in our experiments the electronic temperature was only ~ 14000°K and did not vary essentially from point to point (Fig. 1). The distance d was generally markedly less than the free path of an atom in the discharge in order to avoid scattering of the latter during their flight from the probe to the collector. However, the use of a discharge in mercury vapor in one of the variations of our investigations might appear not completely successful for the following reason; the surfaces of all cold parts of the apparatus, among them being the probe under investigation, might be partially covered with a thin film of mercury which could to some extent lower the effectiveness of the impact of the bombarding ion, i. e., its sputtering effect[6],

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[5] M. Devienne, J. phys. radium, **13**, 53 (1952); **14**, 257 (1953). N. Morgulis, V. Gavriliuk, and A. Kulik, Dokl. Akad Nauk SSSR **101**, 479 (1955).

[6] L. Koller, Physics **7**, 225 (1936). A. Patiokha, Usp. fiz. nauk **28**, 208 (1946).

and consequently could increase somewhat the value obtained for the quantity V_0. Moreover, this influence might be somewhat different at various energies of the bombarding ions, etc. Since it would be very difficult in our experiments to heat the probe to a high temperature and thereby preclude the presence of mercury films[6,7], similar measurements were also performed (second case) in the atmosphere of an inert gas-argon. However, a comparative investigation of the sputtering of a cold cathode of thoriated tungsten, carried out with mercury vapor as well as with inert gases Ne and A at not too great values of the quantity V_p[8], showed that the error associated with the possible role of the mercury film did not require undue consideration in this case.

Measurements were performed in the following manner. After the discharge was started, the probe characteristic was obtained, the required value of the quantity V_p was established, and the value of I_p was measured. After the target-probe was bombarded for the time t, the tube was opened, the collector with its deposit of active sputtered material was removed, and the number of counts N given by it during the time τ was determined (using an apparatus of type B). The thickness of the layer on the collector was so small that its self-absorption of the radiation could be neglected. The quantity thus obtained, $\nu = N/I_p t\tau = A\mu$, is proportional to the coefficient of sputtering μ atoms/ion, where the coefficient of proportionality A depends on 1) the relative (constant) location of the target and collector, in connection with the flight of the sputtered atoms in the different directions; 2) the specific activity of the initial material of the target; 3) the relative (constant) locations and shapes of the collector with the radioactive deposit and the radiation detector; 4) the counting efficiency. All that was required for the determination of this quantity A was the assumption that the flight of the sputtered particles in the various directions obeyed a cosine law, as is done at the present time[1].

As sputtering targets we used an alloy of nickel with 1% radioactive cobalt^{60} of one activity, and pure nickel, electrolytically coated with a layer of radioactive cobalt^{60} of another activity. The latter emits β-rays of energy 0.32 mev and γ-rays of energies 1.17 and 1.32 mev, has a large specific activity and a long half-life (5.3 years), i.e., is extremely convenient to work with. Its activity is determined in the usual way by γ-radiation.

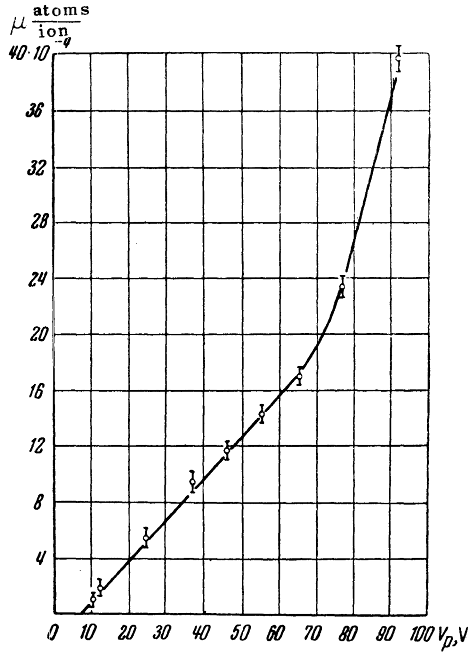

FIG. 2

A typical example of the results obtained by such means for the relation $\mu = f(V_p)$ is shown in Fig. 2 for the case of the alloy Ni + 1% Co^{60}. Here the coefficient of sputtering μ by ions of mercury refers to the common sputtering of Co as well as Ni. Since the physical properties of these two metals are very similar (in particular they have almost the same heat of evaporation, $Q = 4.0$ ev for Ni and 4.1 ev for Co) it is reasonable to expect that the ratio of these components in the sputtered deposit on the collector will be the same as in the sputtered target. A curve of $\nu = f(V_p)$ is shown in Fig. 3 for the case of the sputtering of a layer of pure Co^{60} by ions of mercury. This curve has precisely the same character as the curve in Fig. 2. Finally, Fig. 4 shows the results in the form of the curve of $\mu = f(V_p)$ of the sputtering by ions of argon of the same target made of the alloy Ni + 1% Co^{60}; the method used for these measurements was essentially the same as that used with ions of mercury, described above. On all of the curves in Figs. 2-4, the vertical "antennae" indicate the limits of the statistical error in the measurements of each point; moreover, the points representing very small values of the quantity V_p were redetermined as a control (with the same results).

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[7] S. Sonkin, Phys. Rev. **43**, 788 (1933).

[8] L. Koller, Phys. Rev. **47**, 806 (1935).

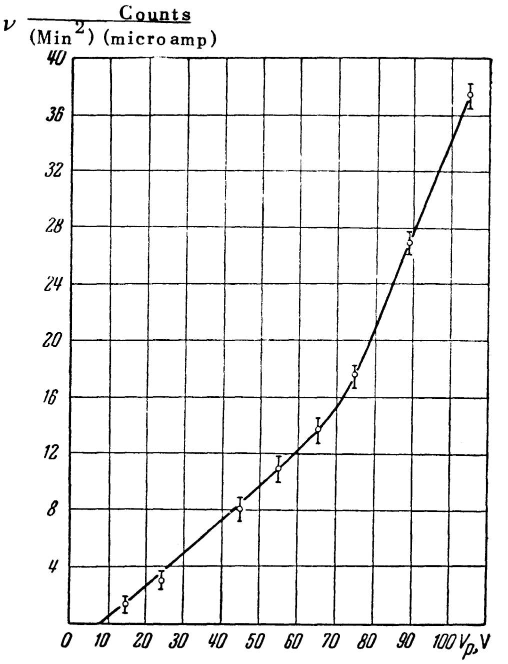

FIG. 3

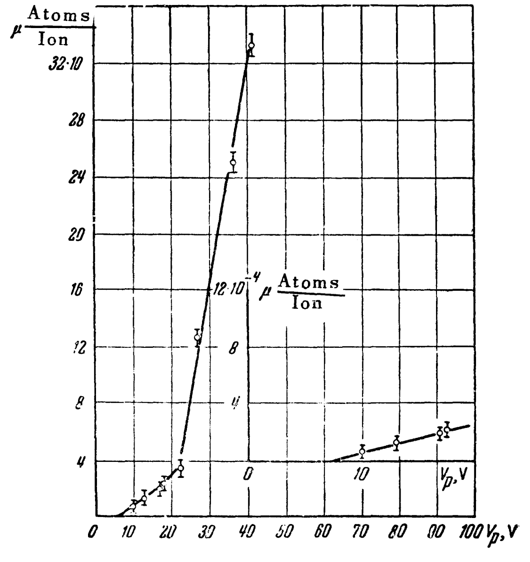

FIG. 4

An examination of the results obtained, presented in Figs. 2-4, leads us to the following conclusions:

1) As the energy of the sputtering ion is diminished and approaches the threshold of sputtering, the basic relation $\mu = f(V_p)$ does not preserve its linear form, but acquires a more concave character, somewhat similar to that obtained by us in reference 2 for the sputtering of thoriated tungsten (see above) Therefore, the method used in the past for the determination of the threshold energy of sputtering V_0, involving a linear extrapolation from the region of large and medium values of V_p to small values is inadmissible, because it may lead to incorrect (too large) values of the quantity V_0. Such an extrapolation to the axis $\mu = 0$, for the determination of the quantity V_0, may be made only in our case, where, by using the method of radioactive tracer atoms, we were able to carry out measurements of sputtering down to energies V_p almost equal to V_0. Our concrete case of the sputtering of mettalic cobalt indicates that $V_0 \simeq 8$ ev.

2) For a preliminary examination of certain peculiarities of sputtering by mercury ions in the near threshold region, we compared the results obtained herein for cobalt with the results obtained in the past for the case of an activated thermocathode made of a coating of thorium on tungsten[8,9], and also, an activated film of an oxidized cathode[4]. The results of such a comparison are presented in the table below. We see here the presence of indications of some regular correlation between the characteristic quantities V_0 and μ of sputtering, and such an important parameter of the sputtered target as the heat of evaporation Q^1.

Sputtered Material	V_0 ev	$\mu \frac{\text{Atoms}}{\text{Ion}}$ for $V_p = 40$ V	Q, eV
Co (Ni)	8	$10 \cdot 10^{-4}$	4.1
Th — W	13	$1 \cdot 10^{-4}$	7.8
Oxidized Cathode	$\leqslant 5.6$	—	3,3

3) A comparison of Fig. 4 for sputtering by ions of argon with the corresponding Fig. 2 for ions of mercury indicates that: 1) the threshold of sputtering has approximately the same value $V_0 \approx 7$-8 ev and 2) the coefficient of sputtering μ in the case of A^+ is considerably larger than in the case of Hg^+; for example, for $V_p = 40$ v we obtain the values $\mu = 30 \times 10^{-4}$, and 10×10^{-4}, respectively. The last relation does not correspond to that which might be expected in this case from the relation between the masses of the sputtering ions and the value of the coefficient of accomodation implied herein; however, owing to the possibility here of a partially masking in-

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[9] N. Morgulis and M. Bernadiner, Z. tekhn. fiz. 5, 1231 (1935).

fluence of a mercury film on the target(see above), to insist on this at the present time is still premature.

4) Also of great interest is the question of the degree of condensation on the surface of the collector under the conditions of a gaseous discharge of comparatively rapid sputtered atoms[10]. Our preliminary experiments indicate that such condensation on the collector of particles of metal might be incomplete, because during the coating of the collector by a layer of lamp-black, the quantity of sputtered atoms condensed on it could become somewhat larger.

Thus the results of the present work show the possiblity of obtaining reliable and interesting data concerning the question of the sputtering of various solid bodies in the previously inaccessible near threshold region by using radioactive tracer methods. Therefore, similar investigations are being expanded by us at present to a whole range of other objects, with the aim of establishing the general laws governing this phenomenon.

Translated by D. Lieberman and M. Mestchersky
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[10] R. Ditchburn, Proc. Cambr. Phil. Soc. 29, 131 (1933); *Proc. Roy. Soc.* (London) A 141, 169 (1933). R. Tr. fiz. otd. Kharkov State University 2, 65, 73 (1950). Zaichik, Tr. fiz. otd. Kharkov State University 2, 65 73 (1950).