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REFLECTION OF SLOW ELECTRONS FROM THE SURFACE OF PURE TUNGSTEN AND FROM TUNGSTEN COVERED WITH THIN FILMS. II

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Studies have been made of the reflection of slow electrons from monocrystalline tungsten and from tungsten crystals on which a layer of tungsten has been deposited by evaporation, under conditions of ultra-high vacuum ($p < 10^{-9}$ mm Hg) to ensure a clean surface. Anomalous changes occur in the reflection coefficients when thin layers of Ba, BaO, and Ba-O are applied to a tungsten single crystal; a possible explanation is discussed. The experimental results are compared with calculations for the case of slow electrons reflected from a potential barrier possessing a maximum. The diffraction of slow electrons from monocrystalline tungsten has been studied, and also the effect of depositing thin films of barium.

A previously published paper[1] contained a report of the results of studies on the reflection of slow electrons from the surface of a polycrystalline tungsten ribbon, and from thin films of barium and oxygen upon the tungsten. The experiments were carried out in sealed-off tubes at total pressures of the order of 10^{-9} mm Hg, and with equivalent pressures of 10^{-10} to 10^{-11} mm Hg in the adsorption regions, thus preventing any distortion of the results by the adsorption of residual gas. The reflection coefficient R for slow electrons from the surface of the tungsten target showed an unusual type of dependence on the energy of the incident primary electrons, V_p; as the energy increased, so did the reflectivity. Meyer[2] has obtained similar results with tungsten. This behavior of the reflection coefficient cannot be explained as an interaction of the electrons with the potential barrier at the metal-vacuum boundary, for any reasonable barrier shape must give a decrease in reflectivity as the energy increases. In these experiments there was still an unknown factor in the polycrystalline nature of the sample, which could conceivably have altered the results, both for the pure tungsten and for the systems with thin films which were studied. For this reason, experiments have been carried out on monocrystalline tungsten targets. The results of these experiments, and of further studies of the effects of thin films on the reflection of slow electrons, are reported in this paper.

MEASUREMENT TECHNIQUES

Reflection coefficient measurements were carried out in sealed-off tubes whose construction was basically the same as that already described[1] (cf. Fig. 1 of Ref. 1). Unlike the ones used previously, these tubes used a glass sphere 35 mm in diameter, coated internally with Aquadag, as the collector for the reflected and secondary electrons. Thanks to this improved collector geometry, the saturation of the elastically-reflected electrons was much sharper in the retarding-potential curves. In the side wall of the collector, at an angle of 45° from the normal to the target, was an opening 1 mm in diameter. A special detector was inserted through this opening when it was desired to study the diffraction of slow electrons. The determination of the contact potential difference between the cathode of the electron gun and the target, and the separation of the current due to elastically reflected electrons from the current of true secondary electrons and inelastically scattered electrons, were carried out as in Ref. 1. As in the earlier work, the utmost attention was paid to obtaining and maintaining a sufficiently high vacuum in the tube (the total pressure in the tubes was of the order of 10^{-9} mm Hg, and was 10^{-10} mm or less in those parts of the apparatus where adsorption took place) using getters and ionization pumps.[3] In every case, measurements were made only after the high vacuum had been attained, and immediately after a prolonged high-temperature bake-out (about 2800° K) of the target by electron bombardment, or after the deposition of the material to be investigated.

REFLECTION OF SLOW ELECTRONS FROM MONOCRYSTALLINE TUNGSTEN

The measurements were carried out on a single crystal of tungsten which was at our disposal, consisting of a plate 7 × 7 × 2 mm in size, cut parallel to the (111) plane and chemically etched to remove the layer of material deformed by the mechanical treatment.

The curves of reflection coefficient R and secondary emission δ as a function of the primary electron energy V_p, measured immediately after heating the tungsten single crystal in high vacuum, are shown in Fig. 1 (curves 1). From a comparison of these curves with the data previously obtained using rolled tungsten ribbon,[1] it could be seen that, except for a small difference in absolute magnitude, the curves were quite similar. Consequently, either the rolled tungsten ribbon consists of a uniformly oriented array of crystallites — a widely accepted view at the present time[4] — and is therefore similar to the monocrystal, or else the polycrystallinity of the target plays no role in increasing the reflectivity at higher electron energies. In any case, the single-crystal experiments have not removed the anomaly in the behavior of the reflection coefficient, namely its increase with increasing electron energy.

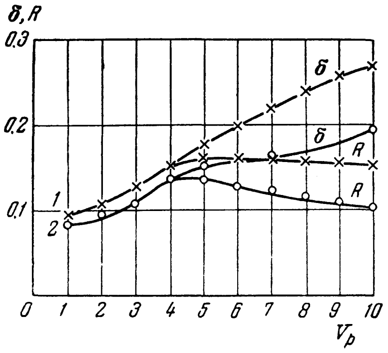

FIG. 1. Variation of R and δ with energy of the primary electrons. 1 – monocrystalline tungsten. 2 – the same covered with a layer of evaporated tungsten.

We have also carried out experiments in which a layer of tungsten was evaporated onto the monocrystalline tungsten target. Such experiments were essential, in view of the fact that in many cases a layer deposited in vacuum has a higher purity than one which has been merely heated, even to the extremely high temperature of the target bake-out. The results obtained with a layer of tungsten evaporated onto the tungsten single crystal are shown in curves 2 of Fig. 1. Deposition of the tungsten has somewhat altered the absolute values of R and δ and has produced a slight falling-off of the curve for R, above $V_p = 5$ ev. Nevertheless, the anomalous behavior in the low-energy region is still there. Hence the increase in reflectivity with increasing energy can hardly be due to contamination of the target surface.

On the basis of MacColl's work[5] and the comments of Herring and Nichols on it (Sec. 28 of Ref. 4) it is possible to attempt a different explanation of these experimental results. MacColl's treatment differed from the earlier work[6,7] by taking into account the periodic nature of the potential within the metal. This led to the prediction of sharp maxima in the curve $R = f(V_p)$, at which R reaches 100%. The effect of inelastic collisions between the electrons inside the metal is to increase the width and greatly decrease the height of these maxima. A distribution in the orientation of individual crystallites, roughness of the sample surface, or a spread in the energies or angles of incidence of the primary electrons, would have the same effect. Consequently it is conceivable that for some metals, because of a particular structure of their internal potential, the reflection coefficient might increase with increasing V_p in a limited region. The idea that the reflection of slow electrons is affected by the potential energy structure of tungsten, in particular, is supported by the correlation between the decreased density of electronic energy states in tungsten and the increased reflection of electrons in the low-energy region. A similar correlation has been observed for copper.[9] The deposition of an active layer which radically alters the profile of the potential at the metal-vacuum boundary can lead to an increased reflection at the surface. This reflection will be of the normal type (a decrease of R with increase of V_p) such as occurs when thin layers of Ba, BaO, or Ba-O (see below) are deposited on tungsten.[1]

REFLECTION OF SLOW ELECTRONS FROM MONOCRYSTALLINE TUNGSTEN COVERED WITH THIN FILMS

To study the potential barrier at the boundary between vacuum and a metal covered with a thin active film, we investigated the reflection of slow electrons from the surface of monocrystalline tungsten covered with a film which reduced the work function. First of all, the experiments with a thin layer of barium atoms[1] were repeated, using the single crystal. This was necessary, since the results previously obtained with a rolled tungsten ribbon coated with barium might have been affected (even if only partially) by the change in the retarding field due to the contact potential of the target after depositing the film. Measurements with a barium film on the tungsten single crystal confirmed our earlier results (see Ref. 1, Fig. 5). Hence the increase in reflectivity as the surface is progressively covered is due to a change in the shape of the potential barrier, and not to a change in electron reflection due to the retarding field.

Layers of the dipolar molecule BaO, which greatly reduce the work function, are of particular interest to the study of cathode electronics. The properties of such systems have been relatively little studied. In this connection, we have experimentally investigated the reflection of slow electrons from a single crystal of tungsten covered with thin layers of BaO molecules. The source of the barium oxide was a spiral of pure platinum covered with barium carbonate, which was heated to convert it to BaO. The amount of barium oxide deposited on the tungsten could be determined from the shift in contact potential, using the data of Gavriliuk.[10] Although it has been stated that the barium oxide deposited from a platinum evaporator is pure,[11] it was considered essential in our particular case to test the evaporation products, and especially to confirm the absence of any appreciable amount of Ba, with a mass spectrometer. These tests* proved that the products of evaporation from our source were in fact pure barium oxide.

Figure 2 shows the results of measuring the coefficient of elastic reflection for the system BaO on monocrystalline tungsten. The shifts in contact potential are indicated in the figure caption. These results depend upon the fraction of the surface covered, which was of the order of one monatomic layer or less. The results obtained with a thick layer of barium oxide will be reported in a separate paper. When barium oxide is deposited, there is a sharp increase in reflectivity in the low-energy region, and the curve $R = f(V_p)$ resumes the normal shape (reflectivity decreasing with increasing energy). The absolute values of R in the low-energy region are considerably larger for the BaO-W system than for the Ba-W system. The results for the latter system are shown as a dashed line in Fig. 2 (see also Fig. 6 of Ref. 1).

In addition to the study of dipolar BaO layers on the tungsten, we also studied the variation in slow electron reflection caused by the evaporation of barium atoms onto a tungsten surface previously covered with a layer of atomic oxygen. The method of producing a layer of atomic oxygen on tungsten is described in Ref. 1. In the present experiment the deposition of oxygen led to an increase of 1.2 ev in the work function, which is somewhat less than the value 1.7 ev obtained in Ref. 1. A comparison of the slow electron reflection from the

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*All the work of testing the BaO source with the mass spectrometer was carried out by G. Ia. Pikus, to whom the author extends his sincere thanks.

BaO-W surfaces with that from the Ba-O-W system shows that, when approximately one atomic layer of material has been deposited, the temperature coefficients are essentially the same and have the same dependence upon energy. From this we conclude that the two systems are largely similar.

It has been suggested several times in the literature that the reduction in work function when a layer of active metal is deposited corresponds with the formation of a potential maximum close to the surface. Some authors[12] consider that the height of this maximum must not exceed the potential level of the vacuum, while others[13] take a different point of view. A study of the reflection of slow electrons may give some evidence to resolve this question; for this purpose, however, we must investigage the interaction of slow electrons with potential barriers containing a maximum. The existing work on this subject[14,15] must be considered unsatisfactory, since the authors make only the crudest assumptions about the shape of the barrier. As a result they obtain an increase in the reflection coefficient at low energies ($R = 1$ at $V_p = 0$). Some calculations have been carried out to find the reflection coefficient from a potential barrier with a maximum lying below the vacuum potential level, formed by superposing the image potential upon the linear potential of a double layer. The results of this calculation predict a relatively small increase in the reflectivity upon going from the pure metal to the metal with an activating layer on its surface, in contradiction to our experimental results. To obtain a satisfactory agreement between theoretical calculations and experiment, it is apparently necessary to assume that when an active metal layer is deposited upon a metal surface, the potential maximum is higher than the vacuum potential level. Such an assumption, and the height of the maximum required to reconcile the results of theory and experiment, is very likely to provide information on the shape of the barrier for this interesting case, which is of such great importance in the study of cathode electronics. It would be very desirable to have such a calculation made. Our preliminary exploratory calculations have shown that, for a potential maximum about 1 ev above the vacuum potential level, it was possible to have a reflection coefficient of the order of 0.2 to 0.4 — i.e., close to the experimental value — for incident electrons of low energy (0 to 1 ev). The difference between the reflection of slow electrons from Ba-W and BaO-W surfaces, on this assumption, implies a difference in the height or width of the potential maximum. In the case of barium oxide the barrier must be considerably wider, and because of the oxygen layer it must also be considerably higher; this would therefore account for the reflection of a greater fraction of the electrons falling on it.

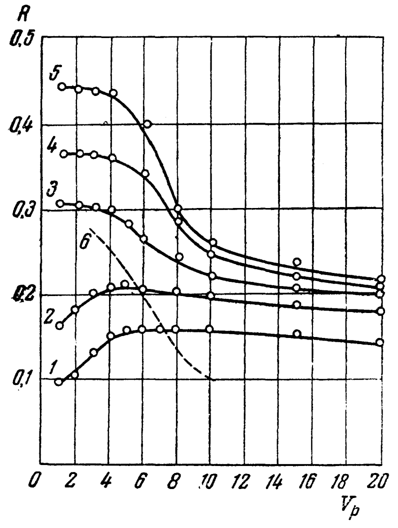

FIG. 2. Variation of R with primary electron energy for BaO-W systems. 1 — $\Delta\varphi = 0$. 2 — $\Delta\varphi = 0.7$ ev. 3 — $\Delta\varphi = 1.7$ ev. 4 — $\Delta\varphi = 2.7$ ev. 5 — $\Delta\varphi = 3.1$ ev. 6 — $\Delta\varphi = 2.4$ ev (Ba-W system).

From our measurements on the systems Ba-W, O-W (Ref. 1), BaO-W and Ba-O-W, it is possible to estimate the effect of slow electron reflection on the size of the constant A in the Richardson-Dushman formula. The reflection of electrons can lead to a reduction in A for such surfaces to 30 — 50% of its theoretical value, 120 amp/cm^2 deg^2. The small values of A quoted in the literature for these systems are apparently connected with other factors, such as the temperature dependence of the work function, non-uniformity of the surface, etc.

DIFFRACTION OF SLOW ELECTRONS FROM MONOCRYSTALLINE TUNGSTEN COVERED WITH A LAYER OF BARIUM

Working with monocrystalline tungsten targets has made it possible for us to observe the diffraction of slow electrons, by using a special collector placed at an angle of 45° to the normal from the target surface. This collector was made positive with respect to the cathode by 3 to 5 volts, so that only the electrons which were elastically scattered from the target would fall on it. The collector was connected to the input of a dc amplifier using a 1E1P tube, giving a sensitivity of 10^{-12} amp/mm. As the potential of the target was varied, the collector current went through a series of maxima (Fig. 3, curve 1).

It is difficult to assign calculated positions to

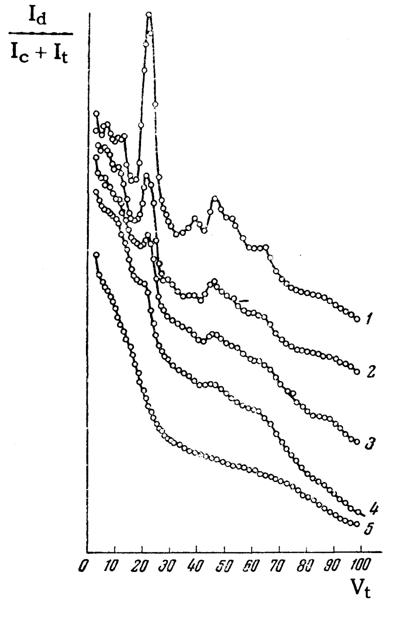

FIG. 3. Variations in diffraction pattern as a layer of Ba is deposited on W. 1—$\Delta\varphi = 0$ (tungsten). 2—$\Delta\varphi = 1.1$ ev. 3—$\Delta\varphi = 1.9$ ev. 4—$\Delta\varphi = 2.7$ ev. 5—$\Delta\varphi = 2.0$ ev (thick layer of Ba).

these maxima, since this requires a knowledge of the order of the reflection and the azimuth angle of the collector, i.e., the angle between a particular crystallographic axis in the target and the projection of the line joining the target to the collector. Furthermore, the problem is complicated by the variation of refractive index with energy for electrons in this velocity range, and by the appearance of a large number of supplementary maxima.[16]

Leaving this calculation for the future, we have in the meantime followed a different route and have studied the effect of foreign material deposits on the intensities of the diffraction maxima. In the experimental and theoretical study of slow electron diffraction it is very important to know how many atomic layers are involved. An answer to this question would make it possible to decide between the volume and surface treatments of this effect.[17] However, the number of papers on this subject is not very large. Lashkarev[18] has estimated the number of effective layers from the width of the diffraction maxima. Farnsworth[19] used for this purpose a foreign material (Ag) deposited on monocrystalline copper and gold, and watched the disappearance of the substrate pattern and the appearance of the maxima for silver (the layer of silver deposited on the gold crystal was obtained in an oriented state). In the present work, however, it would be hard to avoid large errors in determining the amount of material deposited, since the method of control is inaccurate. The rate is determined by weighing the amount deposited over a period of a number of hours.

As a matter of interest, we have estimated the number of layers of Ba required to destroy the substrate diffraction pattern, determining the amount of barium deposited from the contact potential change, using Gavriliuk's measurements.[20] Figure 3 shows the diffraction curves obtained by depositing barium onto a cold single crystal of tungsten; for clarity, the curves are displaced slightly along the vertical axis. Curve 4 shows that a barium layer for which $\Delta\varphi = 2.7$ ev almost completely destroys the diffraction pattern. Here the concentration of barium atoms on the surface is approximately monatomic, i.e., 3 to 4×10^{14} atoms/cm^2. It may be concluded from this that the formation of the diffraction pattern is due almost entirely to the uppermost layer of atoms. This argues in favor of Kalashnikov's theory,[17] which treats the diffraction of slow electrons as a purely surface effect. A study of the position of the sharp diffraction maximum at $V_t = 25$ ev shows that as barium is deposited, the maximum shifts in the direction of lower energy, by an amount equal to the change in contact potential. This corresponds with the effects previously described above for slow electron diffraction. We have studied a thick layer of barium (curve 5), as well as thick layers of gold and germanium, all of which gave smooth curves without any maxima, in agreement with our previous work (see Lashkarev[18], p 93).

To study the structure of thin adsorbed layers of barium on metallic tungsten, we have studied the diffraction of slow electrons from a monocrystalline tungsten surface covered with less than a monatomic layer of barium. Two different methods were used for finding the mean shift in the work function $\Delta\varphi$: one measurement was made after the barium had been evaporated onto a clean, cold substrate; and a second was made after the same value of $\Delta\varphi$ was attained by the gradual evaporation of a thick layer of barium when the substrate was heated. Experiment showed that the diffraction patterns obtained from these two surfaces were different. Invariably the surface obtained by evaporating barium off a heated target gave the more sharply defined maxima. In explaining these results it must be kept in mind that when heat is applied to the uniform layer of barium obtained by deposition onto a cold surface, it may result in the formation of an ordered array of barium atoms, and also in the formation of isolated patches of barium. In the latter case, part of the tungsten surface becomes uncovered, resulting in the appearance of its characteristic diffraction maxima.

In general the work of a surface is determined experimentally by the use of electrons with a phase wavelength of the order of a few tens of angstroms, while at the same time diffraction patterns are measured by using electrons with wavelengths of only a few angstroms. Because of this, surfaces with the same average work function may give different diffraction patterns, depending upon their degree of homogeneity. Hence it must be concluded that the change $\Delta\varphi$ in work function is not by any means a single-valued function of the state of a composite monatomic surface.

In conclusion, I wish to express my thanks to Prof. N. D. Morgulis for suggesting the subject, and for his constant interest in the work.

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MEASUREMENT OF THE SURFACE IMPEDANCE OF SUPERCONDUCTORS AT 9,400 MCS

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The surface impedance of several superconductors has been measured at a frequency of 9400 Mcs. The temperature dependence of the surface impedance of thin films of Sn and Hg has been studied. The penetration depth was investigated for these films and found to be in accord with the critical magnetic field measurements. The effective conductivity of the films in the superconducting state increases as the temperature is reduced. An investigation was also made of the relation between the impedance of thin Sn and Hg films and the magnetic field at $T < T_0$. The dependence of penetration depth on field strength is in satisfactory agreement with the Landau-Ginzburg theory.

1. A knowledge of the complex impedance of a superconductor at frequencies on the order of 10^{10} cps makes it possible to find the dielectric permittivity of the metal:

$$\varepsilon = \varepsilon_0 - c^2/\omega^2\delta_0^2. \qquad (1)$$

Determining the penetration depth of the magnetic field δ_0 from static measurements, it is then possible to estimate the value of ϵ_0 due to the presence of bound electrons in the superconductor.[1,2] On the other hand, if $\epsilon_0 \ll c^2/\omega^2\delta_0^2$, it is possible, using these measurements, to find the penetration