fields, noted above, were computed according to formulas (10) using the values of $g_{||}$ and δ obtained previously.

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54

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The Inelastic Scattering of Electrons from a Copper Oxide Surface

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An electrical differentiation method in a spherical condenser is used to study the inelastic scattering of electrons from a copper oxide surface. Inelastically scattered electrons with discrete energy losses were observed from both monocrystalline and polycrystalline surfaces. The discrete energy losses are equal to the energies required to transfer an electron of the crystal lattice from a filled band to an allowed one. It is shown that the inelastically scattered electrons, with discrete energy losses, are produced by the same mechanism as accounts for the group of genuinely secondary electrons, of discrete energies, which we had discovered previously.

INTRODUCTION

WHEN a group of electrons enters matter, individual electrons undergo changes in their direction of motion, and also lose energy. Little is known about the mechanism by which electrons of low and medium energies (tens and hundreds of ev) lose energy as they pass through a solid body. A study of this mechanism is both interesting for its own sake, and relevant to the understanding of many phenomena, in particular the emission of secondary electrons.

In the course of investigations on the distribution of secondary electrons emitted from metal and metal oxide surfaces, several authors[1-4] have observed inelastically scattered primary electrons with small, discrete energy losses. The primary electrons had energies of approximately 100 volts in these investigations and the inelastically scattered groups were observed at energies close to the primary energy. The fundamental method used in the study of spontaneous energy losses was that of magnetic analysis. This method has high resolving power, but suffers from the disadvantage that only a small fraction of the electrons can be analyzed: namely those traveling in a small solid angle determined by the slit of the analyzer. The retarding field and spherical condenser method proposed by Lukirskii[5] avoids this drawback, but does not have high enough resolving power to investigate the fine structure in the energy distribution of the electrons. As our experiments on the energy distribution of genuinely secondary electrons[7] have shown, the use of electric differentiation in the spherical condenser method[6] increases the resolution considerably and allows one to study the fine structure.

In the present work we examine the fine structure in the energy distribution of secondary electrons at energies close to the primary electron energy. As samples to be investigated, we used copper

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[2] L. J. Hawort, Phys. Rev. 48, 88 (1935); 50 216 (1936)

[3] E. Rudberg, Phys. Rev. 50, 138 (1936); Proc. Roy. Soc. (London) A 127, 111 (1930).

[4] A. R. Shulman and E. I. Miakinin, Dokl. Akad. Nauk SSSR 91, 1075 (1953).

[5] P. I. Lukirskii, Zh. Rus. Fiz-Khim Ob., Ch. Fiz. 57, 463 (1925).

[6] N. B. Gornyi and L. M. Rakhovich, J. Exptl. Theoret. Phys. U.S.S.R. 26, 454 (1954).

[7] N. B. Gornyi, J. Exptl. Theoret. Phys. U.S.S.R. 27, 171 (1954).

covered with copper oxide. In previous work with these samples, the distribution curve had been observed to have fine structure in the neighborhood of the genuinely secondary electrons.

APPARATUS

The principles of the electrical differentiation method for investigating the energy distribution of secondary electrons are described in references 6,7. The circuits described in reference 6 were set up in our laboratory by Rakhovich and are described in detail in his dissertation[8]. Fig. 1 shows a block schematic of the electrical circuit used. The generator frequency was chosen to be 29 cps. The effective value of the alternating component of the potential applied to the collector by the generator was $\Delta V_k = 0.42$ volts. With the preamlifier we could observe currents as low as 10^{-13} amp. In order to measure currents of this size, it was necessary to choose a very narrow band width for the amplifier--no larger than 0.5 cps. This was attained by using a phase sensitive detector.* The phase sensitive detector consists of a synchronous detector and a low frequency filter. As a synchronous detector we used a mechanical relay, which works well at low frequencies. The synchronous detector scheme worked quite satisfactorily. A microammeter at the output gave not only the magnitude of the derivative, but also its sign.

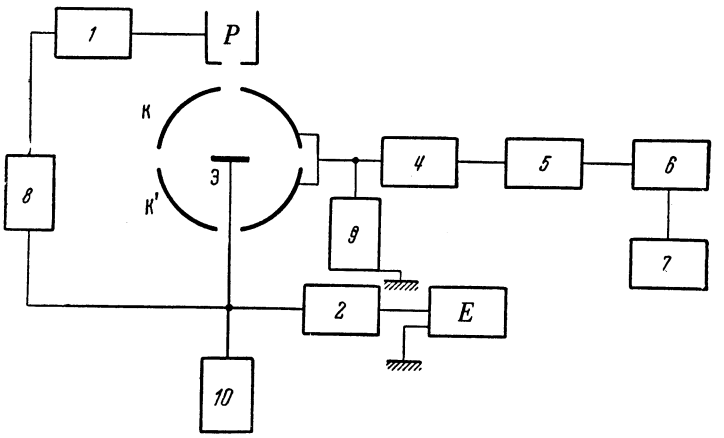

FIG. 1. Block-schematic of the apparatus: *E*-emitter, *P*-electron gun, *K* and *K'*-collectors, *1*-electron gun power supply; *2*-source for D.C. component of the collector potential; *3*-generator of the alternating, sinusoidal potential, *4*-preamplifier, *5*-band pass amplifier, *6*-synchronous filter, *7*-measuring apparatus *8* and *9*-galvanometers, *10*-apparatus for outgassing the emitter.

Our investigations led us to the conclusion that the experimental energy distributions of the secondary electrons, as obtained by the retarding field method, can be distorted in the spherical condenser.[9] This distortion is due to the fact that the number of tertiary electrons knocked out of the collector by secondary ones, subsequently reaching the emitter, depended on the retarding potential. The distortion of the experimental distributions is significant for small values of V_k, and for the values of interest to us--those near the potential V_a accelerating the primary electrons. In order to correct for this distortion, we used the vacuum apparatus shown schematically in Fig. 2. In the present investigations we used the same mono- and polycrystalline samples of copper oxide on copper as used earlier.[7,9] The electrode with the emitter E was cylindrical, and contained a bifilar heating filament inside it. The diameter of the cylindrical electrode was 20 mm, while its length was 17 mm. The construction of the electrode and emitter was similar to that in reference 9. The preparation and outgassing of the apparatus was as in

* We are grateful to A. M. Bonch-Bruevich for pointing out to us the advantages of using a synchronous filter in our apparatus.

[8] L. M. Rakhovich. Thesis, Leningrad Institute for Electrotechnical Communications, 1953.

[9] N. B. Gornyi, J. Exptl. Theoret. Phys. U.S.S.R. 26, 327 (1954).

reference 7. Before the getter was unsoldered and fired, the pressure in the apparatus was less than 10^{-6} mm of mercury at 450°C.

The spherical part of the apparatus was 120 mm in diameter, covered inside with aquadag, and served as the collector. The collector was divided into two hemispheres K and K', insulated from each other and such that the secondary electrons could reach only the upper hemisphere K. The secondary electrons 2 (Fig. 2) incident on the collector K

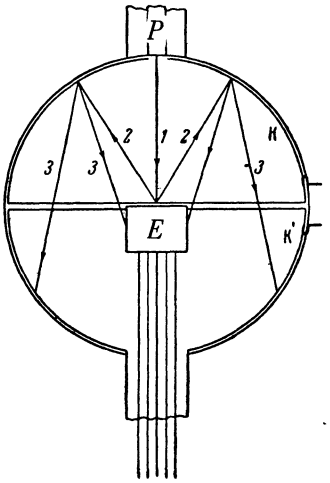

FIG. 2. The vacuum apparatus. E-electrode with the emitter, P-electron gun; K and K'-collectors.

knock out tertiary electrons 3, which can go back to the collector K, to the electrode with emitter E, or to the collector K'.

Our data were taken with both collectors K and K' in the measuring circuit, (arrangement 1), with only one collector K (arrangement 2) and with only the collector K' (arrangement 3). In all runs the collectors K and K' were at the same potential. In order to avoid distortion of the electron trajectories by the earth's magnetic field, the latter was compensated for by two coils of 1 m diameter.

RESULTS OF THE MEASUREMENTS, AND APPLICATION OF CORRECTIONS FOR THE TERTIARY ELECTRONS

Figure 3 shows the curves obtained using a monocrystalline copper oxide surface (111) with V_a=128.5 volts and emitter temperature 420°C. The bias curves 1,2,3 and the distribution curves 4,5,6 were obtained using the first, second and third measuring arrangements respectively. Curves 1--6 are shown on an enlarged scale in the voltage region $V_k \approx V_a$.

We note that curves 1 and 2 fall off for $V_k \approx V_a$; the fall off is due to a decrease in the number of elastically scattered electrons, and occurs over a voltage range of about one volt. Corresponding to this, the width of the elastically scattered peak is also about one volt. This indicates that there is a spread in the energy of the primary electrons, the spread being due to the thermal energy distribution of electrons leaving the cathode filament, or to the non-uniformity of the filament potential. In the following, "primary electron energy" shall mean the energy at the maximum of the peak due to the elastically scattered electrons.

Curve 3 in Fig. 3 shows how the current from the collector K' varied. Since secondary electrons from the emitter cannot reach K', curve 3 evidently gives the tertiary electron current between K and K' as a function of V_k, while curve 6 gives the "speed" relative to changes in V_k with which this current varied. Curve 1 (Fig. 3) becomes negative for values of V_k between 16 and 80 volts. It follows that in this voltage range the number of secondary electrons from the collector (tertiaries) which fall on the emitter is larger than the number of secondary electrons (from the initial scattering of the primaries) falling on the collector K. This is quite possible, since the secondary emission coefficient δ of aquadag depends strongly on the conditions of preparation, as well as on the temperature of the initial heating, and can be larger than unity. Thus, according to the measurements of Titkov [10], aquadag which has been warmed for 6 hours at a temperature of 500°C at a pressure of 2 x 10^{-6} mm mercury has $\delta > 1$ for V_a about 100 to 500 volts. As the (negative) values of V_k are increased above 70-80 volts, curves 1 and 2 (Fig. 3) become positive. This corresponds to a decrease in the number of tertiary electons emitted, the decrease being due to the lower energy of the scattered primary electrons which reach the collector.**

When V_k reaches ~ 126 volts, i. e., when the

** The bias curve 2 (Fig. 3) becomes negative at a smaller V_k (13 volts) than does curve 1 (16 volts), and becomes positive again at a larger V_k (90 volts) than curve 1 (80 volts). This can be explained by the fact that the bias curve 2 is distorted by the large tertiary electron current to the emitter and collector K', while curve 1 is distorted only by the tertiary electron current reaching the emitter.

[10] A.S. Titkov, Zh. Tekhn. Fiz. 23, 1361 (1953).

energy of the elastically scattered primary electrons reaching the collector has decreased to V_a-V_k~2-3 volts, then curves 1 and 2 increase sharply. Correspondingly, the distribution curves (4 and 5) become negative in the same region of V_k, while the "rate-of-change" curve 6 for the current of tertiary electrons reaching the collector K' exhibits a small peak. These data indicate that the tertiary electron current decreases the current of secondary electrons when V_k is less than about 126 volts. When the energy V_a-V_k of the electrons falling on the collector decreases to 2-3 volts, the efflux of tertiary electrons either ceases or becomes negligibly small.

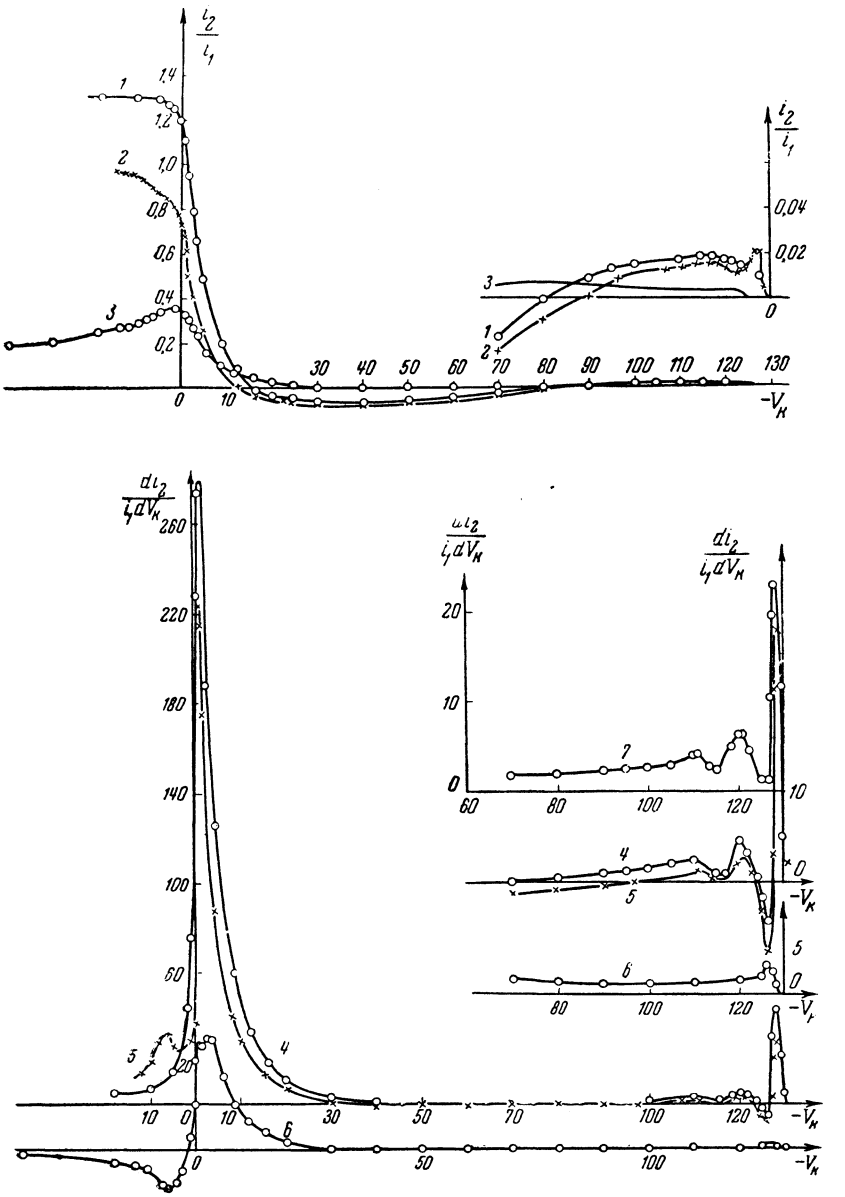

FIG. 3. Curves obtained for a monocrystalline copper oxide surface (111) with V_a=128.5 volts and t=420°C.; *1,2* and *3*-bias curves; *4,5*, and *6*-distribution curves, obtained by electrical differentiation, *7*-corrected distribution curve.

Let us consider how to find the ratio of the tertiary electron currents $I_{k'}$, reaching the collector K', and I_E reaching the emitter E (Fig. 2). Since $I_{k'}$ is measured directly, knowledge of this ratio would allow us to correct for the distortion due to the current I_E. When the space between emitter and collector is field free, i.e., when V_k=0, then both the tertiary and secondary electrons move in straight lines. If we assume that the angular distributions of the secondary electrons leaving the emitter, and of the tertiary electrons leaving the collector K, are given by the cosine law, then from the geometry and a direct measurement of $I_{k'}$ one can calculate the tertiary electron current I_E falling on the electrode E. Calculations showed that in our experimental set up, the current I_E was about $(1/2)\, I_{k'}$.

In the present work, we are particularly interested in the possibility of corrections at large negative values of V_k, close to V_a. Under these conditions, the electric field could bend the trajectory of the tertiary electrons in such a way as to deflect some tertiary electrons from the collector K' to the electrode E, thus increasing I_E. We could try to calculate the increase in I_E by finding the trajectories of tertiary electrons moving in a radial electric field. However, such calculations would be difficult because of the large, but unknown, spread in tertiary electron energy at a definite V_k. In addition, since the electrode with the emitter is cylindrical, the field is not strictly radial and hence the calculations would not be exact. Hence, we looked for another method of correcting the experimental data. It turned out that it was possible to make this correction by obtaining the values of I_k, and I_E, for V_k in the region of interest (close to V_a), directly from the experimental data.

The bias curves 1 and 2 (Fig.3) rise sharply as V_k approaches V_a ($V_k \sim 126$ volts); as mentioned above, this is explained by the decrease(to zero or a negligible value) in tertiary electron emission as the energy of the primary electrons scattered from the emitter and reaching the colector K decreases to 2-3 volts. In making the corrections for tertiary electron current, we assume that the emission of tertiary electrons under the above described conditions ceases completely. The rise in curve 1 is due to the tertiary electrons reaching only the electrode with the emitter E, while the rise in curve 2 is due to the tertiary electrons reaching the electrode E and the collector K'. Clearly, the difference between the maximum ordinate in curve 1 for $V_k \sim V_a$, and the minimal ordinate for $V_k \sim 125$ volts gives I_E, while the corresponding difference in curve 2 gives $I_E + I_{k'}$. Since the ordinates of curves 1 and 2 in this region are small, the value of I_E and $I_{k'}$ found by this method will be quite inaccurate. I_E and $I_{k'}$ can be determined more accurately from curves 4--6 (Fig. 3).

The distortion of the bias curves by the tertiary electron currents corresponds to a distortion in the distribution curves (curves 4 and 5, Fig. 3) caused by changes in the tertiary electron current with changes in V_k. Curve 4 is lowered by the changes in the number of tertiary electrons reaching only the electrode E, while curve 5 is lowered by the changes in the number of tertiary electrons reaching the electrode E and the collector K'. Since curve 6 (Fig. 3) gives the "speed", relative to changes in V_k, of the changes in the number of tertiary electrons reaching only the collector K', then clearly the differences of the ordinates in curves 4 and 5 should equal the value of the ordinate in curve 6 at the corresponding V_k. This is indeed the case. The negative peaks in curves 4 and 5 at $V_k \sim 126$ volts, and the peak in curve 6 at the same voltage, correspond to the above mentioned abrupt decrease in the tertiary electron current as the energy of the primary electrons falling on the collector K decreases to 2--3 volts. Clearly, the area of the negative peak in curve 4 corresponds to the tertiary electron current I_E, the area of the negative peak in curve 5 to the sum of the tertiary electron currents I_E and $I_{k'}$, and the area of the peak in curve 6 corresponds to the current $I_{k'}$, all at the same value of V_k.

From this we can find the ratio of the tertiary electron currents I_E and $I_{k'}$. The distortion introduced by changes in the tertiary electron current I_E for given values of V_k can be found by changing the ordinates in curve 6 in the ratio of the currents I_E and $I_{k'}$. Correcting curve 4 for the "speed" of changes in tertiary electron current reaching the electrode E, and curve 5 for the "speed" of changes in the tertiary electron current reaching the electrode E and collector K', we obtain curve 7, shown in Fig. 3. Curves similar to those shown in Fig. 3 were obtained for other values of V_a and on other samples of copper oxide. Only for low energy electrons (V_a = 21.5 volts) did the results differ from those given above.

Figure 4 shows the curves obtained with the same monocrystalline copper oxide surface (111) for V_a =21.5 volts and emitter temperature t=400°C. The bias curves 1, 2 and 3, and the distribution curves 4, 5 and 6 were obtained with the first, second, and third measuring arrangements respectively. Curves 1 and 2, Fig. 4, differ from the curves 1 and 2, Fig.

3, in the absence of a sharp rise in i_2/i_1, as V_k increases to V_a. Correspondingly, the distribution curves 4 and 5 in this range of V_k do not become negative. One might conclude from these data that in this case (V_a=21.5 volts) there is no decrease in the tertiary electron emission as the energy of the electrons falling on the collector decreases to ~2 volts, and the distribution curves for the secondary electrons are not distorted by changes in the tertiary current. However, the presence of a rather distinct peak at V_k=20 volts in curve 6, which gives the "speed," relative to changes in V_k, of changes in the tertiary current falling on the collector K, indicates an abrupt decrease in the tertiary electron emission in this case. This contradiction can be explained if one supposes that when electrons of energy 21.5 volts are scattered there appears a group of inelastically scattered electrons with energy loss ~2 volts, which compensate for the decrease in the tertiary electron emission. The existence of this group of inelastically scattered electrons is supported by the small peaks in curves 4 and 5 (Fig. 4) at V_k=19 volts.

In Fig. 5 there are shown several corrected curves giving the relative number of electrons falling on the collector as a function of the energy loss in the emitter, obtained at various values of V_a and on various samples of copper oxide.

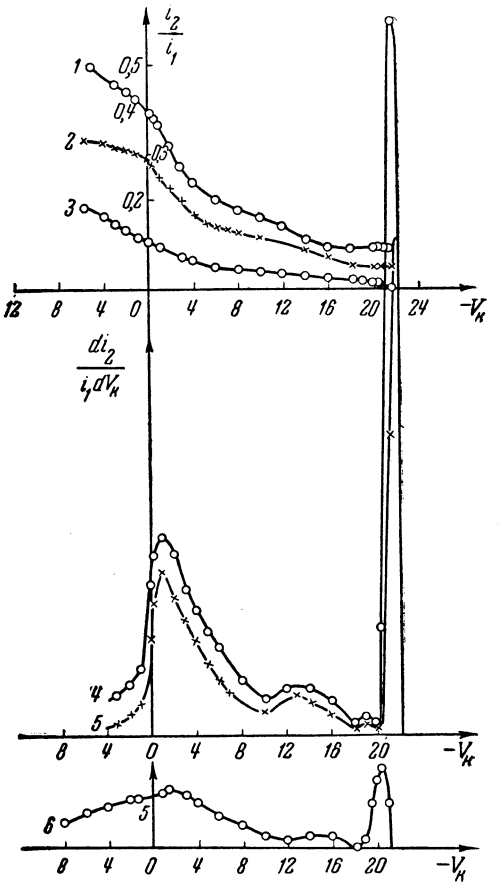

FIG. 4. Curves obtained for a monocrystalline copper oxide surface (111) with V_a=21.5 volts and t=400°C.; *1,2,* and *3*-bias curves; *4,5,* and *6*-distribution curves, obtained by electrical differentiation.

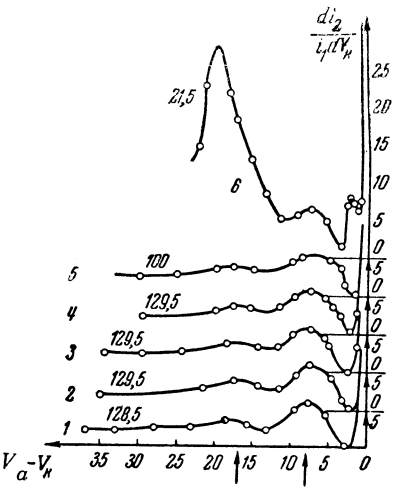

FIG. 5. Curves obtained at 400°C. for the relative number of electrons as a function of their energy loss at various values of V_a, which are shown next to the appropriate curve. *1,2* and *6*-for a monocrystalline (111) surface, *3* and *5*-for a monocrystalline (110) surface, *4*-for a polycrystalline surface. Curve 6 has been magnified by a factor 3.34 relative to other curves. The peak in curve 6 at V_a-V_k=20 volts corresponds to a maximum in the genuinely-secondary electrons .

DISCUSSION OF THE RESULTS

In our study of the energy distribution of genuinely-secondary electrons from a monocrystalline surface of copper oxide[1], we demonstrated the existence of fine structure--i.e., supplementary

maxima. These supplementary maxima are due to transitions which the electrons of the crystal lattice can make from the filled energy band to other allowed bands lying above the Fermi level. Each transition is associated with the absorption of a definite amount of energy from the primary electrons; the possible energy transfers are given by the formula [11,12]

$$W = (h^2/8m\pi^2)(2\pi n/d)^2, \quad (1)$$

where h is Planck's constant, m the electron mass, d the lattice constant, and $n^2 = h^2 + k^2 + l^2$ (h, k, l, being the Miller indices of the appropriate lattice planes).

It is natural to assume that some of the primary electrons can be scattered after losing the energies given above. Then in addition to the elastically scattered primary electrons, there must appear inelastically scattered primary electrons in the distribution curve. In both the corrected curve (Fig. 3, curve 7) and the uncorrected ones (curves 4 and 5), there are 2 distinct peaks, corresponding to primary electrons which have lost energy $V_a - V_k = 8.5$ and 17.5 volts. These values agree well with formula (1), which for $n^2 = 1$ and 2 gives 8.3 and 16.5 volts, respectively. There are also well defined peaks in all the curves giving the relative numbers of electrons as a function of energy loss (Fig. 5). The arrows under the curves give the energy losses computed from formula (1) for $n^2 = 1$ and 2 (8.3 and 16.6 volts).

The energy loss of the primary electrons, as obtained from the peaks, agrees well with the calculated energy loss in all cases except one, namely for $V_k \sim 2$ volts in the distribution curve corresponding to $V_a = 21.5$ volts. An energy of 2 ev is apparently insufficient to take an electron from the full band of copper oxide to the outside. The investigations of Zhuze and Ryvkin on the photoconductivity of copper oxide[13] showed that the maximum in the photoconductivity is at $\lambda = 0.63\,\mu$, which corresponds to a quantum energy of 1.96 ev. It is natural to suppose that the discrete energy loss of 2 ev which we obtained, and which agrees well with the 1.96 ev mentioned above, is the energy required to transfer an electron from the full band to the conduction band.

It should be noted that, while in the investigation of the energy distribution of genuinely-secondary electrons there were supplementary maxima only for monocrystalline surfaces of copper oxide[7], peaks due to the inelastically scattered electrons exist in the distribution curves obtained both with monocrystalline and polycrystalline surfaces. It should also be noted that as the energy V_a of the primary electrons is decreased, then the second peak, corresponding to inelastically scattered primary electrons with energy loss 16.6 volts, decreases also. In the distribution curve obtained with $V_a = 100$ volts this peak is weaker than in the distribution curve at $V_a \sim 130$ volts, while in the distribution curve at $V_a = 21.5$ volts, the peak has disappeared.

The vacuum apparatus used in our earlier work[7] on the distribution of genuinely-secondary electrons from the monocrystalline surface (111) of copper oxide differed from the present one in having the collector covered with powdered platinum; data were there taken on the distribution of secondary electrons over the whole energy range right up to $V_k = V_a$. Figure 6 shows these data. Curves 1 and 2 give the distributions obtained with the electrical differentiation method at $V_a = 75$ and 76 volts respectively, while curve 3 gives the bias curve at $V_a = 75$ volts. The electrical differentiation circuit used in obtaining these curves did not have a synchronous filter, and used a vacuum tube AC voltmeter on the output. As a consequence of this, the negative peaks do not differ from the positive ones. However, the slope of the bias curve (Fig. 6, curve 3) for values of V_k corresponding to the first peak (near the elastically scattered maximum in curves 1 and 2) clearly shows that the first peak must be negative. The second peak must be positive for similar reasons.

The reason for the increase in the bias curve as V_k approaches V_a, and the subsequent negative peak, is the same as applies to the curves considered above (Fig. 3)--the decrease in the tertiary electron emission associated with a decrease in the energy of the elastically scattered primary electrons reaching the collector to 2-3 volts. Thus in the distribution curves for $V_a = 75$ volts, we have only one peak, corresponding to inelastically scattered primary electrons with an energy loss of 8 volts, which agrees well with calculations from formula (1) (8.3 volts).

Reference 4 gives several distribution curves for nickel which were obtained at various values of V_a in the range 20--50 volts by differentiating the bias curves. These distribution curves also exhibit one peak, corresponding to inelastically scattered electrons. The energy at which this peak occurs

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[11] A. Ia. Viatskin, J. Exptl. Theoret. Phys. U.S.S.R. 20, 547 (1950); 20, 557 (1950); 21, 851 (1951).

[12] D. E. Woolridge, Phys. Rev. 56, 562 (1939).

[13] V. P. Zhuze and S. M. Ryvkin, J. Exptl. Theoret. Phys. U.S.S.R. 20, 152 (1950).

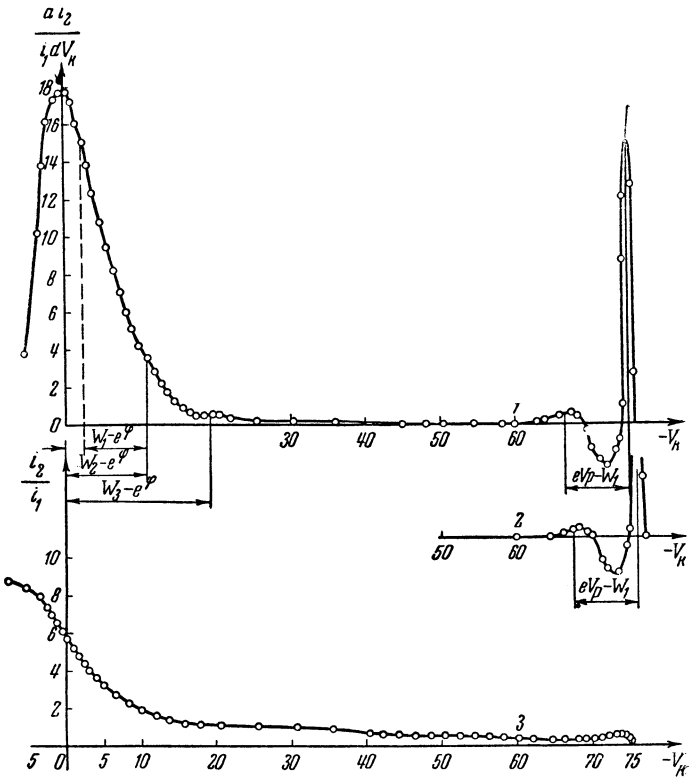

FIG. 6. Curves obtained for a monocrystalline copper oxide (111) surface at t=450 °C. *1* and *2*-distribution curves, obtained at V_a=75 and 76 volts respectively; *3*-bias curve at V_a=75 volts.

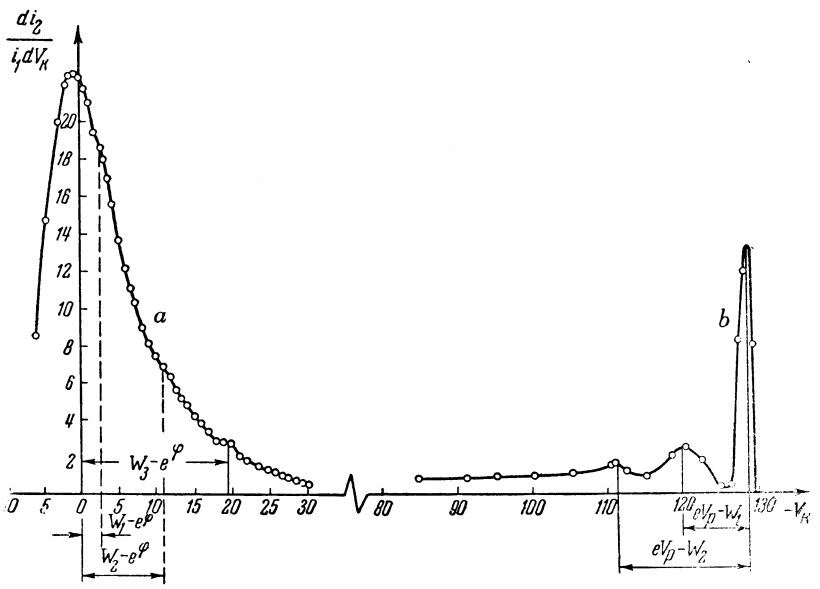

FIG. 7. Distribution curves obtained for a monocrystalline copper oxide (111) surface. Curve *a* obtained with V_a=140 volts and *t*=450 °C, curve *b* obtained with V_a=128.5 volts and *t*=420 °C.

agrees well with the energy calculated from formula (1) for nickel, using $n^2 = 1$ (12.2 volts).

These investigations on inelastically scattered electrons confirm Viatskin's theory of discrete energy losses associated with band transitions[11]. The mechanism by which these energy losses occur consists in the transfer of definite energies from primary electrons moving in the solid body to the electronsof the crystal lattice in this body; these definite energies correspond to transitions from one energy band to another lying above the Fermi level. The electrons from the crystal lattice form, upon emerging from the solid body, the discrete energy groups which we observed when investigating the energy distribution of genuinely --secondary electrons[7].

Under the distribution curves of Figs. 6 and 7, we show the calculated energy losses $ev_a - W_1$ and $ev_a - W_2$ (the latter only on Fig. 7) of inelastically scattered electrons, where W is calculated from formula (1), and also the calculated energies $W_1 - e\varphi$, $W_2 - e\varphi$, and $W_3 - e\varphi$ which electrons of the crystal lattice must have on leaving the emitter as secondary electrons. On curve 1, of Fig. 6, we see that the peak due to inelastically scattered electrons with energy loss $ev_a - W_1$ corresponds to the fine structure maximum in the energy distribution of genuinely-secondary electrons at an energy $W_1 - e\varphi$, while in Fig. 7, in addition, the second inelastically scattered peak, with energy loss $ev_a - W_2$, corresponds to the second maximum in the fine structure with energy $W_2 - e\varphi$.

Thus we see that the inelastically scattered electrons of discrete energy loss which we have discovered in this work, and the groups of discrete energy genuinely-secondary electrons discovered previously,[7] are due to the same mechanism .

CONCLUSIONS

1. The bias curves obtained with the spherical condenser method, and the energy distribution curves obtained by electrical differentiation, are distorted by tertiary electron currents in the voltage region $V_k \sim V_a$.

2. Electrons are scattered both elastically and inelastically, the latter undergoing discrete energy losses.

3. The appearance of inelastically scattered electrons with discrete energy losses is due to electron transitions between the full and allowed bands of the crystal lattice, the necessary energy coming from the primary electrons.

4. The inelastically scattered electrons with discrete energy losses are produced by the same mechanism as accounts for the genuinely-secondary electrons of discrete energies.

In conclusion, I should like to express my gratitude to L. M. Rakhovich for his help in setting up the apparatus and carrying out the experiments. I am also thankful to V. Bolotin, R. Breslav and A. Reitsakas for their help in taking data.

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20