Nachdem die betreffende Flüssigkeit während 4 Std. ruhig gestanden hatte, waren die meisten der darin noch schwebenden Teilchen bereits kleiner als 1 μ. In vielen Fällen dauerte es 10 bis 20 Std. bevor die Flüssigkeit völlig klar war. Aus der Tatsache, dass eine so lange Zeit zur Klärung erforderlich war, lässt sich schliessen, dass jedenfalls Teilchen vorhanden waren, deren Dimension weniger als 1 μ betrug.

13. In den Versuchen mit diesem Material, welche wir in unserer nächsten Mitteilung zu beschreiben beabsichtigen, handelt es sich demnach um Teilchen, welche nach der Gleichung von WI. OSTWALD–FREUNDLICH eine messbar grössere Löslichkeit aufweisen müssten, und dementsprechend müssten auch deren gesättigten Lösungen ein messbar grösseres Leitvermögen besitzen.

VAN 'T HOFF-*Laboratorium.*

Utrecht, Dez. 1939.

Physics. — *Cathode sputtering in a magnetic field.* By F. M. PENNING and J. H. A. MOUBIS. (Natuurkundig Laboratorium der N.V. Philips' Gloeilampenfabrieken, Eindhoven, Holland.) (Communicated by Prof. G. HOLST.)

(Communicated at the meeting of December 30, 1939.)

§ 1. *Sputtering coefficient and sputtering efficiency according to earlier experiments.*

The elementary process in the sputtering of a metal surface is the liberation of one or more metal atoms [1]) by the collision of one ion against the surface. For this process a probability ϑ may be introduced, which we shall call the sputtering coefficient, giving the mean number of atoms liberated per ion from the surface and not again diffusing back to it. ϑ proves to be a function of the nature and the energy V_p of the ion, the nature and surface condition of the target metal, the gas density p_0 (gas pressure reduced to 0° C) and the geometrical form of the apparatus used. When the target metal is an infinite plate at a distance d of another infinite plate (collector), the two latter variables may be combined into one, viz. the product $p_0 d$. In this case the sputtering shows analogy to the evaporation of atoms from a plate through a gas atmosphere to another plate [2]); the fraction f_1 of the evaporated atoms reaching the collector plate being, under certain approximations [3]):

$$f_1 = \frac{2{,}3\,\lambda_1}{p_0\,d + 2{,}3\,\lambda_1} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

(λ_1 = mean free path of the evaporated atoms in a gas of 1 mm pressure). A similar value of f_1 should be expected in the case of sputtering; calling ϑ_0 the total number of metal atoms liberated by one ion, inclusive those returning back again, then:

$$\vartheta = f_1\,\vartheta_0 .$$

---

1) From oxidized metal surfaces also ions may be liberated (comp. § 5). The electrochemical sputtering, where the gas ions react with the metal is left out of account here; comp. A. GÜNTHERSCHULZE, Z. Phys. **36**, 563 (1926).

2) A. GÜNTHERSCHULZE, Z. Phys. **38**, 575 (1926) (sputtering of a large number of metals in a hydrogen glow discharge).

3) W. DE GROOT, Physica **8**, 23 (1928); H. POSE, Z. Phys. **52**, 428 (1928); H. BARTELS, Z. Phys. **55**, 507 (1929). The coefficient 2,3 holds for the case that the evaporating atoms have the same mean energy and the same mass as the gasatoms, the persistence being taken into account (without persistence the coefficient for the three-dimensional case is 4/3).

The experiments in the literature from which ϑ may be derived consist of two groups, one with $\lambda_1 \gg p_0 d$ and $f_1 = 1$, giving directly values for ϑ_0, the other with $\lambda_1 \ll p_0 d$, giving only values for ϑ, whilst ϑ_0 has to be calculated with Eqs. (1) and (2) where, however, the value of λ_1 is very uncertain (comp. § 4).

For the experiments of this first group up to now a discharge tube with at least 3 electrodes was necessary, the ions being formed in an auxiliary discharge (usually with a heated filament as cathode) and being accelerated to a target electrode serving at the same time as collector; the energy V_p of the impinging ions (in volts) here is equal to the potential difference between the place of origin and the target electrode.

Most experiments on cathode sputtering belong to the second group, where the target electrode is also the cathode of a glow discharge. In the case of parallel plates, considered here, the collector often at the same time serves as anode for the glow discharge. In this kind of experiments the distance d is at least of the same order of magnitude as the thickness of the Crookes dark space d_c; as usually $\lambda_1 \ll p_0 d_c$ this involves $\lambda_1 \ll p_0 d$. Here the ions arrive at the cathode with an energy V_p which is smaller than the cathode fall V_c for two reasons: firstly, part of the ions are formed within the Crookes dark space and so pass only a fraction of the cathode fall V_c, secondly the ions lose energy in the Crookes dark space in collisions with gas atoms. Both effects we take into account [4] by introducing a factor f_2 (< 1). Calling $\vartheta_c(V_c)$ the number of atoms liberated from the cathode and not returning to it for a cathode fall V_c, we have in connection with the preceding Eqs.:

$$\vartheta_c(V_c) = f_2\,\vartheta(V_c) = f_1 f_2\,\vartheta_0(V_c) \quad . \quad . \quad . \quad . \quad . \quad . \qquad (2)$$

For the determination of ϑ the pos. ion current i_p to the target electrode should be known. Taking into account that one ion liberates γ electrons from the cathode, the actually measured current i is given by:

$$i = i_p (1 + \gamma),$$

As γ usually is not known, the quantity directly resulting from the experiment is $\vartheta/(1+\gamma)$ or $\vartheta_c/(1+\gamma)$.

In Fig. 1 a few results for $\vartheta(500)/(1+\gamma)$ obtained according to both methods are summarised. The dependence on $p_0 d$ at low pressures should be considered as only approximately right as the collector was not a plate parallel to the cathode. The values for tungsten given by the Research

[4]) At the values of $V_c \geq 500$ V the percentage of the ions formed within the Crookes dark space is usually small (comp. a forthcoming article of M. J. Druyvesteyn and F. M. Penning in the Rev. Mod. Phys.). Moreover in the factor f_2 the number of atoms liberated by fast gas atoms originated from charge transfer has to be included. For low values of p_0 and high values of V_c, due to these circumstances the factor f_2 will be not much lower than 1.

Staff G.E.C. [5]) are only relative ones as the velocity of the ions is not known exactly. They were reduced to absolute values by equalling the

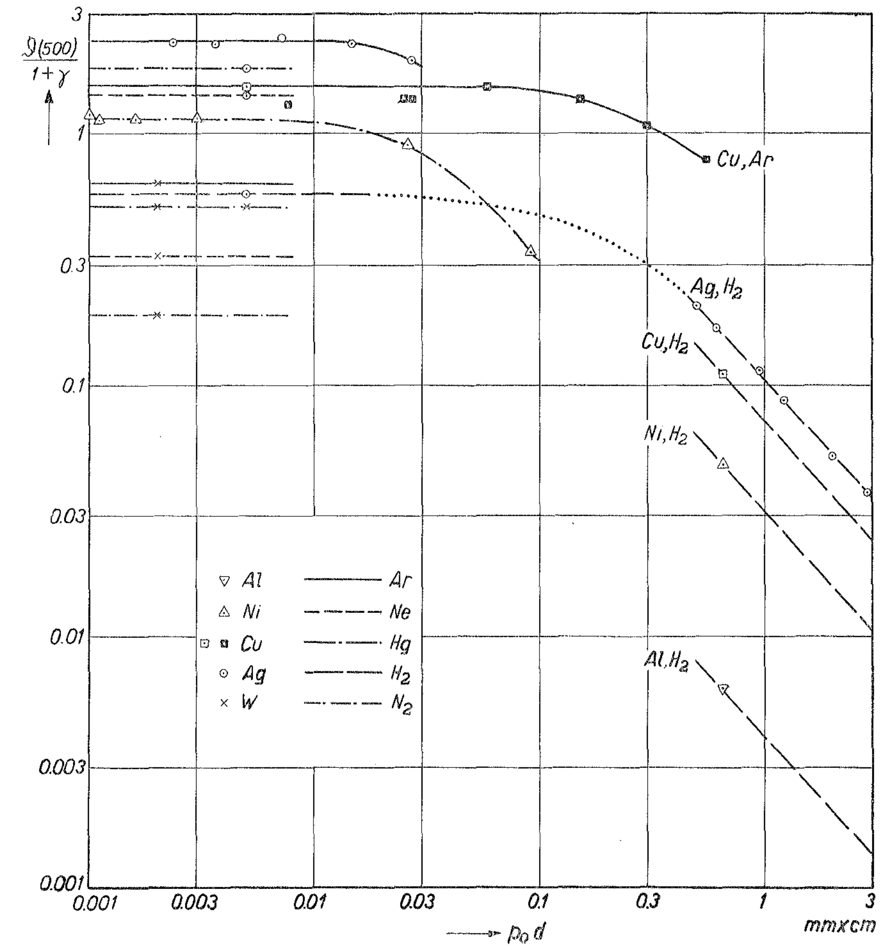

Fig. 1. Values of $\vartheta(500)$, the number of atoms liberated from and not returning to the cathode, according to different authors, reduced to 500 V. Solid points: some of the results obtained in the present article.

result for Hg^+ to that given by Meyer and Güntherschulze. The other points are due to Güntherschulze c.s. [2, 6, 7]) with exception of the solid squares which give some of the results obtained in the present article (cylindrical electrode arrangement). The measurements, made at higher voltages than 500 V, were reduced to 500 V with a reduction factor derived

[5]) Research Staff G.E.C. Phil. Mag. **45**, 98 (1923) (first method; W in H_2, He, N_2, Ne, Hg, Ar).

[6]) A. Güntherschulze and K. Meyer, Z. Phys. **62**, 607 (1930) (first method; Ag in He, H_2, Ne, N_2, Ar; Cu in Ar).

[7]) K. Meyer and A. Güntherschulze, Z. Phys. **71**, 279 (1931) (first method; 16 metals in Hg vapour).

from GÜNTHERSCHULZE's results. The values for Cu, Ni and Al at $p_0 d \geqq 0.5$ were calculated in the supposition that here $d = d_c$.

At low values of $p_0 d$ the independence of ϑ from $p_0 d$ shows that $\vartheta = \vartheta_0$; at the high values of $p_0 d$ (second method) f_2 probably [4]) may be taken ~ 1, so that the actually measured quantity ϑ_c is not much lower than ϑ.

Roughly spoken ϑ_0 is, for $V_p > 500$ V, proportional to V_p, so that for the sputtering of one atom an energy of $V_p/\vartheta_0\,(V_p)$ (volts) is required. The minimum energy necessary for the liberation of one atom is the evaporation energy W. So we may define a sputtering efficiency ε as:

$$\varepsilon = \frac{W}{V_p/\vartheta_0\,(V_p)} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

According to GÜNTHERSCHULZE c.s. [6, 7]) this efficiency is only of the order of a percent.

As to the dependence of ϑ_0 on the nature of the ion and the sputtered metal up to now no adequate theory has been given which covers the whole field, although several attempts in this direction have been made and for a small number of ions and atoms an agreement could be stated [8]).

§ 2. *Cathode sputtering in a magnetic field.*

As was remarked already in § 1 the sputtering in a glow discharge could not be performed at such low pressures that $\lambda_1 \rangle\rangle\, p_0 d$ and therefore ϑ_0 could be obtained from these experiments only by a rather strong extrapolation. Moreover, as the current density at constant cathode fall is proportional to p_0^2, it is so small at low gas pressures that the sputtering times become very long.

These inconveniencies may be eliminated by the application of a properly designed magnetic field which increases the lengths of the electron paths and consequently the amount of ionisation. As was discussed elsewhere [9]) this influence is particularly large when the cathode is a cylinder with end plates perpendicular to the axis in a coaxial magnetic field. In this arrangement the current density may be one or more orders of magnitude larger than without magnetic field, so that the sputtering time is reduced very considerably. This is important for the technical coating of surfaces with metal and, as a matter of fact, the experiments of which some results are given below, were undertaken for this purpose.

Apart from this advantage, the sputtering in a magnetic field may be performed in the pressure region where $\lambda_1 \rangle\rangle\, p_0 d$, giving direct results for the sputtering coefficient ϑ_0 and so bridging the gap between the two

[8]) A. VON HIPPEL, Ann. d. Phys. **80**, 672 (1926); **81**, 1043 (1926); here also the older literature; E. BLECHSCHMIDT, Ann. d. Phys. **81**, 999 (1926); E. BLECHSCHMIDT and A. VON HIPPEL, Ann. d. Phys. **86**, 1006 (1928); G. HOLST, Physica **4**, 68 (1924); R. SEELIGER and K. SOMMERMEYER, Z. Phys. **93**, 692 (1935); K. SOMMERMEYER, Ann. d. Phys. **25**, 481 (1936).

[9]) F. M. PENNING, Physica **3**, 873 (1936).

groups of experiments, mentioned in § 1 (solid squares Fig. 1). Therefore in the course of the experiments mentioned also a few measurements were made to determine the coefficient ϑ_0. Although the accuracy of the quantitative results is not very great and might be improved by a better design of the apparatus, we believe that this new method is worth to be described and that the preliminary results obtained give a welcome complement to those obtained with quite different experimental arrangements.

Another advantage of this method is that the three variables, current density (j), voltage (V) and gas density (p_0) may be changed independently of each other by the use of the magnetic field, whilst in the ordinary glow discharge one of them is determined by the two others.

§ 3. *Experimental arrangement.*

One of the tubes used is shown in Fig. 2. It consists of 2 parts, the fixed part II with a ground glass edge, and a part I with a ground flange which could be taken off for a new filling. Between the two grindings a rubber ring was inserted; the atmospheric pressure proved to be large enough to ensure a vacuum of 10^{-5} mm at the running pump.

The cathode C is a water cooled hollow copper cylinder with two flat end plates, soldered to a chrome iron one, which in its turn is sealed to a cylindrical glass tube. The metal-glass joint is protected from the discharge by mica foil and a silica tube. For the water cooling a hollow brass tube is screwed into the chrome iron part of the cathode.

For the sputtering of Ni and Ag the copper tube was electrolitically covered with one of these metals; the sputtering of Al was performed with a hollow Al-cylinder as a cathode.

The amount of sputtered material was determined by weighing small mica plates M before and after the experiment. The holder for these mica plates can be moved magnetically, with the aid of the iron piece F, within a hollow brass case B; through a window of this case successively all the plates M in the holder are exposed to the sputtering. By special measurements it was stated that the amount of sputtering did not change much along the length of the cathode cylinder.

The tube was placed within a coil, giving a magnetic field H parallel to the cathode cylinder.

The experiments were made in flowing argon, the arrangement being shown schematically in Fig. 3.

Immediately after the sputtering tube another gas discharge tube in a magnetic field ("magnetic manometer") was provided for the permanent survey of the gas pressure [10]) and the spectroscopic controll of the gas purity. The value of the argon pressure was measured on a Mc LEOD further on in the tube to the pump. According to the resistance of the

[10]) F. M. PENNING, Physica **4**, 71 (1937).

tubings this presure had to be multiplied with a certain factor in order to obtain the pressure of the flowing gas in the sputtering tube. Experimentally this factor was determined to about 1.7.

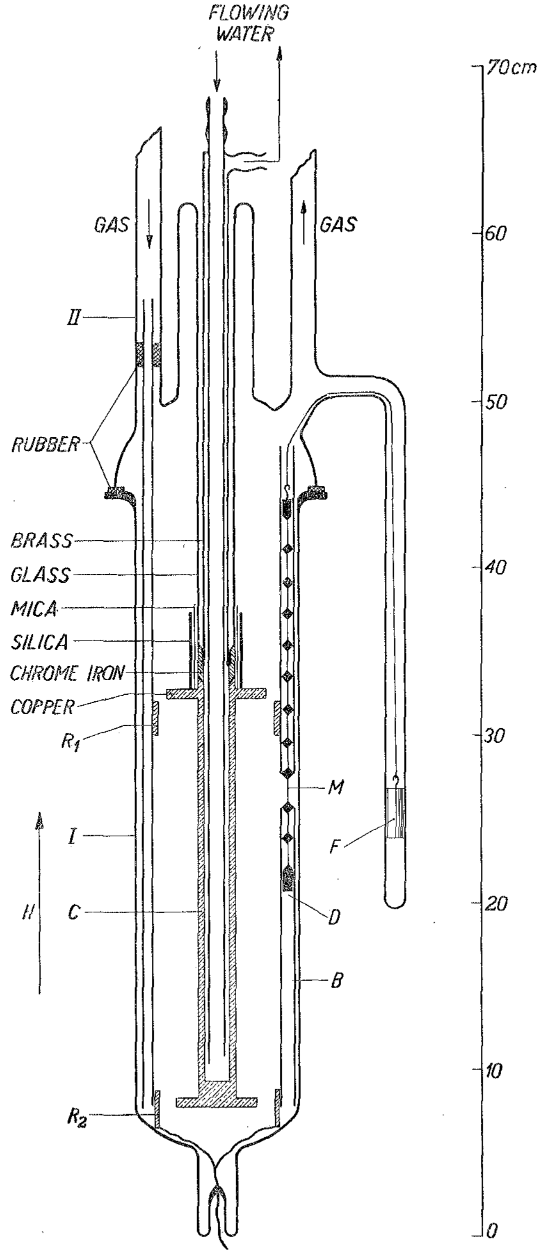

Fig. 2. Discharge tube for the sputtering of Cu.

§ 4. *Results for Cu.*

In Fig. 4 a few current voltage characteristics are given, showing the large influence of the magnetic field H. For $H = 0$ the current below

2000 V anode voltage was zero. With magnetic field the energy required to cover the mica with a layer of 0.001 mm was about 5 kilowattmin, which

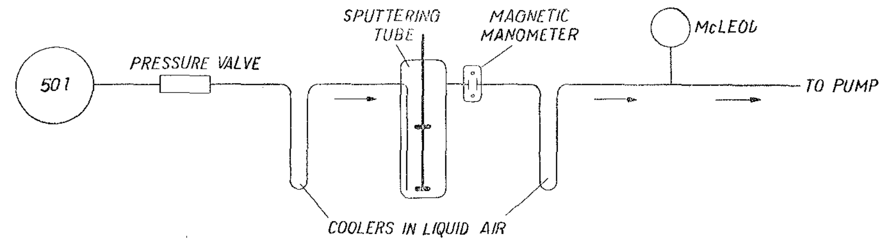

Fig. 3. Schematic survey of the experimental arrangement.

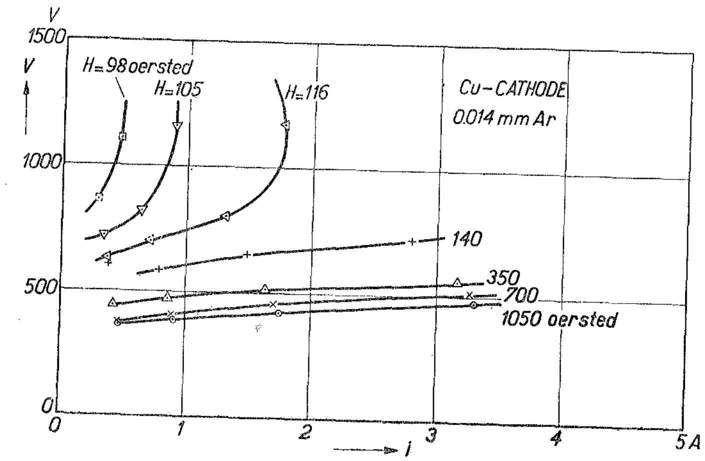

Fig. 4. Characteristics of the discharge in 0.014 mm Ar with Cu-cathode. Parameter: magnetic field strength H.

gives e.g. a time of only $3\frac{1}{2}$ min. at 500 V, 3 A. This time is very short as compared with that in the usual sputtering arrangements. With the tube mentioned in the end of § 4 it could be reduced to $\frac{1}{2}$ min.

The value of $\vartheta/(1+\gamma)$ was determined as a function of i, V and p (see the end of § 2), changing one of these quantities and holding constant the other two by a proper value of the magnetic field. The following series were measured:

500 V ;	1.65 A ;	p variable
1.7 A ;	0.022 mm Ar ;	V variable
0.022 mm Ar ;	500 V ;	i variable

The results for $\vartheta/(1+\gamma)$ as $f(p)$ are given in Fig. 5; the two series measured showing the same behaviour but a constant difference in the value of ϑ, perhaps due to a somewhat different distance from the cathode to the mica plate. Without magnetic field, with a current of 0.4 A (0.35 mm Ar), a value for $\vartheta/(1+\gamma)$ of only 0.15 could be obtained.

In Fig. 6, which gives $\vartheta/(1+\gamma)$ as $f(V_c)$ also the points have been

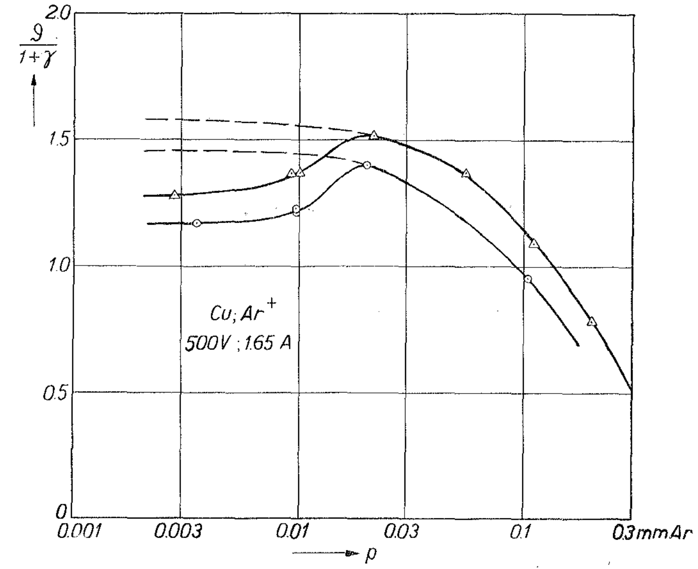

Fig. 5. Values of $\vartheta/(1+\gamma)$ as a function of the pressure for Cu and Ar+. ϑ is the number of Cu-atoms reaching the collector per Ar+-ion reaching the cathode, γ is the number of electrons liberated from the cathode per pos. ion.

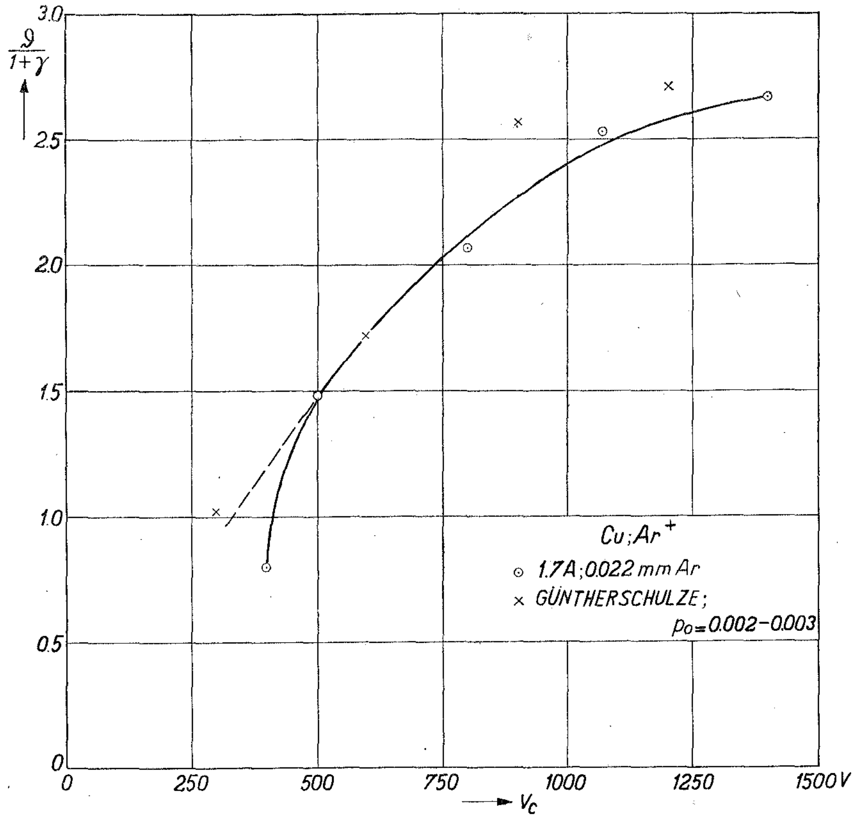

Fig. 6. Values of $\vartheta/(1+\gamma)$ as a function of the voltage V_c between cathode and anode.

plotted obtained by GÜNTHERSCHULZE and MEYER [6]) at much lower gas densities [11]) and a cathode temperature of 800° K. (first method of § 1). The agreement is good, the more so as our point for 400 V, should be discarded (see below). Fig. 7 shows the dependence of $\vartheta/(1+\gamma)$ on i.

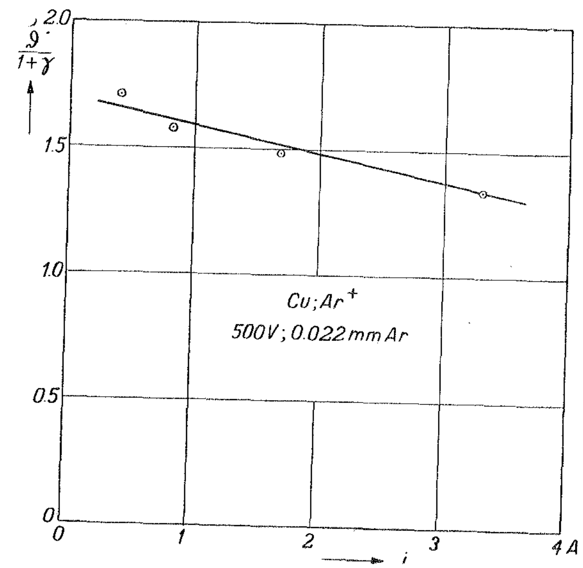

Fig. 7. Values of $\vartheta/(1+\gamma)$ as a function of the discharge current i.

In order to enable a rough survey of the experimental conditions in table I a few quantities, important for the discharge are given:

λ_e = mean free path of the electrons [12]) with energy $\frac{1}{2}V_c$.

λ_p = mean free path of the ions [13]) with energy $\frac{1}{2}V_c$.

λ = mean free path of the Ar-atoms [14]) at the pressure p.

d_c = thickness of the Crookes dark space, calculated after the space charge formel of LANGMUIR [15]) $d_c^2 = 5.462 \cdot 10^{-8} M^{-\frac{1}{2}} V^{\frac{3}{2}}/j$ (M = atomic weight of the ion, V = potential difference in volts, j current density in A/cm 2).

r = $3.37 \sqrt{V}/H$, radius of the circle described by an electron of energy V in the magnetic field H.

l = $11.38\, E/H^2$, maximum distance of the cathode, reached by an electron starting from the cathode with zero velocity in an homogeneous electric field [16]) of strength $E = V_c/d_c$.

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11) In order to reduce the amount in gram/ampère as given by G. and M. to ϑ it has to be multipled with $26.8/M$ (M = atomic weight of the sputtered metal).

12) P. LENARD, Ann. d. Phys. **12**, 715 (1903).

13) F. WOLF, Ann. d. Phys. **29**, 33 (1937); A. ROSTAGNI, Nuovo Cim. **15**, 117 (1938).

14) LANDOLT–BÖRNSTEIN, Physikalisch-Chemische Tabellen I (1923), p. 119.

15) I. LANGMUIR, Rev. Mod. Phys. **3**, 191 (1931).

16) F. M. PENNING, Ned. T. Natuurk. **3**, 141 (1936).

TABLE I.

Mean values of several quantities in the experiments of Figs. 5—7 (Cu and Ar+)

p (mm)	V (volt)	i (ampère)	H (oersted)	$\frac{\vartheta}{1+\gamma}$	$\frac{\vartheta(500)}{1+\gamma}$	γ	ϑ (500)	λ_e (cm)	λ_p (cm)	λ (cm)	d_c (cm)	r (cm)	l (cm)
0.003	500	1.65	850	1.23	1.23	0.10	1.35	31	2.6	1.6	0.098	0.089	0.081
0.01			430	1.30	1.30		1.43	9.3	0.77	0.48		0.17	0.31
0.03			204	1.43	1.43		1.57	3.1	0.26	0.16		0.35	1.4
0.10			127	1.06	1.06		1.16	0.93	0.077	0.048		0.59	3.6
0.30			83	0.37	0.37		0.41	0.31	0.026	0.016		0.91	8.2
0.022	400	1.65	1050	0.90	1.12	0.06	1.19	3.6	0.34	0.22	0.083	0.064	0.050
	500		251	1.48	1.48	0.10	1.65	4.2	0.35		0.098	0.30	0.92
	750		128	2.02	1.35	0.18	1.60	6.0	0.38		0.13	0.72	3.9
	1000		115	2.39	1.19	0.27	1.52	6.8	0.39		0.16	0.93	5.2
	1250		114	2.62	1.05	0.36	1.43	7.9	0.40		0.20	1.0	5.6
	1400		114	2.74	0.98	0.41	1.38	8.5	0.41		0.21	1.1	5.7
0.022	500	0.5	147	1.64	1.64	0.10	1.80	4.2	0.35	0.22	0.17	0.51	1.5
		1	178	1.59	1.59		1.74				0.12	0.42	1.4
		2	297	1.48	1.48		1.62				0.087	0.25	0.74
		3.5	675	1.32	1.32		1.46				0.067	0.11	0.19

γ = number of electrons liberated by one Ar+ ion of energy V_c from a Cu cathode after GÜNTHERSCHULZE, BÄR and WINTER [17].

$\vartheta/(1+\gamma)$, according to Figs. 5—7.

ϑ, calculated with the value for γ given in the table.

ϑ (500) = value of ϑ for a voltage of 500 V, assuming that

$$\vartheta(500) = 500\,\vartheta(V_c)/V_c \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

The values of r and l are given in order to compare roughly the path of an electron starting from the cathode with the thickness d_c of the Crookes dark space. The maximum distance from the cathode, reached by an electron, will lie between $r + d_c$ and $2r + d_c$ for $r \gg d_c$ or $3\sqrt{V} \gg Hd_c$ and will be approximately equal to l for $3\sqrt{V} \ll Hd_c$ (see Fig. 8). For $3\sqrt{V} \sim Hd_c$ it will have an intermediate value.

The table shows that generally $r > d_c$ and $\lambda_e > d_c$ so that only few ions will be formed within the Crookes dark space. Only at very low pressures r and l are $\sim d_c$. Here however, r and l are also $\ll \lambda_e$ so that most of the electrons liberated from the cylindrical part of the cathode will

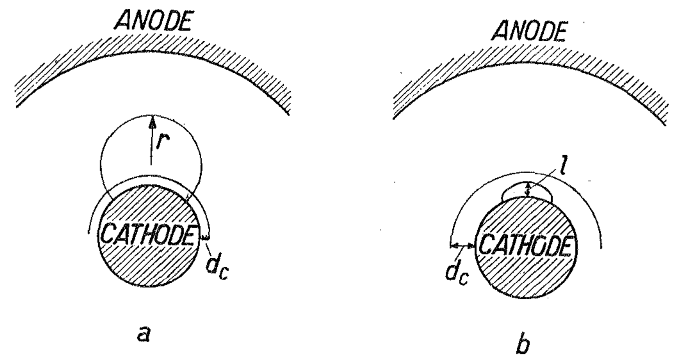

Fig. 8. Path of an electron liberated from the cathode for $3\sqrt{V} \gg Hd_c$ (case a) and $3\sqrt{V} < Hd_c$ (case b). $H \perp$ plane of drawing.

return to it without having performed any collision. In these circumstances it is possible that the electrons are liberated mainly from the end plates of the cathode [9]) so that the discharge has an abnormal character. We suppose that the decrease of ϑ at the lowest pressures is due to this effect and that the real course of the curves in Fig. 5 should be approximately as given by the dotted lines.

According to the table usually d_c is $< \lambda_p$ and $\ll \lambda_e$ so that the factor f_2 of § 1 is equal to 1. Only at the highest pressures used it possibly has to be taken into account ($p = 0.3$ mm).

[17]) A. GÜNTHERSCHULZE, W. BÄR and A. WINTER, Z. Phys. **111**, 208 (1938). These authors give γ as a linear function of V, γ becoming zero at a certain value u_0 of usually several hundred volts. As it is known that in the rare gases γ is still different from zero at $V = 0$, the values of GÜNTHERSCHULZE c.s. should not be used below the voltage region in which they were determined (500—3000 V.).

As to the dependence of ϑ on V the table shows that ϑ, after correction for the value of γ, is roughly proportional to V, so that the value of ϑ (500) calculated after Eq. (4) and corrected for the value of γ is approximately constant (see Fig. 9). The point for 400 V in Fig. 6 is

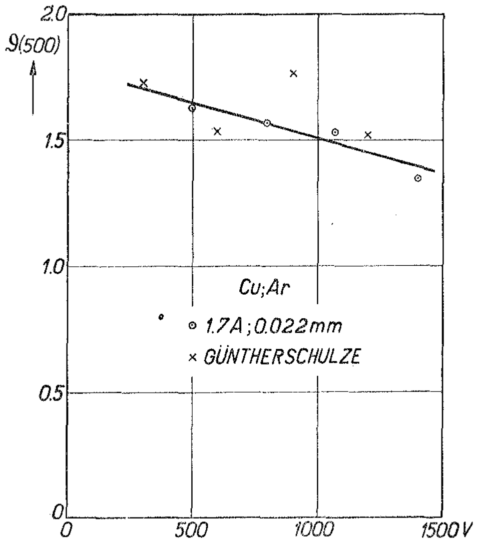

Fig. 9. Values of ϑ (500) being the value of ϑ reduced to 500 V after Eq. (4) and corrected for γ.

abnormally low, but as here also r and l are $< d_c$ this point was rejected for the reason given higher up. We do not know if the systematic small decrease of ϑ (500) with V in Fig. 9 which nevertheless remains, is real or is possibly due to the accepted value [18]) of γ.

The reason for the decrease of ϑ with increasing i in Fig. 7 is unknown. One could suppose that it was due to the ionisation of sputtered Cu-atoms which should return as ions to the cathode; the strong Cu-spectrum emitted by the discharge points in the same direction. This hypothesis, however, was not confirmed by the experiments with still larger current densities (see below).

It is remarkable that $\vartheta/(1+\gamma)$ has decreased with increasing p with a factor $\frac{1}{2}$ only at $p = 0.2$, corresponding to $pd = 0.5$. When the Cu-atoms were liberated from the cathode with room temperature velocity, however, the value of λ_1 (mean free path for 1 mm pressure) would be 0.005, and according to Eq. (1) the decrease of ϑ with a factor $\frac{1}{2}$ should occur already at $p_0 d = 0.011$ which makes a difference of a factor 45! A similar deviation from the expected value, although quantitatively much less, was observed by GÜNTHERSCHULZE for Ag with Ar^+ and for Ni with Hg^+ and is also

[18]) The value of γ is known to be very sensible to variations of the cathode surface, see A. GÜNTHERSCHULZE and H. BETZ, Z. Phys. **108**, 780 (1938); F. M. PENNING, Proc. Kon. Akad. v. Wetensch., Amsterdam, **33**, 841 (1930).

shown by Fig. 1 when the results for Ag in H_2 at high values of $p_0 d$ are compared with those at low values of $p_0 d$. For the explanation the following circumstances have to be taken into account:

1. probably the velocity of the Cu-atoms starting from the cathode is much larger than corresponds to room temperature [19]);
2. the mass of the Cu-atoms is larger than that of the Ar-atoms so that its velocity $\perp$ cathode cannot be reduced to zero by one collision;
3. the temperature of the gas is, due to the large energy used, much higher than room temperature [20]) $(p_0 < p)$;
4. in the cylindrical arrangement used here the number of returning atoms is smaller than in the case of parallel plates, considered in deriving Eq. (1).

The influence of 3 was confirmed by later measurements with a smaller cathode, where four times larger current densities could be applied. In this case the decrease of ϑ with increasing values of pd was still smaller. Combining the results from the measurements with both tubes we obtain as a mean value of $\vartheta_0(500)/(1+\gamma)$:

$$\vartheta_0\,(500)/(1+\gamma) = 1{,}7$$

and, correcting for the value of γ and applying Eq. (4):

$$500 < V_p < 1400\ V \qquad \vartheta_0\,(V_p) = 0{,}0037\ V_p \qquad \text{Cu and Ar}^+$$

The accuracy of this result we estimate as $\pm$ 25 %. As has been remarked already the agreement with the results of GÜNTHERSCHULZE and MEYER [6]) is very good.

§ 5. *Results for Al.*

In the literature one often finds the statement that the sputtering rate of aluminium is very small, BLECHSCHMIDT [8]) e.g. gives a value for Ar^+-ions which is only $1/20 \times$ that of copper. It has also been stated [8]) that this low value is due to a surface layer on the metal, in the first place of Al_2O_3. To study this phenomenon more quantitatively the following experiment was made. An Al-bar of 1 cm diameter in the axis of a glass tube was exposed intermittently to a heavy glow discharge in an axial magnetic field (40 mA; 900 V; 0.06 mm Ar). In the cylindrical discharge tube another glass tube of a somewhat smaller diameter could be moved in order to expose successively fresh parts of the glasswall to the sputtering. The results for 4 consecutive sputterings are shown in Fig. 10. Obviously an energy of 7600 wattsec is needed to free the cathode from the not sputtering surface layer (I); in the second and third experiment (II and

[19]) F. BAUM, Z. Phys. **40**, 686 (1937) finds a velocity of the sputtered atoms corresponding to the melting point of the metal used.

[20]) In a glow discharge with watercooled electrodes already temperature increases of 150°, were found (H. FISCHER, Z. Phys. **113**, 360 (1939)).

III) the sputtering sets in immediately. One night's standing (between III and IV) in humid air restores again the initial state of the surface [21]).

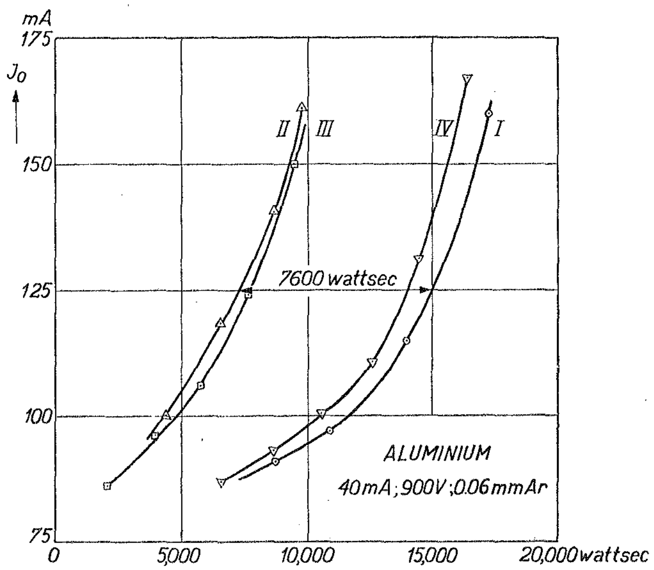

Fig. 10. Consecutive sputterings of an aluminium bar; sequence I, II, III, IV. I was the first sputtering after the pumping and filling of the apparatus, between III and IV the tube was opened and exposed to humid air of 1 atm. The thickness of the sputtered layer on the glass wall is given by the current I_0 through a glowlamp behind the tube, for which the glowing filament was just no longer visible.

The reason is probably the same as for the similar behaviour of MgO : according to GÜNTHERSCHULZE and BETZ [22]) in MgO the Mg^+ ions are sputtered which are drawn back to the cathode by the electric field.

The discharge with an oxide layer on the aluminium cathode is, apart from the small amount of sputtering, also characterised by a lower discharge voltage at the same magnetic field, due to the much larger value of γ for the oxide [23]). Moreover, the disappearance of the oxide layer manifests itself by a rather sudden change in the colour of the discharge due to the appearance of the strong resonance lines of Al (3944, 3962 Å).

Similar optical and electrical phenomena, although much less pronounced, were also found in the experiments with Cu, described in the preceding §§. Also with Cu the first sputtering after a new filling of the tube usually gave too low values for ϑ with, at the same time, deviating values of the cathode fall. Contrary to Al, however, the cathode fall was higher for the

[21]) Similar phenomena were already found by L. L. CAMPBELL, Phil. Mag. (6), **28**, 347 (1914).

[22]) A. GÜNTHERSCHULZE and H. BETZ, Z. Phys. **106**, 365 (1937).

[23]) For Mg GÜNTHERSCHULZE and BETZ l.c. found a decrease in λ from 1.88 to 0.37, when the oxide layer was removed. Compare the "spray discharge" phenomenon described by A. GÜNTHERSCHULZE and H. FRICKE, Z. Phys. **86**, 451 and 821 (1933).

unclean than for the clean Cu-surface. In the clean state, the colour of the discharge was of a deep green; especially at high pressures the Cu-lines were the strongest of the spectrum.

The values of ϑ for Al could be determined with an apparatus as that of Fig. 2 but with a hollow Al-cylinder instead of the copper one. The curves for ϑ (500) (corrected for the γ) are given in Figs. 11 and 12.

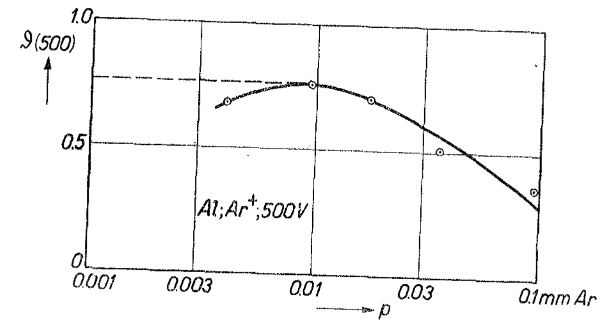

Fig. 11. Values of ϑ (500) for Al and Ar^+.

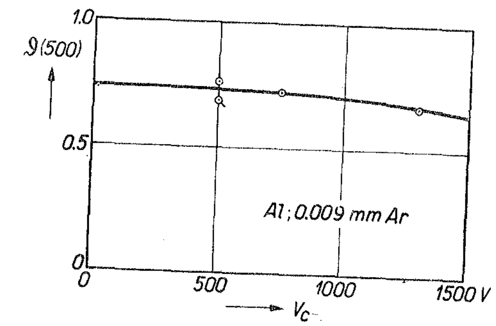

Fig. 12. Values of ϑ (500) for Al and Ar^+.

The dependence on i was not determined. Here also ϑ is approximately proportional to V. The decrease of ϑ with increasing pressure is stronger than for Cu, which may be due to the smaller mass of the Al-atoms. The most probable value of ϑ_0 following from these measurements is:

$$500 < V < 1400 \qquad \vartheta_0 (V_p) = 0{,}0015\ V_p \qquad \text{Al and Ar}^+$$

which is about half the value obtained for Cu.

§ 6. *Experiments with Ni and Ag.*

For these metals no special measurements of ϑ were made. Only a rough value could be obtained from earlier experiments in Ar and H_2, where a number of holders with mica plates were placed behind rings as R_1 and R_2 in Fig. 2. As here always the first sputtering after a new filling must be used, the metal was not as clean as in the measurements of the preceding §§, the more so as the sputtering did not occur in flowing gas.

§ 7. *Survey of the results obtained.*

In table II the results for ϑ_0 are summarized, together with the values of the evaporation heat W [24]) and the efficiency ε, calculated after Eq. (3). For Cu and Ag also the values according to GÜNTHERSCHULZE and MEYER are given (cathode temperature about 800° K.); for Ag and Ar the difference is rather large, in the other 2 cases the agreement is very good.

TABLE II.
Summary of the results for ϑ_0 (500)

Metal	Gas	$\frac{\vartheta_0\ (500)}{1+\gamma}$	ϑ_0 (500)	Estimated accuracy	W (volts)	ε	$\frac{\vartheta_0\ (500)}{1+\gamma}$ G. and M.
Al	Ar	0.75	0.83	± 25 %	2.92	0.5 %	
Cu	Ar	1.7	1.9	± 25 %	3.51	1.3 %	1.5
Ni	Ar	1.2	1.3^5	± 50 %	4.25	1.1 %	
Ag	Ar	1.4	1.5	± 50 %	2.99	0.9 %	2.7
Ni	H_2	0.14	0.15	± 50 %	4.25	0.01 %	
Ag	H_2	0.47	0.56	± 50 %	2.99	0.03 %	0.58

The conclusion of GÜNTHERSCHULZE and MEYER that the sputtering efficiency is only of the order of 1 % and lower, is confirmed.

[24]) LANDOLT–BÖRNSTEIN, 3ter Erg. Bd. III (1936), p. 2709. According to some authors the efficiency should be calculated by taking for W not the evaporation heat, but adding to it the melting heat and the energy required to heat the metal to the boiling point. This, however, makes not more difference in ε than a factor 1.3 at maximum for the metals considered here.

Physics. — *Recherches sur quelques phénomènes d'interférence des courbes de vibration* (suite). Par J. W. N. LE HEUX. (Communicated by Prof. P. ZEEMAN).

(Communicated at the meeting of December 30, 1939.)

1. Dans un travail antérieur, nous avons dit, qu'aucune des figures de la table IV, qui représente les diverses images de la formule

$$x = a \cos \alpha \cos \varphi$$
$$y = a \cos (\alpha + \theta) \cos (\varphi + \triangle)$$

ne peut donner l'image des lemniscates d'un cristal biaxial. Cependant, il y a deux figures, $2D$ et $4D$, qui présentent quelque ressemblance avec l'image précitée. En étudiant les particularités de ces figures, nous allons établir les conditions nécessaires et suffisantes pour chacune des deux spirales elliptiques, dont la superposition donne exactement l'image des lemniscates avec toutes ses variations. Quelques résultats des expériences à l'aide d'un appareil à quatre pendules sont réunis dans les tables VIII et IX.

2. Dans la numération de la table IV, les chiffres se rapportent à la différence de phase θ et les lettres à la différence de phase $\triangle$. Donc, les figures $2D$ et $4D$ ont une même différence de phase $\triangle$, près de 90°, mais la valeur de θ est pour $2D$ près de 0° et pour $4D$ près de 90°. Autrement dit: figurons-nous une ellipse aplatie E_2, qui diffère très peu de deux droites parallèles et une ellipse E_4, qui diffère très peu d'un cercle. La figure $2D$ peut être regardée comme l'ensemble des ellipses, presque cercles, inscrites dans les rectangles sur les doubles coordonnées des points de l'ellipse E_2 et la figure $4D$ comme l'ensemble des ellipses, presque cercles, inscrites dans les rectangles sur les doubles coordonnées des points de l'ellipse E_4. Il est évident, que la première bissectrice de l'angle des coordonnées est un axe de symétrie, donc les côtés des rectangles de la partie supérieure de l'ellipse E sont perpendiculaires aux côtés correspondants de la partie inférieure.

3. Observons ces rectangles avec plus de précision.

Nous distinguons dans la demie-ellipse $EGHKL$ (table VII, fig. A):

1. dans l'intervalle EG: un petit carré sur EL comme diagonale, des rectangles horizontaux, une droite horizontale GN.
2. dans l'intervalle GH: une droite horizontale GN, des rectangles horizontaux, un grand carré sur HP comme diagonale.