Physics. — *The mechanism in the positive column of a discharge.* By L. S. ORNSTEIN, H. BRINKMAN and T. HAMADA. (Communication from the Physical Institute of the University of Utrecht.)

(Communicated at the meeting of February 29, 1936).

SUMMARY.

The measurements by HAMADA [1]) of the temperature T of the gas along the diameter of a narrow tube, containing the positive column of a discharge in nitrogen, are discussed. The importance of the determination of T for the mechanism of a gas-discharge is emphasized. It is shown that the velocitydistribution of the electrons, depending on the parameter $E_0 . \lambda$ (E_0 axial fieldstrength, λ electronic mean free path) varies along the tube-diameter; this variation is only due to the radial variation of T.

For the discharge in nitrogen the fact is reported that $E_0\lambda$ ($=$ 0,066 Volt) is independent on the discharge conditions for pressures between 10 and 30 mm. At lower pressures $E_0\lambda$ increases, thus electrons with high energies become more probable.

The different shape of the intensitycurves over the tube-diameter for N_2 and N_2^+ bands are discussed. It is proved qualitatively that the effect of the increase of the total excitationprobability of the N_2 bands with temperature T (due to the excitation from N_2 molecules in higher vibrational states) must be taken into account in order to explain the experimental facts.

In an appendix an elementary deduction of the velocitydistribution of electrons in a gasdischarge is given.

In a previous communication [1]) one of us has given a survey of the main results of temperature-measurements in the positive column of a discharge in nitrogen. Together with the theoretical interpretation of these results, we will give in this paper a more general description of the mechanism in the positive column of a discharge.

§ 1. *The temperature of the gas in a discharge.*

In a discharge one of the most important facts to be known is the temperature T of the gas as a function of the place. If in a cylindrical tube the temperature has a maximum in the axis of the tube, the gasdensity has a minimum. The electrons and ions moving in axial direction, thus have the highest mobility in the axial regions of the tube. So the current-

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[1]) T. HAMADA, Proc. Royal Acad. Amsterdam, **39**, 50 (1936). Determination of the temperature in the column of a discharge from the intensity-measurement of rotational band spectra. — See this paper for experimental details.

density as a function of radius shows a still more prominent maximum than the radial electron distribution does. The radial temperature distribution is therefore strongly related with the so called radial "contraction" of the discharge, an effect that is easily noticed as the gaspressure increases.

Besides in many other aspects the temperature of the gas is of high interest. The phenomena in the discharge in which the mean free path of the particles plays a rôle (diffusion, velocitydistribution of the electrons and excitation of the gas by electrons) depend on the gastemperature; further in the effects depending on the v.d. (velocitydistribution) of the gasparticles, as there are the effectivity for collisions of the second kind, the contribution to the luminosity by thermal excitation of the gas, the widths of spectral lines, etc.

In the figs. 6—12 of T. HAMADA's paper the temperature as a function of radius is shown for a number of pressures and total currents. These functions can be considered as parabolae:

$$T = T_0 - \beta r^2.$$

Near the wall of the tube the measured temperatures may be too high. For it is difficult to determine T as the N_2-bands have a low intensity compared with the intensity in the axis of the tube and further, due to the way in which the end-on observations have been performed and measured, the averaging over a certain region of the tube-radius results in a systematic error.

The determination of T from the intensity-distribution in bandspectra is in our case accurate within 5 %. For this reason, together with the fact that in some cases only a few points fix the curve, the values of β, determining the shape of the radial temperature distribution, are inaccurate. They don't show a well defined dependence upon pressure, currentstrength and watt-input. We can only say that the observed values of β increase with increasing wattinput per cm in the column, in accordance with the theoretical expectation from the energy-equation.

The values of the temperature in the axis, T_0, are better defined. In fig. 5 of the cited communication of T. HAMADA the linear relation between temperature and the product p.i has been shown. More than this curve the plot of E_0 (E_0 = fieldstrength in the axis of the tube) against $\frac{p}{T_0}$ has a direct physical meaning (fig. 1). These curves show the relation between fieldstrength and gasdensity $\left(\backsim \frac{p}{T_0}\right)$. It suggests the constancy of $E_0\lambda$ $\left(\lambda \backsim \frac{T}{p}\right.$, the mean free path of the electrons$\left.\right)$ with varying discharge conditions; only at low pressures $E_0\lambda$ increases. These results are not in accordance with the relation between E_0 and λ given by GÜNTHERSCHULZE [2]).

[2]) A. GÜNTHERSCHULZE, Zs. f. Phys. **41**, 718 (1927); **42**, 763 (1927).

In fig. 2 (upper curve) we plotted the value of $E_0\lambda$ in electron-Volts as a function of gaspressure. Each value is the mean for all the currents at

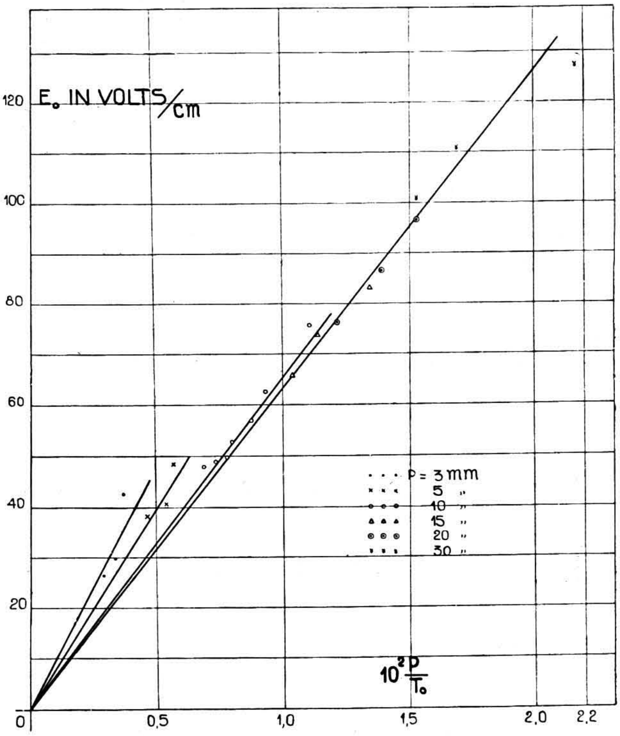

Fig. 1.

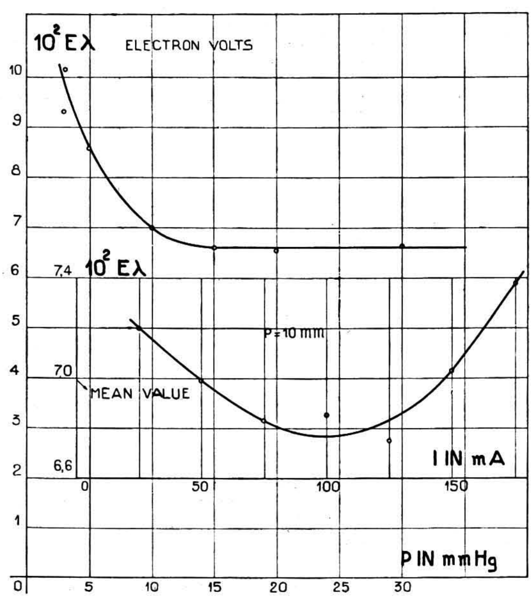

Fig. 2.

which experimental values of E_0 and T were available. $E_0\lambda$ for different currents at constant pressure deviates from the average less than 7 %, except for $p = 3$ mm, in which case $E_0 \lambda$ differs from the mean value + 15.7 %, —8.7 %, —7.0 % for $i = 25$, 50 and 75 mA. respectively.

For the calculation of $E_0 \lambda$ we have used as a value for the mean free path of electrons in N_2 at 1 mm. pressure and 0° C 2.85 . 10^{-2} cm. This is a mean value for electrons moving in N_2 with velocities aequivalent with about 5 e.V.[3])

§ 2. *Conclusions about the velocitydistribution (v.d.) of the electrons at different discharge conditions.*

The value of $E_0 \lambda$ is the parameter which fix the v.d. of the electrons. In different ways[4]) one can show that the number of electrons having energies between ε and $\varepsilon + d\varepsilon$ is given by

$$n_e(\varepsilon) \, . \, d\varepsilon = C \, . \, \varepsilon^{1/2} \, . \, e^{-a\left(\frac{\varepsilon}{E_0 \lambda}\right)^2} \, . \, d\varepsilon \quad . \; . \; . \; . \; . \quad (1)$$

$$\left[a = \frac{3m}{M \varrho^2}, \; m = \text{mass of electron}, \; M = \text{mass of gasparticle}, \right.$$

$$\left. \varrho = \text{electronic charge} \right]$$

assuming only elastic collisions of the electrons with the gasparticles, very small kinetic energies of the gasparticles compared with the energy of the electrons, small $E_0 \lambda$ values ($E_0 \lambda < 1$ Volt), λ independent on the electron velocities and no electrostatic interaction of the electrons among each other.

With this distributionfunction one can prove that the mobility μ of the electrons[4]) is given by

$$\mu = \frac{c \, . \, \lambda}{\sqrt{E_0 \lambda}} . \qquad (c = \text{constant}) \; . \; . \; . \; . \; . \; . \; . \quad (2)$$

If the excitation and ionisation of the gasparticles is taken into account the above mentioned simple form of the v.d. function alters in a way typical for the gas in question[5]). But still the value of $E_0 \lambda$ is the most important parameter. Complications arise if the gastemperature T is so

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3) R. B. BRODE, Rev. Mod. Phys. **5**, 257 (1933) (fig. 11).

4) M. J. DRUYVESTEYN, Physica **1**, 1003 (1934); Physica (old series) **10**, 61 (1930).
A. M. CRAVATH, Phys. Rev. **46**, 332 (1934).
B. DAVYDOV, Sowj. Phys. **8**, 59 (1935).
P. M. MORSE, W. P. ALLIS and E. S. LAMAR, Phys. Rev. **48**, 412 (1935).
For an elementary deduction of equation (1) see the appendix to this paper.

5) M. J. DRUYVESTEYN, Physica **3**, 65 (1936) (Neon).
J. A. SMIT, to be published shortly in Physica (Helium).

high, that the kinetic energy of the gasatoms is not very small compared with the electronenergies [6]) and if the mutual electrostatic interaction of the electrons must be taken into account. In these cases $E_0\lambda$, T and n_e determine the v.d. In the nitrogen-tube which T. HAMADA used, we estimate the number of electrons per cm^3 in the axis of the tube to 10^{10} à 10^{11}. The electrostatic interaction may then be neglected and we assume that in our cases the temperature of the gas doesn't have an appreciable influence on the v.d. of the electrons.

From fig. 2 we may thus conclude that the v.d. in the positive column of the nitrogen-discharge in the axis of the tube is independent on the dischargeconditions at pressures between 10 and 30 mm. At lower pressures the distribution varies in such a way that higher electron-energies become more probable.

In the tube-axis the v.d. seems to be for a certain pressure rather independent on the currentstrength. In the case of 10 mm pressure $E_0\lambda$ is observed for a large number of currents. In fig. 2 (lower curve, enlarged scale!) is shown that there exists in the tube axis a small variation of the v.d. with current.

§ 3. *The variation of the electronic v.d. along the diameter of the cylindrical discharge.*

As the temperature of the gas falls down towards the wall of the tube we see that $E_0\lambda$ decreases along the tube-radius proportional to T. Thus the v.d. of the electrons differs for the various cylindric zones of the tube in such a way that high velocities are more probable in the axis of the tube than in the layers near the wall.

On the first sight against this conclusion may be objected that outside the tube-axis a radial component of the field E_r exists and the total field $E = \sqrt{E_0^2 + E_r^2} > E_0$. Qualitatively we shall show, however, that for the electronic v.d. $E_0\lambda$ is the determining parameter and not $E\lambda$.

According to SCHOTTKY [7]) the radial field E_r has such a value that the mean driftvelocity of the electrons in radial direction, due to the combined influences of diffusion towards the wall and radial field action, equals the mean radial driftvelocity of the positive ions.

The radial force acting on the electrons is therefore:

$$-E_r\, e - \frac{d}{n_e}\frac{dn_e}{dr} \;{}^{7a)} \quad . \; . \; . \; . \; . \; . \; . \; . \quad (3),$$

the two components of the force acting in opposite directions.

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[6]) B. DAVYDOV, l.c. and SMIT's paper.

[7]) W. SCHOTTKY, Phys. Z.S. **25**, 342, 635 (1924).
M. J. DRUYVESTEYN, ZS. f. Phys. **81**, 571 (1933).

[7a]) $d(:)\, E_0\lambda$ can be calculated from equation (1).

Between two successive collisions the electron moves in a field of strength $E=\sqrt{E_0^2+E_r^2}$. However, the stationary v.d. is determined after a great number of collisions, that means, depends on the balance of energy-yields and energy-losses. This balancing differs strongly from the case in which no radial diffusion, caused by the radial variation of the electronconcentration, exists and only an electric field is present. For in our case, due to the radial diffusion, the number of energy-losses is larger than the number of energy-yields and the effect of an E larger than E_0 is compensated.

Quantitatively the influence of the radial field and electron diffusion on the v.d. may be calculated on the basis of the fundamental equation of LORENTZ [8]), extended by the work of MORSE, ALLIS and LAMAR [4]). From LORENTZ's equation [8]) one directly sees that the variation of the electron v.d. depends on the value of the force acting on an electron (see our expression (3)) and the value of E_r is just so that this force is very small compared with $e\,.\,E_0$.

A simple and more general proof, that the v.d. of electrons is the same if $E_0\lambda$ remains constant, is as follows. Assuming that the electrical energy-input per element of volume is totally given to the gas by the collisions of electrons with gasparticles, we can write:

$$E_0\,.\,I=\gamma\,.\,n_e\,n$$

[I = current density, γ is a function of the v.d. of the electrons, n = number of gasparticles per cm^3].
Now we know:

$$I=n_e\,.\,e\,.\,\mu\,.\,E$$

(e = electronic charge)

$$\mu=\frac{c\,.\,\lambda}{\sqrt{E_0\lambda}}$$

$$\lambda=\frac{1}{n\,.\,Q}$$

(Q = total crosssection of the gasparticles for collisions with electrons). Thus we have:

$$(E_0\,\lambda)^{3/2}=\frac{\gamma}{Q\,.\,e\,.\,c}.$$

If the v.d. doesn't vary, then we find $E_0\lambda$ is constant.

Conclusion: *The temperature variation over the tube-diameter is the only reason for the radial variation of the velocity distribution of the electrons.*

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[8]) H. A. LORENTZ, Theory of Electrons (1909, Teubner) page 266—274; see equation (65).

§ 4. *The excitation of the molecular spectra in the discharge.*

We shall try to give the interpretation of the curves, giving the intensity of a negative and a positive band of N_2 as a function of the radius. In fig. 13 of HAMADA's paper such curves are given. The intensity scale for these curves, obtained with various currents, must be adjusted in such a way that (— as has been proved by further experiments —) the intensity in the tube-axis is proportional to the current. This holds as well for the N_2^+ 0—0 band (λ 3914 Å) as for the bands 0—3 (λ 4059) and 1—4 (λ 3998) of the second positive bandsystem of N_2. From fig. 2 we have seen the small variation of the electronic v.d. in the tube-axis with current. Thus the currentdensity in the axis is practically proportional with the number of electrons. It is not exact to conclude that thus the intensity is proportional to n_ε, for the function giving the currentdensity at various radiï may differ for different currents. The latter may occur, as the temperatures along the radius are different for different currents (see fig. 9 in HAMADA's paper). However, probably the conclusion that the intensity is proportional to the number of electrons is correct.

From the detailed study of the excitation conditions in a discharge[9]), one can predict that for the rather high pressures with which we are dealing, the intensity of N_2 and N_2^+ bands (assuming excitation directly from the fundamental level of N_2) is independent on the pressure (as has been checked experimentally by T. HAMADA) and proportional to the number of electrons. Using the notation giving in the cited articles[9]) the number of excited particles

$$n_a = \frac{a_e}{\beta} \cdot n_e ,$$

$$a_e = \int_{\sqrt{\frac{2\varepsilon_a}{m}}}^{\infty} v \cdot q(v) \cdot f(v) \cdot dv \quad \text{and} \quad \beta = \int_0^{\infty} V' \cdot Q'(V') \cdot F(V') \cdot dV'$$

$\varepsilon_a =$ energy of the excited state,

$f(v)$ is the distribution function for the electronvelocities,

$q(v)$ the excitation function of the nitrogen bands[10]) in question,

$F(V')$ is the distribution function for the relative velocities of the gas-particles,

$Q'(V')$ the crosssection of the molecules for a collision of the second kind.

We see that β depends on the temperature of the gas.

[9]) L. S. ORNSTEIN and H. BRINKMAN, Physica **1**, 797 (1934).
P. J. HARINGHUIZEN, Thesis, Utrecht (1935).

[10]) L. S. ORNSTEIN u. G. O. LANGSTROTH, Proc. Royal Acad. Amsterdam, **36**, 384 (1933).
G. O. LANGSTROTH, Proc. Roy. Soc. London, **146**, 166 (1934); **150**, 371 (1935).
O. HERRMANN, Ann. d. Phys. **25**, 166 (1936).

In molecular spectra the intensity of one band is the total intensity of all rotational lines together.

In the investigation of the excitation of molecular spectra we must account for the following effect. If the temperature of the gas rises, the population of the higher vibrational states of the N_2 molecule (in the fundamental $^1\Sigma$-state) increases. At 1500° K the ratio of the number of molecules in the vibrational state with quantumnumber 0 to the number of molecules in the vibrational state with quantumnumber 1 evaluates $\frac{1}{10}$. If the excitationprobability from the 0-state is small compared with the excitationprobability from the 1-state the total excitationprobability turns out to increase with the temperature of the gas! This is the case for the bands of the second positive bandsystem of N_2. That can be seen directly from the relative position of the molecular-potential curves on the nuclear-distance scale [11]), together with the application of the FRANCK-CONDON-principle for excitation [10]). For the N_2^+ band this effect does not occur. In both cases no calculations of excitationprobabilities from higher vibrational states have been carried out; so we cannot calculate the magnitude of this effect. It is very probable that the relation between intensity and current for the positive bands is affected by the reported phenomena.

In the case of 10 mm pressure the intensitydistribution along the diameter of the tube has been measured for the band λ 3914 of N_2^+ as well as for the band λ 3998 of N_2. The ratio of the intensity of the N_2 band and of the N_2^+ band shows a maximum near the tube axis (see fig. 3; computed

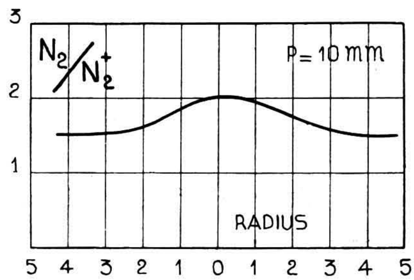

Fig. 3.

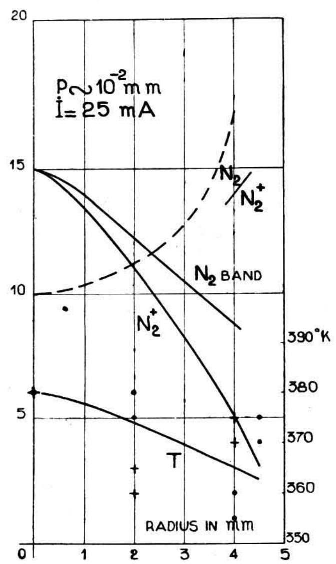

Fig. 4.

from fig. 13 in HAMADA's paper). As we have seen the v.d. of the electrons shifts to the lower velocities if we go from the axis towards the wall. Thus

[11]) W. JEVONS, Report on Bandspectra (1932).

the remarkable way in which the intensity ratio of positive and negative bands varies with the radius can only be understood if the effect of the variation of the v.d. is more than compensated by the effect of the increase of the total excitationprobability with increasing gastemperature.

For the very low pressure of about 10^{-2} mm (λ about 4 cm; in this case SCHOTTKY's theory is not valid), the experimental facts are quite different from the case stated above. It has been found by HAMADA (see fig. 12 of his paper) that the temperature is practically constant over the tube-diameter. The small temperature variation in radial direction is shown on larger scale in our fig. 4 (• • T derived from N_2^+ band, ++ T derived from N_2 band). Thus there will be a radial variation of the v.d. of the electrons. As the temperatures are low the total excitationprobability for the N_2 band is the same at different radiï. The intensity ratio of N_2 and N_2^+ bands has in this case a minimum at the tube-axis and increases with 70 % near the wall of the tube, indicating the variation of the electron v.d. along the diameter.

APPENDIX.

Elementary deduction of the v.d. of electrons moving in a gasdischarge with a constant gradient.

The following elementary deduction of the velocitydistribution law is perhaps of some interest as this law is of utmost importance for the investigation of the discharge.

We assume that the mass M of the atoms (or molecules) is very large in comparison to that of the electrons, so that $\frac{m}{M}$ may be considered as small compared to unity. Further we assume that the velocity of the atoms is small (low temperature of the gas). The electrons move in an electric field of strength E. Under these assumption the energy of the electrons is changed by the field between the collisions with the atoms in positive or negative sense, and this energy is changed after each collision in the average with the factor $1 - \frac{2m}{M} = 1 - \beta = \alpha$.

As a first approximation the distribution law shows a spherical symmetry.

Let us now follow an electron on its way during n free paths ($n - 1$ collisions) and let $l_1, l_2, \ldots\ldots, l_n$ be the successive paths between the collisions, which form angles $\vartheta_1, \vartheta_2, \ldots\ldots, \vartheta_n$ with the direction of E. Let the energy at the beginning of the path be ε_0, than we have:

$$\varepsilon_n = (\ldots[\{(\varepsilon_0 + Eel_1 \cos\vartheta_1)\,\alpha + Eel_2 \cos\vartheta_2\}\,\alpha + Eel_3 \cos\vartheta_3]\,\alpha + \ldots \\ \ldots)\,\alpha + Eel_n \cos\vartheta_n$$

or

$$\varepsilon_n = \varepsilon_0\,\alpha^{n-1} + Ee\,.\,[l_1 \cos\vartheta_1\,.\,\alpha^{n-1} + l_2 \cos\vartheta_2\,\alpha^{n-2} + \ldots + l_n \cos\vartheta_n] \quad . \quad (I)$$

The mean value of ε_n will therefore be:

$$\overline{\varepsilon_n}^{\varepsilon_0} = \varepsilon_0 \, a^{n-1}$$

as the mean value $\overline{\cos \vartheta} = 0$. After a very large number of collisions the particle, with initial energy ε_0, has the energy 0.

Let us now determine $\overline{\varepsilon_n^2}^{\varepsilon_0}$; for this quantity we get:

$$\overline{\varepsilon_n^2} = \varepsilon_0^2 \, a^{2(n-1)} + E^2 \, e^2 \, . \, [l_1^2 \cos^2 \vartheta_1 \, . \, a^{2(n-1)} + \ldots + l_n^2 \cos^2 \vartheta_n]$$

as

$$\overline{\cos \vartheta_p \, . \, \cos \vartheta_q} = 0 \qquad (p \neq q).$$

Now $\overline{l_1^2 \cos^2 \vartheta_1} = \frac{2\,\lambda^2}{3}$, where λ means the mean free path of the electrons. Thus we get:

$$\overline{\varepsilon_n^2} = \varepsilon_0^2 \, a^{2(n-1)} + \frac{2\,E^2 \, e^2 \, \lambda^2}{3} \cdot \frac{1 - a^{2n}}{1 - a^2}$$

$$= \varepsilon_0^2 \, . \, a^{2(n-1)} + \frac{E^2 \, e^2 \, \lambda^2 \, M}{6\,m} \, . \, (1 - a^{2n})$$

for very large values of n we find:

$$\overline{\varepsilon^2} = \frac{E^2 \, e^2 \, \lambda^2 \, M}{6\,m}.$$

Now the change of the energy distribution of the electrons is given by a generalised diffusion equation (PLANCK-FOKKER) in which $\beta \, \overline{\varepsilon_n^2}$ $(n \rightarrow \infty)$ is the diffusion constant. This can be proved taking (I) as an EINSTEIN-LANGEVIN equation of the form

$$\frac{d\varepsilon}{dn} = -\beta \, \varepsilon + F \; {}^{*)} \, , \; \beta = \frac{2\,m}{M}$$

and deriving the diffusion equation in the common way. The distribution in energy is then a GAUSSian distribution with modulus $2\,\overline{\varepsilon^2}$; however, taking into account that for a given energy the velocity can have any direction with the same probability, we find for the velocitydistribution equation (1) of this paper.

*) Compare: G. E. UHLENBECK and L. S. ORNSTEIN, On the theory of the BROWNian Motion, Phys. Rev. 36, 823 (1930). See especially §§ 5 and 6.