THE THEORY OF CASEOUS ISOTOPE SEPARATION IN A FORCE FIELD WITH APPLICATION TO THE ULTRACENTRIFUGE

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The possibility of isotope separation in the ultracentrifuge was pointed out by Lindemann and Aston,¹ who gave the equilibrium theory both for an ideal gas and for an incompressible fluid. Mulliken² and Harkins³ examined the possibility more critically, and Humphreys⁴ has considered the departure from equilibrium conditions when the continuous flow method of centrifugation is employed. The problem of the rate of sedimentation in a continuous medium in a constant force field has been worked out by Mason and Weaver⁵, ⁶.

Lamm⁷ has given an equation for the transport of molecules in a field of force, and from it has derived a differential equation governing diffusion in such a field. His diffusion equation has been solved by Faxen,⁸ Oka,⁹ and Archibald.¹⁰ Only Archibald’s solution is suitable for numerical calculation, however. Lamm’s equation, and all the solutions of it which have been given, are restricted to cases in which the sedimentation and diffusion coefficients are constant. They are therefore suitable for the study of sedimentation in liquids, but do not apply to diffusion in gases if there is great variation in density. It is true that Archibald writes the diffusion equation in a more general form but, because he was interested in sedimentation in liquids, he did not use it in that form.

When diffusion in gases in a force field is considered it seems necessary to re-examine the basis on which Lamm’s equations rest,

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¹ Lindemann and Aston, Phil. Mag. 37, 523 (1919).
⁴ Humphreys, Phys. Rev. 56, 684 (1939).
⁵ Mason and Weaver, Phys. Rev. 28, 412 (1924).
⁶ Weaver, Phys. Rev. 27, 499 (1926).
¹⁰ Archibald, Phys. Rev. 53, 746 (1938); 54, 371 (1938).
and the variation in the sedimentation and diffusion coefficients must be considered. Investigation shows that these latter depend on the mean free path which, in turn, is a function of the density. We give here a kinetic theory derivation of the transport equation, write the diffusion equation in its most general form, and solve it for cases in which the variation of the mean free path must be taken into account. A typical calculation is then made.

THE TRANSPORT EQUATION

We want to find the number of molecules of a given kind crossing an area of one square centimeter in a plane which is perpendicular to a field of force, in one second. The analysis will apply to each component of a mixture of any number of component gases.

Let \( N_1 \) be the number of molecules per cm.\(^3\) of one kind. We assume an acceleration \( a \) in the x-direction, which may be taken as constant over an interval equal to the mean free path \( L_1 \). \( L_1 \) will depend on the kind of molecule considered and also on \( N \), the total number of molecules per cm.\(^3\). The molecules are all supposed to have the same velocity \( u_1 \), and a perfectly random distribution as to direction of motion, so that the mean free path in the x-direction is \( \frac{L_1}{\sqrt{3}} \). On the average, the molecules crossing a plane located by the coordinate \( x \), and proceeding in the positive x-direction, come from a level \( x - \frac{L_1}{\sqrt{3}} \) at which \( N_1 = N_1(x) = \frac{L_1}{\sqrt{3}} \frac{\partial N_1}{\partial x} \), and only one half of them will have a component of motion in this direction. They cross the plane with a velocity \( u_{1x} + \frac{a L_1}{u_{1x} \sqrt{3}} \) provided we take the gain in velocity in traversing \( \frac{L_1}{\sqrt{3}} \) as much less than \( u_1 \) itself. This is allowable even where \( a \) is quite large, as may be shown by a simple calculation.

Then the number of molecules crossing one cm.\(^2\) of the plane per second, and travelling in the positive x-direction is
\[ \frac{1}{2} \left( N_1 - \frac{L_1}{\sqrt{3}} \frac{\partial N_1}{\partial x} \right) \left( u_{1z} + \frac{aL_1}{u_{1z}\sqrt{3}} \right), \]
and the corresponding number travelling in the negative x-direction is
\[ \frac{1}{2} \left( N_1 + \frac{L_1}{\sqrt{3}} \frac{\partial N_1}{\partial x} \right) \left( u_{1z} - \frac{aL_1}{u_{1z}\sqrt{3}} \right). \]
So the net number crossing one cm.² of the plane per unit time in the positive x-direction is
\[ Z_1 = \frac{1}{\sqrt{3}} \left( \frac{aL_1 N_1}{u_{1z}} - \frac{L_1 u_{1z} \partial N_1}{\partial x} \right) = \frac{aL_1 N_1}{u_1} - \frac{L_1 u_1}{3} \frac{\partial N_1}{\partial x}. \]
The flow across a plane with any orientation can readily be found by replacing \( a \) in (1) by the component of the acceleration normal to the plane.

If in (1) we set \( s_1 \), the sedimentation coefficient, equal to \( \frac{L_1}{u_1} \), and \( D_1 \), the diffusion coefficient, equal to \( \frac{L_1 u_1}{3} \) we get the transport equation of Lamm.⁷

The transport equation (1) can also be obtained by the method of Kennard,¹¹ who shows that in a constant force field there is an equivalent \( \frac{\partial N_1}{\partial x} \) given by
\[ \frac{\partial N_1}{\partial x_{\text{equiv.}}} = - \frac{N_1 m_1 a}{kT} \]
where \( m_1 \) is the mass of a molecule, and \( k \) and \( T \) have the usual meanings of Boltzmann constant and absolute temperature respectively. Using the method of Kennard we have
\[ Z_1 = - D_1 \left( \frac{\partial N_1}{\partial x} - \frac{N_1 m_1 a}{kT} \right) = \frac{N_1 L_1 a u_1 m_1}{3kT} - \frac{L_1 u_1}{3} \frac{\partial N_1}{\partial x} \]
if we put \( u_1^2 = \frac{3kT}{m_1} \) and \( D_1 = \frac{L_1 u_1}{3} \).

A kinetic theory derivation, not given here, which makes use of the velocity distribution law, indicates that a more accurate transport equation is

where $\Omega_1$ is the average velocity $\sqrt{\frac{8kT}{\pi m_1}}$, or

$$Z_1 = \sqrt{\frac{8}{3\pi}} \left( \frac{N_1 a L_1}{G_1} - \frac{L_1 G_1}{3} \frac{\partial N_1}{\partial x} \right)$$

where $G_1$ is the root-mean-square velocity $\sqrt{\frac{3kT}{m_1}}$. If we identify $u_1$ of the derivation given above, with $G_1$ in (3), then the factor $\sqrt{\frac{8}{3\pi}}$ will simply change the time scale in our final results. Because of this, because it is nearly equal to unity, and because such coefficients in kinetic theory are never very certain, we suppress it and use the transport equation (1).

**THE GENERAL DIFFUSION EQUATION**

The transport equation can be used to derive the general equation governing diffusion in a force field as follows.

Consider an element of volume $dx \cdot dy \cdot dz$, where $dx$, $dy$, $dz$ are much greater than $L_1$, but so small that the variations in $N_1$, $a$, and $L_1$ are sensibly linear as we pass through it. The rate of increase of the number of molecules of a given kind in the element is

$$\frac{\partial}{\partial t} (N_1 \ dx \ dy \ dz) = \frac{\partial}{\partial x} (N_1 a z) dy \ dz + \frac{\partial}{\partial y} \left( \frac{N_1 a x}{u_1} \right) dx \ dz + \text{etc.}$$

Inflow $= L_1 \left[ \left( \frac{N_1 a z}{u_1} - \frac{u_1}{3} \frac{\partial N_1}{\partial x} \right) dy \ dz + \left( \frac{N_1 a x}{u_1} - \frac{u_1}{3} \frac{\partial N_1}{\partial y} \right) dx \ dz + \text{etc.} \right]$.

Outflow $= \frac{1}{u_1} \left( L_1 N_1 a z + \frac{\partial}{\partial x} (L_1 N_1 a z) \ dx \ dz \right) + \frac{1}{u_1} \left( L_1 \frac{\partial N_1}{\partial x} + \frac{\partial}{\partial x} (L_1 \frac{\partial N_1}{\partial x}) \ dx \ dz \right)

whence

$$\frac{\partial N_1}{\partial t} = \frac{u_1}{3} \frac{\partial}{\partial x} (L_1 \frac{\partial N_1}{\partial x}) - \frac{1}{u_1} \frac{\partial}{\partial x} (L_1 N_1 a z) + + + -$$

$$= \frac{u_1}{3} \text{ div } (L_1 \text{ grad } N_1) - \frac{1}{u_1} \text{ div } (L_1 N_1 \vec{a})$$

where $\vec{a}$ denotes the vector acceleration.

An equation of this type holds for each component of the mixture, $u$ and $L$ being in general different for each component.
In the case of the ultracentrifuge we may assume that $L_1 = L_1 (r, t), N_1 = N_1 (r, t), a = a(r)$, and transforming to polar coordinates, we obtain for the general diffusion equation

$$\frac{\partial N_1}{\partial t} = \frac{u_1 L_1}{3} \left[ \frac{\partial}{\partial r} \left( L_1 \frac{\partial N_1}{\partial r} \right) + \frac{L_1}{r} \frac{\partial N_1}{\partial r} \right] - \frac{1}{u_1} \left[ \frac{\partial}{\partial r} \left( L_1 N_1 a \right) + \frac{1}{r} \left( L_1 N_1 a \right) \right].$$  \hspace{1cm} (5)

If in this equation we set $\frac{u_1 L_1}{3} = D_1 = \text{a constant}, \ a = \omega^2 r$, as in the centrifuge, and $\frac{L_1}{u_1} = s_1$, the sedimentation constant, then we obtain the familiar sedimentation equation of Lamm,

$$\frac{\partial N_1}{\partial t} = D_1 \left( \frac{\partial^2 N_1}{\partial r^2} + \frac{1}{r} \frac{\partial N_1}{\partial r} \right) - \omega^2 s_1 \left( \frac{\partial N_1}{\partial r} + 2 N_1 \right).$$

**Boundary Condition and Steady State Distribution**

At a fixed boundary (as of an enclosing vessel) the flow ($Z_1$) across the bounding surface is zero. If the surface is perpendicular to the $x$-axis, then from (1)

$$\frac{L_1 N_1 a_x}{u_1} - \frac{L_1 u_1}{3} \frac{\partial N_1}{\partial x} = 0,$$

or

$$\frac{\partial N_1}{\partial x} = \frac{3 N_1 a_x}{u_1^3} = \frac{m_1 N_1 a_x}{k T}.$$  \hspace{1cm} (6)

The steady state distribution can be found either by setting the flow ($Z_1$) everywhere equal to zero, in which case (6) is valid at all points, or by setting $\frac{\partial N_1}{\partial t} = 0$ and using (4).

For simplicity let us assume that the distribution of the molecules depends on one space coordinate only, as does $a$. We obtain as the steady state equation

$$\log N_1 = \frac{3}{u_1^3} \int a(x) \cdot dx + \log N_{10}.$$  \hspace{1cm} (7)

**Application to the Ultracentrifuge**

The solution of the diffusion equation (4), when applied to the ultracentrifuge, depends on an ordinary differential equation for which we were unable to find a solution for the general case in which high speeds are considered. For a particular type of centrifuge, however, it was found possible to obtain graphical sloutions on the differential analyzer. For that reason we consider a centrifuge in which the gas is enclosed in a tube whose length is parallel to the $x$-axis.
and perpendicular to the axis of rotation. We take the mean free path \( L_1 \) as inversely proportional to the total number of molecules per cm.\(^3 \) \( (N) \). It is by considering the variation of the mean free path that our treatment differs from those which have previously been given. For simplicity we consider a mixture of two gases, although the method can be extended to a mixture of any number. Quantities descriptive of the one component for which we give the full analysis have the subscript \((1)\) appended to them; quantities description of the other component bear the subscript \((2)\).

Then

\[
L_1 = \frac{c_1}{\sqrt{N}}, \quad N = N_1 + N_2, \quad a = \omega^2 x.
\]

Putting these values in \((4)\) we obtain

\[
\beta_1 \frac{\partial N_1}{\partial t} = \frac{\partial}{\partial x} \left( \frac{1}{N} \frac{\partial N_1}{\partial x} \right) - \alpha_1 \frac{\partial}{\partial x} \left( \frac{N_1 x}{N} \right)
\]

(8)

where

\[
\beta_1 = \frac{3}{c_1 u_1}, \quad \alpha = \frac{3 \omega^2}{u_1^2} = \frac{\omega^2 m_1}{kT} = \frac{\omega^2 M_1}{RT},
\]

\( m_1 \) is the mass of a molecule of the first kind, \( M_1 \) is the chemical molecular weight of the component considered, and \( R \) is the universal gas constant. A similar equation holds for the second gas.

This equation is to be solved subject to the boundary condition at the outer boundary

\[
\frac{\partial N_1}{\partial x} = \frac{m_1 \omega^2}{kT} x N_1 = \alpha_1 x N_1
\]

which follows from \((6)\), and the boundary condition at the axis of rotation \( \frac{\partial N_1}{\partial x} = 0 \) which follows the symmetry which exists with respect to that axis.

From \((7)\), the steady state distribution will be

\[
N_1 = N_{10} e^{-\frac{\alpha_1 x^2}{2}},
\]

which is the same as the result obtained by Lindemann and Aston.\(^1\)

We assume that initially \((t = 0)\) the gas and rotor have attained a steady angular velocity \( \omega \), that the gas as a whole has so distributed itself as to assume the quasi-steady state distribution given by

\(^{12}\) S. Chapman, *Phil. Trans. 217 A*, § 13 g (1917).
\[ N = N_0 e^{\frac{\alpha x^2}{2}}, \]

and that thereafter \( N \) does not change much with time. Here

\[ \alpha = \frac{\omega^2 m}{kT} = \frac{\omega^2 M}{RT} \]

and

\[ M = \frac{M_1 N_1 + M_2 N_2}{N_1 + N_2} \]

is the molecular weight which would be determined experimentally for the mixture.

Inserting this expression for \( N \) in (8), we obtain

\[ N_0 \beta_1 \frac{\partial N_1}{\partial t} = \frac{\partial}{\partial x} \left( e^{-\frac{\alpha x^2}{2}} \frac{\partial N_1}{\partial x} \right) - \alpha_1 \frac{\partial}{\partial x} \left( e^{-\frac{\alpha x^2}{2}} N_1 x \right). \tag{9} \]

It will simplify matters to replace \( x \) by the dimensionless variable

\[ y = \sqrt{\frac{\alpha}{RT}} x = \sqrt{\frac{M}{RT}} \omega x \]

and to define a function \( \Psi \) by

\[ N_1 = \Psi(x, t) N_0 e^{\frac{\alpha_1 x^2}{2}} = \Psi(y, t) N_0 e^{\frac{\alpha_1 y^2}{2}} \]

\[ = \Psi(y, t) N_0 e^{\frac{M_1 y^2}{M}}. \]

Then the function \( \Psi \) tells us how the component (1) approaches its steady state, and (9) becomes

\[ \gamma_1 e^{\frac{y^2}{2}} \frac{\partial \Psi}{\partial t} = \frac{\partial^2 \Psi}{\partial y^2} + b_1 y \frac{\partial \Psi}{\partial y} \tag{9a} \]

where

\[ \gamma_1 = \frac{N_0 \beta_1}{\alpha}, \quad b_1 = \frac{\alpha_1 - \alpha}{\alpha} = \frac{M_1 - M}{M}. \]

For one component \( b > 0 \) and for the other \( b < 0 \).

The boundary conditions now reduce to

\[ \frac{\partial \Psi}{\partial y} = 0 \]

at \( y = 0, \sigma \) where \( y = \sigma \) determines the outer boundary.

If a solution is assumed of the form

\[ \Psi = Y(y) T(t) \]

then it is found that

\[ T = e^{-\frac{p^2 t}{\gamma_1}} \]

where \( -p^2 \) is the separation constant, and \( Y \) satisfies
\[
\frac{d^2 Y}{dy^2} + b_1 y \frac{dY}{dy} + \frac{b_1 y^2}{4} Y = 0. \tag{10}
\]

Before discussing this equation let us remove the middle term by the substitution

\[
Y = e^{-\frac{b_1 y^2}{4}} v(y). \tag{11}
\]

Then

\[
\frac{d^2 v}{dy^2} + \left( \frac{b_1 y^2}{2} - \frac{b_1 y^2}{4} \right) v = 0
\]

or approximately, when \( b_1 \) is small,

\[
\frac{d^2 v}{dy^2} + \frac{b_1 y^2}{2} v = 0. \tag{12}
\]

No exact analytic solution of this equation was found, but through the courtesy of the Moore School of Electrical Engineering of the University of Pennsylvania, graphical solutions of (10) and (12) were obtained on their differential analyzer. In the problem of isotope separation \( b_1 \) will not exceed one, and it was found that for values of \( b_1 \) of this order of magnitude the solution of (10) is quite accurately given by the substitution (11), where \( v \) is a solution of (12). This is shown in Table I below. Since the solutions are oscillatory we chose to compare their maximum and minimum values. The cases of \( b_1 = 0.04343 \) \( n_2 \) where \( n = 1, 2, \ldots, 8 \) were investigated.

**Table I**

Maxima and Minima of the Solution of Equation (10) for the case \( n = 5, b_1, = 1.086, p = 5. \)

<table>
<thead>
<tr>
<th>( y )</th>
<th>0.000</th>
<th>0.604</th>
<th>1.120</th>
<th>1.530</th>
<th>1.840</th>
<th>2.080</th>
</tr>
</thead>
<tbody>
<tr>
<td>Analyzer Solution</td>
<td>1.000</td>
<td>-0.873</td>
<td>0.613</td>
<td>-0.408</td>
<td>0.271</td>
<td>-0.914</td>
</tr>
<tr>
<td>Substitution (11)</td>
<td>1.000</td>
<td>-0.872</td>
<td>0.615</td>
<td>-0.405</td>
<td>0.269</td>
<td>-0.187</td>
</tr>
</tbody>
</table>

**BOUNDARY CONDITIONS**

The values of \( p \) must be so chosen as to satisfy the boundary condition \( \frac{dy}{dy} = 0 \) for \( y = 0, \sigma \), which will be satisfied if \( \frac{dY}{dy} = 0 \) for \( y = 0, \sigma \). All the solutions obtained on the analyzer satisfy \( \frac{dY}{dy} = 0 \) for \( y = 0 \), and they show that if one value of \( p \) (say \( p = p_1 \)) can be found for which \( \frac{dY}{dy} = 0 \) at \( y = \sigma \), then the values \( p = p_1 n \), where
n is an integer, also lead to solutions which satisfy this condition. This is true, at least, with all the values of \( b_1 \) and \( p \) which we considered.

The curves obtained further demonstrate that when \( \frac{dY}{dy} = 0 \) at \( y = 0, \sigma \), where \( Y \) is the analyzer solution of (10), so too is \( \frac{dv}{dy} = 0 \) for \( y = 0, \sigma \), where \( v \) is the analyzer solution of (12). In Fig 1, which is typical, (A) is the solution of (10) and (C) is its derivative curve; (B) is the solution of (12) and (D) is its derivative curve. The case is that for which \( b_1 = 1.086 \) and \( p = 5 \).

This fortunate simultaneous vanishing of the derivatives of \( Y \) and \( v \) makes it easy to use the solutions of (12), and easy to prove them orthogonal, but it is not what we would expect from the substitution (11). The latter predicts that if \( \frac{dY}{dy} = 0 \) at \( y = 0, \sigma \), then

\[
\frac{dv}{dy} = 0 \quad \text{at} \quad y = 0, \quad \text{but for} \quad y = \sigma \\
\frac{dv}{dy} - \frac{b_1 y v}{2} = 0.
\]

Nevertheless, the outer boundary condition predicted by the differential analyzer is in pretty good agreement with that which follows from the substitution (11) because the term \( \frac{b_1 y v}{2} \) will, in general, be small. The discrepancy is no greater than might be expected from the approximate nature of (12), which was obtained by dropping two small terms.

The solutions of (12) in the interval \( 0 < y < 2.08 \), as given by the analyzer for various values of \( p \) (\( p = 1, 2, \ldots, 8 \)), are shown in Figs. 2 and 3.* It would have been desirable to secure them in a larger interval, for then calculations could have been made for higher speeds, but the machine ran off scale shortly beyond 2.08. The end values of the various solutions do not differ very much, an average being 0.58. In the case to be calculated below \( b_1 = 0.04343 \), so for \( y = 2.08 \)

\[
\frac{dv}{dy} = \frac{b_1 y v}{2} = 0.026,
\]

corresponding to a slope of \( 1^\circ 30' \), a disagreement with the analyzer solutions which we may safely neglect.

* See Table II for values of \( v \) and \( Y \) at maxima and minima. These values are differential analyzer scale readings.
We will therefore take as the boundary condition
\[ \frac{dv}{dy} = 0 \]
\( y=0, \sigma, \) where, as stated before, \( \sigma \) locates the outer boundary. It is, in fact \( \sqrt{\frac{M}{RT}} \omega x_{\text{max}} \).

**ORTHOGONALITY OF THE FUNCTIONS (v)**

It is easy to prove that the solutions of Equation (12) are orthogonal in an interval at both ends of which the above boundary condition is satisfied. Choose any two values of the separation constant \( p \) which cause the boundary condition to be satisfied. Let these values be \( p_m \) and \( p_k \), and let the corresponding solutions be \( v_m \) and \( v_k \).

Then
\[ \frac{d^2v_m}{dy^2} + p_m^2 e^{\frac{v^2}{2}} v_m = 0, \]
\[ \frac{d^2v_k}{dy^2} + p_k^2 e^{\frac{v^2}{2}} v_k = 0. \]

Multiply the first of these equations by \( v_k \, dy \) and the second by \( v_m \, dy \), subtract and integrate. We get
\[ \int_0^\sigma \left[ v_k \frac{d^2v_m}{dy^2} - v_m \frac{d^2v_k}{dy^2} \right] \, dy + (p_m^2 - p_k^2) \int_0^\sigma e^{\frac{v^2}{2}} v_m v_k \, dy = 0. \]

Integrating the first term by parts, we have
\[ \left[ v_k \frac{dv_m}{dy} - v_m \frac{dv_k}{dy} \right]_0^\sigma + \int_0^\sigma \left[ \frac{dv_m}{dy} \frac{dv_k}{dy} - \frac{dv_k}{dy} \frac{dv_m}{dy} \right] \, dy \]
\[ + (p_m^2 - p_k^2) \int_0^\sigma e^{\frac{v^2}{2}} v_m v_k \, dy = 0. \]

The expression in brackets vanishes at both limits because of the boundary conditions; the second term vanishes because the integrand is identically zero; so
\[ \int_0^\sigma e^{\frac{v^2}{2}} v_m v_k \, dy = 0, \quad k \neq m. \]

Now consider a function \( f(y) \) developed in terms of a series of the functions \( v \).
\[ f(y) = \sum B_kv_k(y). \]
Multiply by \( e^{\frac{y^2}{2}} v_m \, dy \) and integrate between the limits 0 and \( \sigma \). Then
\[
\int_0^\sigma v_m \, e^{\frac{y^2}{2}} \, f(y) \, dy = \sum B_k \int_0^\sigma e^{\frac{y^2}{2}} v_k v_m \, dy.
\]
Each term in the right member vanishes except that for which \( m = k \), and solving for \( B_k \) we obtain
\[
B_k = \frac{\int_0^\sigma e^{\frac{y^2}{2}} v_k f(y) \, dy}{\int_0^\sigma e^{\frac{y^2}{2}} v_k^2 \, dy}. \tag{13}
\]
In the absence of an analytical expression for the functions \( f(y) \), we can plot the integrands in the expression (13) and evaluate the integrals by mechanical integration. In the typical calculation to be given below the integrals were evaluated by means of a planimeter.

The theory just developed can be applied to the study of isotope separation in the ultracentrifuge as follows.

Let \( Q_1 \) be the total number of molecules of kind (1); \( Q_2 \), the total number of molecules of kind (2); \( Q \), the total number of molecules of both kinds. Then for a centrifuge tube of unit cross-section
\[
Q = 2N_0 \int_0^\sigma e^{\frac{y^2}{2}} \, dx = \frac{2N}{\sqrt{\alpha}} \int_0^\sigma e^{\frac{y^2}{2}} \, dy. \tag{14}
\]
This equation determines \( N_0 \).

Initially
\[
N_1 = \theta N_0 \, e^{\frac{y^2}{2}}
\]
where \( \theta \) is defined to be \( \frac{Q_1}{Q} \) In the steady state
\[
N_1 = N_{10} \, e^{\frac{M_1 \, y^2}{2}}.
\]
Since the number of molecules of kind (1) remains fixed, we have the relation
\[
\frac{\sqrt{\alpha} \, Q_1}{2} = \theta N \int_0^\sigma e^{\frac{y^2}{2}} \, dy = N_{10} \int_0^\sigma e^{\frac{M_1 \, y^2}{2}} \, dy. \tag{15}
\]
This equation determines \( N_{10} \).

Corresponding to each permissible value \( \theta_k \) of \( p \) there will be
a solution of (9a) of the form

\[ \Psi_k = e^{-\frac{p_{k_1}^2}{\gamma_1}} e^{-\frac{b_{12}y^2}{4}} v_k. \]

In order to satisfy the initial and final conditions we must express \( N_1 \) by a series of the form

\[ N_1 = N_{10} e^{\frac{M_1 y^2}{2}} \sum_{k=0}^{\infty} B_k \Psi_k (y, t), \]

or

\[ N_1 = N_{10} e^{\frac{M_1 y^2}{2}} \sum_{k=0}^{\infty} B_k e^{-\gamma_1^t Y_k(y)}. \]

To make this reduce to the correct steady state distribution when \( t = \infty \), we must define \( p_0 = 0 \) and \( B_0 \ Y_0 = 1 \), definitions which are consistent with (10).

Then

\[ N_1 = N_{10} e^{\frac{M_1 y^2}{2}} \left[ 1 + \sum_{k=1}^{\infty} B_k e^{-\gamma_1^t Y_k(y)} \right] \]

\[ = N_{10} e^{\frac{M_1 y^2}{2}} \left[ 1 + e^{-\frac{b_{12}y^2}{4}} \sum_{k=1}^{\infty} B_k e^{-\gamma_1^t v_k(y)} \right] \] (16)

from the substitution (11).

The outer boundary condition can be satisfied by properly choosing \( p_1 \). Then \( p_2 = 2p_1, p_3 = 3p_1 \), etc. In the calculation which we make we choose \( p_1 = 1 \) and find that the outer boundary is then determined by \( \sigma = 2.08 \). The curves given in Figs. 2 and 3 are useful only if the outer boundary is so determined, or if \( \sigma \) has a smaller value determined by the first zero of the derivative of one of these curves. This means that the experimenter, in order to test our results, must so adjust the centrifuge speed or so determine its radius as to make

\[ \sqrt[\frac{M}{RT}]^{\omega_{\text{max}}} = 2.08, 1.33, \text{ etc.} \]

In order to decide whether the solutions shown in Figs. 2 and 3 are of any real use to the experimenter, let us calculate \( \sigma \) for three sets of conditions under which experiments have been performed. In the absence of information as to the correct value of \( T \) we will

13 Beams and Skarstrom, Phys. Rev. 56, 266 (1939).

* See Table II.
take it as 300°K. We get the following values.

<table>
<thead>
<tr>
<th>Substance</th>
<th>( \omega ) (r.p.s.)</th>
<th>( x ) (cm.)</th>
<th>( \sigma )</th>
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<td>( \text{CCl}_4 )</td>
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Thus we see that for one of these cases, at least, our solutions are of direct use. For cases in which we cannot work out the theory in detail from our curves we will show below that we can, nevertheless, draw some conclusions regarding the rapidity of approach to the steady state.

It would be desirable to express \( p_1 \) as a function of \( \sigma \), but this function was not discovered. However, we can make a pretty good guess at the value of \( p_1 \) corresponding to a given value of \( \sigma \) by referring to Fig. 4, in which \( \frac{1}{\sigma} \) has been plotted as a function of \( p_1 \).

Unfortunately, the form of the curve is most doubtful in the region of greatest interest to us, that for which \( \sigma > 2.08 \).
The coefficients \( B_k \) must be so evaluated as to reduce (16) to the correct initial distribution

\[
N_1 = \theta N_0 e^{\frac{y^2}{2}}
\]

when we put \( t = 0 \). That is

\[
\theta N_0 e^{\frac{y^2}{2}} = N_{10} e^{\frac{M_1 y^2}{2}} \left[ 1 + e^{-\frac{b_{12}^2}{4}} \sum_{k=1}^{\infty} B_k v_k (y) \right],
\]

or

\[
\sum_{k=1}^{\infty} B_k v_k = \frac{\theta N_0}{N_{10}} e^{-\frac{b_{12}^2}{4}} - e^{-\frac{b_{12}^2}{4}}.
\]

This is the function \( f(y) \) which is to be inserted in (13) in order to evaluate the coefficients \( B_k \).

**Typical Calculation**

We shall now use the preceding analysis to make a calculation for a mixture of chlorine molecules of the types \( \text{Cl}_2^{35} \) and \( \text{Cl}_2^{37} \). It is recognized that in neglecting molecules of the type \( \text{Cl}^{35} \text{Cl}^{37} \), we are considering a purely hypothetical case. We do so, however, for the sake of simplicity. It is our purpose merely to show how a calculation can be effected.

The constants have the following values.

\[
M_1 = 74, \quad M_2 = 70, \quad M = 70.92
\]

\[
b_1 = \frac{M_1 - M}{M} = 0.04343, \quad \theta = 0.2300
\]

If we are to use the curves in Figs. 2 and 3 then \( \sigma = 2.08 \). Taking \( T = 300^\circ K \) and \( x_{\text{max.}} = 1 \text{ cm.} \) it is found that the angular velocity corresponding to this value of \( \sigma \) is 6208 revolutions per second. Then from (15).

\[
\theta N_0 \int_0^{2.08} e^{\frac{y^2}{2}} dy = N_{10} \int_0^{2.08} e^{\frac{M_1 y^2}{2}} dy = \sqrt{\frac{N_{10}}{M_1}} \int_0^{2.125} e^{\frac{z^2}{2}} dz,
\]

whence \( \frac{\theta N_0}{N_{10}} = 1.06 \).

The coefficients \( (B_k) \) are to be evaluated so as to satisfy

\[
\sum B_k v_k (y) = \frac{\theta N_0}{N_{10}} e^{-\frac{b_{12}^2}{4}} - e^{-\frac{b_{12}^2}{4}} = 1.06 e^{-0.01086y^2} - e^{-0.01086y^2}.
\]
Using (13), the first five coefficients were evaluated by plotting the integrands and integrating with a planimeter. The remaining three coefficients were found to be too small to determine in this way. The coefficients found were

\[ \begin{align*}
B_1 &= + 0.0589 \\
B_2 &= + 0.0041 \\
B_3 &= + 0.0036 \\
B_4 &= - 0.0021 \\
B_5 &= + 0.0017.
\end{align*} \]

Plotting the successive terms in the series \( \sum B_k v_k \) shows that the first term is the most important, and that the series converges slowly. A good approximation is obtained by taking the first term only, and altering \( B_1 \) to 0.06. It is perhaps more informative to write, not \( N_1 \), but an expression for the relative concentration \( (F_1) \), defined by

\[ F_1 = \frac{N_1}{N} \]

Then

\[ F_1 = \frac{N_{10} e^{\frac{M_1}{M}} 2}{N_0 e^{\frac{y_1}{2}}} \left[ 1 + 0.06 e^{-\frac{t}{\gamma_1}} e^{-\frac{b_{1y}^2}{4}} v_1(y) \right] \]

\[ = \frac{\theta}{1.06} \frac{e^{b_{1y}^2}}{e^{\frac{y_1}{2}}} \left[ 1 + 0.06 e^{-\frac{t}{\gamma_1}} e^{-\frac{b_{1y}^2}{4}} v_1(y) \right] \]

\[ = 0.218 e^{0.0217 y^2} \Psi(y, t), \]

\( \Psi \) being the expression in brackets.

To determine \( \Psi \), we must first determine \( \gamma_1 \).

\[ \gamma_1 = \frac{N_0 \beta_1}{\alpha} = \frac{Q \sqrt{\alpha}}{2 \int_0^\sigma \frac{y^2}{e^2} dy} \frac{3}{c_1 u_1 \alpha} \]

which readily reduces to

\[ \gamma_1 = \frac{\sqrt{3}}{\omega L_{10} \int_0^\sigma \frac{y^2}{e^2} dy} \sqrt{\frac{M_1}{M}} \]

where by \( L_{10} \) we mean the mean free path of the molecules of kind (1) at the pressure at which the gas is put into the centrifuge. For the case under consideration we will take \( L_{10} = 4.47 \times 10^{-6} \). With the previously stated values of the other constants, this gives us \( \gamma_1 = 1.8956 \) and

\[ \Psi(y, t) = 1 + 0.06 e^{-0.5275} e^{-0.01086 y^2} v_1(y). \]
Values of $\Psi$ for various values of $t$ are plotted in Fig. 5. It will be seen that the steady state is approached rapidly.

When the steady state has been reached then, at the outer boundary

$$F_1 = 0.2386, \quad F_2 = 0.7580, \quad F_1 + F_2 = 0.9966.$$  

This justifies the assumption that $N$ does not vary much with time. It is of interest to compare the initial and final values of the ratio $\frac{F_1}{F_2}$ at the outer boundary. Initially $\frac{F_1}{F_2} = 0.2987$, finally $\frac{F_1}{F_2} = 0.3147$, an increase of about 5.35%.
Let us investigate the effect on the rapidity of approach to the steady state of an increase in speed to 8400 r. p. s., giving us \( \sigma = 2.81 \). Neglecting all terms containing the time except the first, in (16), we see that this rapidity is determined by

\[
e^{\frac{p_1^2 t}{\gamma_1}} = e^x p \left( -\frac{\omega L_1}{\sqrt{3}} \int_0^\gamma y^2 dy \sqrt{\frac{M_1}{M}} t \right)
\]

In this case \( \frac{1}{\sigma} = 0.356 \) and referring to Fig. 4, we get \( p_1 = 0.570 \).

The ratio of \( \frac{p_1^2}{y_1} \) in this case to \( \frac{p_1^2}{y_1} \) for the case computed in detail above is, from (17), 1.85. An increase in speed in the ratio 1.35:1 increases the rapidity of separation in the ratio 1.85:1.

We have only outlined here the method of studying the distribution of the heavier component gas. The study of the distribution of the other component proceeds in the same way.

If, as above, we are content with the approximation given by taking only the first term of the series, then the curves we have plotted in Figs. 2 and 3 give us a choice of eight different values of \( \sigma \).

**Approximate Analytic Solutions**

In a few cases it was found possible to obtain approximate analytical solutions of the general diffusion equation. No calculations were made with them because they appear to be laborious to use, and because these cases do not possess the interest for us to the one worked out above. The results are given below, together with outlines of the methods by which they were obtained.

**The Centrifuge Problem in Cartesian Coordinates**

A case in which \( y \) is sufficiently small can be treated by making, in (12), the substitution.

\[
e\frac{\frac{z^2}{2}} = 1 + \frac{y^2}{2}.
\]

Then \( v \) satisfies

\[
\frac{d^2v}{dy^2} + p^2 (1 + \frac{z^2}{2}) v = 0.
\]

Let \( z = \frac{p y^2}{2\sqrt{2}} \) and \( A = \frac{p}{\sqrt{2}} \) and this reduces to
\[ z \frac{d^2v}{dz^2} + \frac{1}{2} \frac{dv}{dz} + (A + z) v = 0. \]

Except for the coefficient \( \frac{1}{2} \) this is the same as an equation discussed at length by Sharpe,\(^{14}\) and may be solved by the methods employed by him.

The solution may be expressed as the confluent hypergeometric function

\[ v = e^{iz} M\left( \frac{1}{4}, \frac{1}{2}; \frac{iA}{2}, \frac{1}{2}, -2iz \right) \]

where by the notation \( M(\alpha, \rho, z) \) we mean a solution of

\[ z \frac{d^2M}{dz^2} + (\rho - z) \frac{dM}{dz} - \alpha M = 0. \]

Using the method discussed in Forsyth, Diff. Equ. Chap. VII we obtain

\[ v = B_k \int_0^\frac{\pi}{2} \csc \frac{\phi}{2} \cos(z \cos \phi + A_k \log \cot \frac{\phi}{2}) \, d\phi \]

provided that we use a sum of such solutions and that

\[ \sum B_k = 0. \]

There is also the series solution

\[ v = \sum a_n z^n \]

where the coefficients satisfy the formula

\[ a_n n(2n - 1) + 2a_{n-1} A + 2a_{n-2} = 0. \]

The result is

\[ v = a (1 - 2Az - \frac{2A^2 + 1}{3} z^2 + \frac{2A(A^2 + 7)}{3 \cdot 5} z^3 + \cdots) \]

\[ + a_{\infty} z^{1/4} (1 - \frac{2A}{3} z - \frac{3 - 2A^2}{15} z^2 + \frac{4A(13 - 2A^2)}{7 \cdot 6 \cdot 15} z^3 + \cdots) \]

For the case in which \( p \) is sufficiently large it can be shown by the B. W. K. method\(^{16}\) that a solution of (12) is

\[ v = C e^{-\frac{y^2}{8}} \sin (p \int_0^y e^{\frac{t^2}{4}} \, dt) \]

where \( C \) is a constant.


\(^{16}\) Watson, Bessel Functions, 100.

\(^{16}\) Kemble, Fundamental Principles of Quantum Mechanics, 90.
In order to satisfy the boundary conditions, \( p \) must be chosen that
\[
p \int_0^\sigma e^{\frac{y^2}{4}} \, dy = n \pi
\]
where \( n \) is an integer. When \( \sigma \) is small this reduces to
\[
p_1 \sigma = \pi,
\]
a relation which is in close agreement with the curve plotted in Fig. 4.

**The Centrifuge Problem in Polar Coordinates**

If the diffusion equation in polar coordinates (5) is used, then by methods completely analogous to those already employed for the treatment of the centrifuge problem in cartesian coordinates, there is obtained as a solution
\[
N_1 = N_{10} e^{\frac{M_1}{M} y^2} \Psi(y, t)
\]
where now \( \Psi \) satisfies the equation
\[
\gamma_1 e^{\frac{y^2}{2}} \frac{\partial \Psi}{\partial t} = \frac{\partial^2 \Psi}{\partial y^2} + \frac{1}{y} \frac{\partial \Psi}{\partial y} + b_1 y \frac{\partial \Psi}{\partial y}.
\]
Then
\[
\Psi = e^{-\frac{p_0^2}{\gamma_1}} Y(y)
\]
and
\[
\frac{d^2 Y}{dy^2} + \frac{1}{y} \frac{dY}{dy} + b_1 y \frac{dY}{dy} + p^2 e^{\frac{y^2}{2}} Y = 0.
\]
It would have been more useful to the experimenter had we solved this equation on the differential analyzer rather than equation (10), but the machine could not handle the term \( \frac{1}{y} \frac{dY}{dy} \). For \( y \) sufficiently small, analytical expressions for the solutions can be obtained. In fact, by suitable substitutions, we are led to the equation of Sharpe referred to above.

Let
\[
e^{\frac{y^2}{2}} = 1 + \frac{y^2}{2}, \quad Y = e^{-\frac{b_1 y^2}{4}} v, \quad z = \frac{p y^2}{2\sqrt{2}}.
\]
Then
\[
z \frac{dv}{dz^2} + \frac{dv}{dz} + (A + z) v = 0.
\]
This is Sharpe's equation,\textsuperscript{14} for which he gets the following solutions,

\[
v = 1 - \frac{z^2}{2^2} + \frac{z^4}{2^4 4^2} - \cdots - zA + z^2 A^2 - \frac{z^3}{36} (A^3 - 5A) + \frac{z^4}{16 \cdot 36} (A - 14 A^2) - \cdots.
\]

\[
= C \int_0^\infty \cos \left( z \cos \phi + A \log \cot \frac{\phi}{2} \right) d \phi,
\]

which reduces to unity for \( z = 0 \) if \( C = \frac{2}{\pi} \cosh \frac{\pi A}{2} \). Watson\textsuperscript{17} points out that the solution may also be expressed as the hypergeometric function

\[
v = e^{\pm iz} M \left( \frac{1}{2} = \frac{iA}{2}, 1, = 2iz \right).
\]

The orthogonality of the solutions may be proved as before.

The final result is

\[
N_1 = N_{10} e^{\frac{M_1}{M} \frac{M^2}{2} \frac{\gamma_1}{4} \sum_{k=0}^\infty B_k e^{-\frac{b_k}{\gamma_1} e^{2\sqrt{2}}} \frac{1}{2} (\frac{iA}{2}, 1, = \frac{i\sqrt{2} k}{\sqrt{2}}).
\]

**Separation in a Constant Force Field**

In a gravitational field \( a = -g \), as before, \( L_1 = \frac{c_1}{N} \). If these substitutions are made in the diffusion equation (4), then

\[
\beta_1 \frac{\partial N_1}{\partial t} = \frac{\partial}{\partial x} \left( \frac{1}{N} \frac{\partial N_1}{\partial x} \right) + \alpha_1 \frac{\partial}{\partial x} \left( \frac{N_1}{N} \right)
\]

where

\[
\beta_1 = \frac{3}{c_1 u_1}, \quad \alpha_1 = \frac{3g}{u_1^2} = \frac{M_1 g}{R T}.
\]

Again a rapid movement of the gas as a whole is assumed such that there is attained the quasi-steady state distribution.

\[
N = N_0 e^{-\frac{m g x}{k T}} = N_0 e^{-\alpha x},
\]

and the function \( \Psi \) is defined by

\[
N_1 = N_{10} e^{-\alpha_1 x} \Psi(x, t) = N_{10} e^{-\frac{M_1 y}{M}} \Psi(y, t)
\]

where \( y = \alpha x \). Then \( \Psi \) satisfies

\textsuperscript{17} Watson, Bessel Functions (Camb. 1922), 105.
\[ \gamma_1 e^{-\gamma} \frac{\partial \Psi}{\partial t} = \frac{\partial^2 \Psi}{\partial y^2} - b_1 \frac{\partial \Psi}{\partial y} \]

where \( b_1 = \frac{M_1 - M}{M} \) and \( \gamma_1 = \frac{N_0 \theta_1}{\alpha^2} \).

It follows that

\[ \Psi = e^{-\frac{p^2}{\gamma_1}} Y(y) \]

where \( Y \) is a function which satisfies

\[ \frac{d^2 Y}{dy^2} - b_1 \frac{dY}{dy} + p^2 e^{-\gamma} Y = 0. \]

Let

\[ Y = e^{\frac{b_1 y}{2}} \]

then

\[ \frac{d^2 v}{dy^2} + \left( p^2 e^{-\gamma} - \frac{b_1^2}{4} \right) v = 0 \]

or, approximately,

\[ \frac{d^2 v}{dy^2} + p^2 e^{-\gamma} v = 0. \]

If \( z = \frac{y}{2} \) this becomes

\[ \frac{d^2 v}{ds^2} + 4p^2 e^{-2z} v = 0. \]

This equation was studied by Lommel\(^{18}\) who gave the solution

\[ v = J_0 \left( 2p e^{-z} \right), \]

\( J_0 \) denoting a Bessel function of order zero.

So, finally,

\[ N_1 = N_{10} e^{-\frac{M_1}{M} y} e^{\frac{b_1 y}{2}} \sum_{k=0}^{\infty} B_k e^{-\frac{p_k^2}{\gamma_1}} J_0 \left( 2p_k e^{-\frac{y}{2}} \right). \]

To satisfy the boundary condition \( \frac{dv}{dy} = 0 \) at \( y = 0 \) we need only pick the \( p_k \) as the half roots of \( J_1 \). The other boundary condition is \( N_1 = 0 \) at \( y = \infty \). This is ensured by the exponential factor

\[ e^{-\frac{M_1}{M} y} e^{\frac{b_1 y}{2}} = e^{\frac{M_1 + M}{M} y}. \]

\(^{18}\) Lommel, *Studien uber die Bessel'schen Functionen* (Leipzig, 1868).
In conclusion the writer wishes to express his thanks to the Moore School of Electrical Engineering of the University of Pennsylvania for the use of their differential analyzer; to Professor J. W. Beams, of the University of Virginia, for suggesting the problem; and especially, to Professor E. E. Witmer, of the University of Pennsylvania, under whose guidance the study was made, for continued assistance and encouragement.

### TABLE II.

Maxima and Minima of the solutions of Equations. (10) and (12).

\[
\frac{d^2 Y}{dy^2} + b \frac{dY}{dy} + \frac{y^2}{\epsilon^2} Y = 0 \tag{10}
\]

When \( b = 0 \), (10) reduces to (12) and \( Y = v \).

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