# The Theory of the Separation of Isotopes by Thermal Diffusion

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The time-dependent partial differential equation of the "separation column" of Clusius and Dickel has been solved exactly. Only an approximate solution, subject to the restriction that the fractional concentration $c$ is everywhere small compared to unity, was known previously. The present work makes a comparison between theory and experiment possible over the full range of concentration values.

## I. INTRODUCTION

THOUGH Chapman[1] had suggested as early as 1919 that the phenomenon of thermal diffusion discovered theoretically by Enskog and himself might be used for separating isotopes, attempts in this direction did not meet with success till Clusius and Dickel[2] pointed out that convection currents could be utilized for greatly enhancing the effect. The apparatus used by them consisted of a long vertical tube closed at both ends with an axial hot wire which could be maintained at any desired temperature by passing an electric current. Introduction of the gaseous mixture of isotopes into this tube gave rise to convection currents rising near the axis and descending near the cooled wall of the tube. When the tube was allowed to remain in this condition for a sufficiently long time, the combined effect of convection and thermal diffusion led to a partial separation of the two constituents of the mixture. The theory of the operation of the "separation column," as it is called, has been worked out by a number of authors. The most notable among them are Waldmann,[3] Furry,[4,5] Jones,[4,5] Onsager, and Debye.[6] The various treatments, though they differ somewhat, lead ultimately to the same transport equation

$$q=Hc(1-c)-K\partial c/\partial z, \tag{1}$$

where $q$ is the transport of the lighter isotope up the tube in g/sec, $c$ is the fractional number density of the molecules of the lighter isotope, $H$, $K$ are constants[4] depending on the nature of the gaseous mixture and the specifications of the column, and $z$ is the height in cm of any cross section of the column from its lower end. A similar equation holds for the transport of the heavier isotope down the tube.

Equation (1) will fail to hold, of course, near the two ends of the column where the convection current turns round; but, as these regions occupy only a small part of the total length, little error is committed by assuming it to be valid throughout the entire length of the column. Conservation of mass then leads to the equation of continuity

$$\mu\partial c/\partial\tau=-\partial q/\partial z, \tag{2}$$

where $\mu$ is the mass of gas in unit length of the column and $\tau$ is the time in sec.

Combining Eqs. (1) and (2), and denoting partial differentiation by a subscript, we have

$$\mu c_\tau=-H(1-2c)c_z+Kc_{zz}. \tag{3}$$

This is the basic equation of the separation column. Its solution subject to the appropriate boundary conditions gives the concentration distribution along the column as a function of time. Both Debye[6] and Bardeen[7] have treated this equation with the restriction that $c$ is small compared to unity, so that the term $c^2$ in Eq. (1) can be neglected. On this assumption the simplified form of the transport equation is

$$q=Hc-Kc_z \tag{4}$$

and that of the equation of continuity is

$$\mu c_\tau=-Hc_z+Kc_{zz}. \tag{5}$$

Equation (5) can be solved easily with the help of the standard theory of linear differential equations; but very often the experimental conditions necessitate a more complete discussion of the differential equation (3) without the above restriction on the values of $c$. In this paper we shall show that it is possible to solve the equation completely by reducing it to the linear form by means of a suitable transformation. Before proceeding to demonstrate this it will be convenient to bring Eq. (3) into the dimensionless form,

$$c_t=-(1-2c)c_x+c_{xx}, \tag{6}$$

by the substitutions,

$$x=Hz/K,\quad t=H^2\tau/\mu K.$$

The same substitutions transform Eq. (1) into

$$q/H=c(1-c)-c_x \tag{7}$$

and the simplified Eqs. (4) and (5) into

$$q/H=c-c_x \tag{8}$$

and

$$c_t=-c_x+c_{xx}. \tag{9}$$

[1] S. Chapman, Phil. Mag. 38, 182 (1919).
[2] K. Clusius and G. Dickel, Z. physik. Chemie B44, 397 (1939).
[3] L. Waldmann, Z. Physik 114, 53 (1939).
[4] R. C. Jones and W. H. Furry, Revs. Modern Phys. 18, 151 (1946). This problem is stated on page 178.
[5] W. H. Furry and R. C. Jones, Phys. Rev. 69, 459 (1946).
[6] P. Debye, Ann. Physik 36, 284 (1939).
[7] J. Bardeen, Phys. Rev. 57, 35 (1940).

## II. TRANSFORMATION OF EQ. (6)

To linearize Eq. (6) we first make the substitution $c=\frac{1}{2}+v_x$, obtaining

$$v_{xt}=(v_x^2)_x+v_{xxx}.$$

Integration with respect to $x$ yields

$$f(t)+v_t=v_x^2+v_{xx},$$

where $f(t)$ is an arbitrary function of $t$. Since the addition of an arbitrary function of time to $v$ leaves $c$ unaltered, we can absorb $f(t)$ in $v$ and write simply

$$v_t=v_x^2+v_{xx}. \tag{10}$$

Next we make the substitution $v=\phi(w)$ in Eq. (10), where the functional form $\phi$ is to be suitably chosen. This gives

$$v'w_t=(v')^2w_x^2+v''w_x^2+v'w_{xx},$$

where $v'=dv/dw$, and $v''=d^2v/dw^2$. If the functional form be so chosen that

$$(v')^2+v''=0, \tag{11}$$

then this equation reduces to

$$w_{xx}=w_t, \tag{12}$$

which is the familiar equation of ordinary diffusion in one dimension.

A solution[8] of Eq. (11) is $v=\ln w$. Thus, the substitution

$$c=\tfrac{1}{2}+w_x/w \tag{13}$$

transforms the nonlinear equation (6) in $c$ into a linear equation in $w$. The same substitution transforms the transport equation (7) into

$$q/H=\tfrac{1}{4}-w_{xx}/w. \tag{14}$$

First, we shall make use of the function $w$ in deriving the solution for the steady state which corresponds to constant $q$. By Eqs. (12) and (14),

$$\tfrac{1}{4}b^2=\tfrac{1}{4}-q/H=w_{xx}/w=w_t/w,$$

whence

$$w=\exp(b^2t/4)\cdot g(x).$$

If Eq. (12) is to be satisfied by $w$, $g(x)$ must be of the form $Ae^{bx/2}+Be^{-bx/2}$. The relation (13) then gives

$$c=\tfrac{1}{2}+\tfrac{1}{2}b(Ae^{bx/2}-Be^{-bx/2})/(Ae^{bx/2}+Be^{-bx/2}).$$

In the special case, $q=0$,

$$c=\tfrac{1}{2}+\tfrac{1}{2}\tanh\tfrac{1}{2}(x-x_0).$$

The expression for $w$ in terms of $c$ is, by Eq. (13),

$$w=g(t)\exp\left[-\tfrac{1}{2}x+\int_l^x cdx\right], \tag{15}$$

where $lK/H$ is the length of the column, and $g(t)$ is an arbitrary function of time. The expression for $w$ in terms of $c$ therefore contains an arbitrary function of time as a multiplier. But the condition that $w$ is to satisfy the differential equation (12) instead of an equation of a more general type obtained by direct substitution of Eq. (13) in Eq. (6), determines $g(t)$ up to a constant factor which still remains arbitrary. This can be seen in the following way: by Eqs. (14), (15), and (2),

$$w_{xx}=[\tfrac{1}{4}-H^{-1}q(x,t)]w$$

and

$$\begin{aligned}w_t&=\left\{[g'(t)/g(t)]+\int_l^x c_t dx\right\}w\\&=\{[g'(t)/g(t)]-H^{-1}q(x,t)+H^{-1}q(l,t)\}w.\end{aligned}$$

Therefore, if Eq. (12) is to hold, we must have

$$g(t)=A\exp\left[(t/4)-H^{-1}\int_0^t q(l,t)dt\right];$$

where the constant $A$ is arbitrary. Hence, the expression (15) can be written as

$$w=A\exp\left[(t/4)-H^{-1}\int_0^t q(l,t)dt-(x/2)+\int_l^x cdx\right]. \tag{16}$$

This expression will be required later in deriving some important results.

## III. THE INITIAL AND BOUNDARY CONDITIONS

We now proceed to discuss the initial and the boundary conditions of the problem. Linearization of the differential equation alone will not be sufficient to make the problem solvable unless the boundary conditions also assume a linear form. Fortunately, this requirement is fulfilled in the two most important cases discussed below.

In the first case the column is closed at both ends, so that the boundary conditions are $q=0$ at both $x=0$ and $x=l$; that is,

$$w_{xx}=\tfrac{1}{4}w,\quad \text{at } x=0 \text{ and } x=l. \tag{17}$$

This will be referred to as Problem (I).

In the second case the column is closed at the upper end and is connected to a reservoir of infinite capacity at the lower end, so that the concentration there remains constant and equal to $c_0$, the concentration in the reservoir. The corresponding boundary conditions are

$$\left.\begin{aligned}&-w_x=(\tfrac{1}{2}-c_0)w=\sigma w,\quad \text{at } x=0\\ \text{and}\quad &w_{xx}=\tfrac{1}{4}w,\quad \text{at } x=l\end{aligned}\right\}. \tag{18}$$

This will be referred to as Problem (II).

[8] This transformation was used by the author in a previous paper (S. D. Majumdar, Phys. Rev. 72, 393 (1947)) to linearize an equation occurring in the general theory of relativity.

The initial concentration will be taken to be constant throughout the length of the column. This is the only case which occurs in practice. If we take $A=\exp(c_0 l)$ in Eq. (16), the corresponding initial condition for $w$ comes out to be

$$w(x,0)=\exp[-x/2+c_0x]=e^{-\sigma x},\quad \text{at } t=0. \tag{19}$$

When Eq. (12) has been solved in conformity with the initial condition (19) and the boundary conditions (17) or (18), Eq. (13) enables us to obtain the expression for $c$ in terms of $x$ and $t$, which, in the cases discussed here, comes out in the form of a ratio of two infinite series. In the special case, $c\ll 1$, a simplified expression for $c$ can be obtained by the following considerations.

In both the cases considered here $q(l,t)=0$ for all values of $t$. Hence the relation (16) reduces to

$$w=A\exp\left[(t/4)-(x/2)+\int_l^x c\,dx\right],$$

whence

$$\exp\left[\int_l^x c\,dx\right]=A^{-1}\exp[-(t/4)+(x/2)]w.$$

If $cl$ is small compared to unity, the left-hand side may be set approximately equal to $1+\int_l^x c\,dx$, whence, on differentiation,

$$c=A^{-1}e^{-t/4}(e^{x/2}w)_x. \tag{20}$$

The expression for $c$ thus obtained will agree (for small $c$) with the solution of the simplified Eq. (9) to a first approximation, but not exactly.

Though the relation (20) has been derived by a process of approximation, it is interesting to note that, when substituted in the simplified Eq. (9), it transforms the latter *rigorously* into an equation of ordinary diffusion for $w$. The substitution (20) therefore leads to an alternative method for solving Eq. (9). Substituting Eq. (20) in Eq. (9) and integrating with respect to $x$, we have

$$w_{xx}=w_t+h(t)e^{-x/2}.$$

Here also the arbitrary function $h(t)$ can be set equal to zero without any loss of generality, so that we have, as in the previous case,

$$w_{xx}=w_t.$$

After these preliminary discussions it is easy to obtain the solutions in an explicit form with the help of the standard theory of linear differential equations. We shall discuss Problem (II) in some detail because of its greater practical importance.

## IV. DISCUSSION OF PROBLEM (II)

The discussion of Problem (II) is perhaps a little simplified by the introduction of a function $u$ defined by

$$u=w-\exp[\sigma^2t-\sigma x],$$

which satisfies the same differential equation,

$$u_{xx}=u_t, \tag{21}$$

but with the altered initial and boundary conditions,

$$u(x,0)=0,\quad \text{at } t=0, \tag{22}$$

$$\left.\begin{aligned} -u_x&=\sigma u, && \text{at } x=0\\ u_{xx}&=\tfrac{1}{4}u+(\tfrac{1}{4}-\sigma^2)\exp[\sigma^2t-\sigma l], && \text{at } x=l\end{aligned}\right\}. \tag{23}$$

Equation (21) is solved most conveniently by making use of a Laplace transformation.[9] We multiply both sides of Eqs. (21) and (23) by $e^{-\gamma t}$, where $\Re(\gamma)$ is sufficiently large, and integrate over $t$ from 0 to $\infty$, obtaining

$$v_{xx}-\gamma v=0, \tag{24}$$

$$\left.\begin{aligned} -v_x&=\sigma v, && \text{at } x=0\\ v_{xx}&=\tfrac{1}{4}v+e^{-\sigma l}\gamma(\tfrac{1}{4}-\sigma^2)/(\gamma-\sigma^2), && \text{at } x=l\end{aligned}\right\}, \tag{25}$$

where $v(x,\gamma)$ is the adjoint function,

$$\gamma^{-1}v(x,\gamma)=\int_0^\infty u(x,t)e^{-\gamma t}dt.$$

The solution of the ordinary differential equation (24), with the boundary conditions (25), is

$$v(x,\gamma)=\frac{\gamma(\frac{1}{4}-\sigma^2)e^{-\sigma l}}{(\gamma-\frac{1}{4})(\gamma-\sigma^2)}\cdot\frac{e^{-s(l-x)}+\epsilon(s)e^{-s(l+x)}}{1+\epsilon(s)e^{-2sl}}, \tag{26}$$

where $s=\sqrt{\gamma}$ and $\epsilon(s)=(s+\sigma)/(s-\sigma)$.

Though the function $s=\sqrt{\gamma}$ has a branch point at $\gamma=0$, it is easy to verify that $v(x,\gamma)$ (looked upon as a function of $\gamma$ alone) is a meromorphic function *devoid of branch points*. Its only singularities are simple poles at $\gamma=\frac{1}{4}$, $\gamma=\sigma^2$, and $\gamma=s_n^2$, where $s_n$ is a root of the equation

$$\exp(2s_nl)=(\sigma+s_n)/(\sigma-s_n); \tag{27}$$

that is, of the equation,

$$\tanh s_nl=s_n/\sigma. \tag{28}$$

At $\gamma=0$, however, there is usually no singularity. Roots of Eq. (28) occur in pairs of opposite signs. Depending on the value of $\sigma l$, there may or may not be any real[10] root other than the one at $s=0$; but there are always an infinite number of pure imaginary[11] roots. Moreover, an examination of Eq. (27) shows that there exists a real number $\alpha_0$ such that $\Re(s_n)<\alpha_0$ for all values of $n$.

The adjoint function $v(x,\gamma)$ is of the familiar type occurring in the theory of partial differential equations,

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[9] R. Courant and D. Hilbert, *Methoden der Mathematischen Physik*, Bd. II (Verlag. Julius Springer, Berlin, 1937).

[10] If $\sigma l<1$ (which includes the case $\sigma l<0$), there is no real root. If $\sigma l>1$, there is a pair of real roots $\pm s_r$ lying between $-\sigma$ and $+\sigma$. In this case $(s_r)^2<\sigma^2<\frac{1}{4}$. If $\sigma l=1$, there is a triple root at $s=0$, which must be taken into account in evaluating residues.

[11] It can be proved that Eq. (27) has no complex roots with nonvanishing real and imaginary parts. This circumstance considerably reduces the labor of numerical evaluation of the roots.

and satisfies all the conditions necessary for the validity of the inversion formula,

$$u(x, t)=(1/2\pi i)\int_{\alpha-i\infty}^{\alpha+i\infty}\gamma^{-1}v(x, \gamma)e^{\gamma t}d\gamma, \quad (29)$$

where the path of integration $L$ is a straight line parallel to the imaginary axis and lying in the region $\mathcal{R}(\gamma)>\frac{1}{4}$. The function $u(x, t)$ thus obtained is the required solution of the differential equation $u_{xx}=u_t$ satisfying Eqs. (22) and (23). To bring it into a form suitable for numerical work we must remove $\gamma$ by carrying out the integration on the right-hand side of Eq. (29). It can be easily verified that on a system of parabolic contours $C_n$, bounded on the right by the straight line $L$, and satisfying the equation $Im(s)=\pm n\pi/l$, this integral tends to zero as $n$ tends to infinity. The value of the integral on $L$ is therefore equal to $2\pi i$ times the sum of the residues at the poles of the integrand. The solution of Problem (II) is thus obtained in the form of an infinite series,

$$w(x, t)=\frac{\exp[-\sigma l+t/4][c_0e^{x/2}+(1-c_0)e^{-x/2}]}{[c_0e^{l/2}+(1-c_0)e^{-l/2}]} +\sum_n \frac{(\frac{1}{4}-\sigma^2)8s_n \exp[-\sigma l+s_n{}^2t] \sinh s_n(l-x)}{\sigma(1-4s_n{}^2)\{1-\sigma l(1-s_n{}^2/\sigma^2)\}}. \quad (30)$$

For the reasons previously stated the root $s=0$ makes no contribution to the series unless $\sigma l=1$. Since two roots $\pm s_n$ of opposite signs correspond to the same pole in the $\gamma$-plane, only one of them should be included in the summation.

The above solution assumes a particularly simple form if we start with a mixture of two isotopes in equal proportions.

The same procedure leads to the following solution[12] of Problem (I)

$$w(x, t)=e^{t/4}[(e^{c_0l}-1)e^{x/2}+(e^l-e^{c_0l})e^{-x/2}]/(e^l-1) +\sum_n \frac{2q_n(\frac{1}{4}-\sigma^2)}{l(\frac{1}{4}+q_n{}^2)(\sigma^2+q_n{}^2)}\cdot[1-(-1)^ne^{-\sigma l}] \times\exp(-q_n{}^2t)\sin q_nx,$$

where $q_n=n\pi/l$, and $n$ runs over all positive integral values.

## V. COMPARISON WITH EXPERIMENT

The solutions worked out in the preceding paragraphs hold for a single column only. In practice a number of columns are usually connected in series by tubes in which a convective circulation of the gas is maintained by non-uniform heating. These connecting tubes act like reservoirs and invariably cause delay in the establishment of equilibrium. This more general case does not lend itself so easily to an exact mathematical treatment. It is desirable that experiments should be performed with a single column over a wide range of concentrations for a more complete test of the theory of Furry and Jones. Meanwhile, we proceed to compare the results obtained here with the experimental data of Clusius and Dickel[2] on the rate of separation of oxygen from air. Their column had the following specifications:

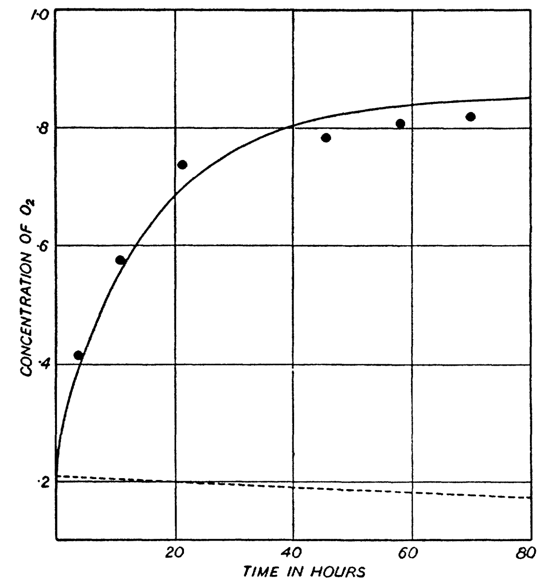


FIG. 1. Theoretical curve for $\alpha=0.014$ and the experimental points of Clusius and Dickel reference 2).

Length of the column = 295 cm;
Radius of the outer tube = 0.42 cm;
Radius of the hot wire = 0.02 cm;
Temperature of the outer tube = 293°K;
Temperature of the hot wire = 923°K.

The upper end of the column was connected with a large reservoir and the whole system filled with air at atmospheric pressure. In course of separation the concentration of $O_2$ in the reservoir fell from its normal value 0.209 to 0.174. The mean, 0.191, of these two is taken to be the initial concentration $c_0$. Since in the equilibrium condition the concentration of $O_2$ varied from 0.174 to 0.83 along the column, calculations are carried out for a mixture of $N_2$ and $O_2$ in equal proportions. For such a mixture[13] at 20°C:

The coefficient of viscosity[14,15] $\eta=1.88\times10^{-4}$ poise, and $\eta$ varies as $T^{0.756}$;

The density $\rho=1.25\times10^{-3}$ g/cm$^3$;

The coefficient of diffusion $D=1.44\eta/\rho$ cm$^2$/sec $=0.216$ cm$^2$/sec.

Making use of the tables and formulas given by Furry

[12] Substituting this solution in Eq. (16) and putting $x=0$, we get for all values of $t$ the expected result $\int_0^l cdx=c_0l$.

[13] The notation of reference 4 is used throughout this section.
[14] M. Trautz and K. G. Sorg, Ann. Physik 10, 81 (1931).
[15] M. Trautz and R. Heberling, Ann. Physik 10, 155 (1931).

and Jones[4] for a maxwellian gas and $\alpha$ independent of temperature, we then obtain the following values for the coefficients $H$, $K$, $\mu$ of the differential equation (3):

$$H/\alpha = 2.12\times 10^{-4} \text{ g/sec};$$
$$K_c = 0.443\times 10^{-4} \text{ g-cm/sec};$$
$$K_d = 2.28\times 10^{-4} \text{ g-cm/sec};$$
$$K = K_c + K_d = 2.72\times 10^{-4} \text{ g-cm/sec};$$
$$\mu = 0.487\times 10^{-3} \text{ g/cm}.$$

These values are substituted in the solution (30) and a series of curves drawn by giving different values to $\alpha$. Of these the curve for $\alpha = 0.014$ which fits best with the experimental points is reproduced in Fig. 1. This may be compared with the value 0.018 found by Waldmann[16] by another method.

In conclusion I wish to thank Professor M. N. Saha, F.R.S., for acquainting me with this subject and for his interest, Professor N. R. Sen for having kindly gone through the paper, Dr. U. C. Guha for his friendly cooperation in checking the calculations, and the National Institute of Sciences of India for the Fellowship.

[16] L. Waldmann, Z. Naturforsch. 1 (1946).

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# On the Definition and Approximation of Feynman's Path Integrals

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A general and compact expression for Feynman's path integral has been obtained. A classical method is given for the computation of such expressions. The example of a Dirac particle in a constant external electromagnetic field is treated by this method.

## I. INTRODUCTION

In order to treat problems involving action at a distance, Feynman has proposed a lagrangian form of quantum mechanics.[1] In this formulation the probability amplitude $K(x^B, x^A)$ for a particle to go from a space-time point $x^A$ to a space-time point $x^B$ is postulated to be given by an expression of the form:

$$K(x^B, x^A) = \int \exp(iS[x]/\hbar)d(\text{paths}), \tag{1}$$

the integral being extended over all paths, $x(\tau)$ from $x^A$ to $x^B$. In this paper we give a general and compact definition for this integral, and we give also a classical method for computing an approximate expression for it.

We make use of the following notation:

$x(\tau)$ is the parametric representation of a world line

$$x = x_\mu. \quad \mu = 1, 2, 3, 4. \quad \tau = \text{proper time}.$$

$\dot{x}(\tau) = dx(\tau)/d\tau$.
$\bar{x}(\tau)$ is the classical path.
$x^k \equiv x(\tau^k)$.
$\epsilon = \tau^{k+1} - \tau^k$.

$$S[x] = \int_{\tau^A}^{\tau} L\{x(\tau), \dot{x}(\tau)\} d\tau;$$

$S[x]$ is a functional of the function $x(\tau)$.
$\bar{S} = S[\bar{x}]$; $\bar{S}$ is the classical action.

## II. DEFINITION OF THE PATH INTEGRALS

In Feynmans' work[1] the definition of the path integrals involves an infinite product of "normalization factors." For his purposes Feynman determined these normalization factors in the cases in which the potential is velocity independent and gave their expressions in rectangular coordinates; and he indicated also the existence of a relationship between these factors and the action, $S$. We shall give here the general formula for the normalization factors valid for all actions and all frames of reference; moreover, we shall give a compact expression for the infinite product of the normalization factors. We shall give first the general formula for $K(x^{k+1}, x^k)$ for two points corresponding to an interval $\tau^{k+1} - \tau^k = \epsilon$ infinitesimally small; then we shall obtain $K(x^B, x^A)$ by iteration. (The essential formulas are given before their proofs.)

① $$K(x^{k+1}, x^k) = \exp[(i/\hbar)\bar{S}(x^{k+1}, x^k)] \times (2\pi\hbar i)^{-\frac{1}{2}l}(\det_{\mu\nu} a^{\mu\nu}{}_{k+1,k})^{\frac{1}{2}}. \tag{2}$$

Here $l$ is the number of degrees of freedom ($l = 4$ in the actual case),

$$a^{\mu\nu}{}_{k+1,k} \equiv \partial^2\bar{S}/\partial x_\mu{}^{k+1}\partial x_\nu{}^k$$

$\det_{\mu\nu}$ means the determinant with respect to the indices $\mu$ and $\nu$.

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† Now at Institut Henri Poincaré, Paris.
[1] R. P. Feynman, Revs. Modern Phys. 20, 367 (1948), hereafter called I. Following the suggestion made in paragraph 14 of I, we have defined a path $x(\tau)$ by four functions $x_\mu(\tau)$ of a parameter $\tau$; the formulas of I are still valid, the quantities $\psi(x, \tau) = \exp(iMc/2\hbar)\psi(x)$ replacing the wave function $\psi(x)$. A proof of this fact is given in connection with the example studied below. For a more complete study of a formalism of relativistic quantum mechanics introducing the wave function $\psi(x, \tau)$, see E. C. G. Stueckelberg, Helv. Phys. Acta 14, 588 (1941), and 15, 23 (1942).