THE EQUATIONS OF THE TRANSVERSE ELECTROMAGNETIC FIELD IN A MEDIUM

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Submitted March 2, 1969; resubmitted August 1, 1969

Zh. Eksp. Teor. Fiz. 58, 558–567 (February 1970)

We develop a perturbation theory for the case where a transverse electromagnetic field interacts with a medium; the interaction is assumed to be weak only as far as the medium is concerned. This theory enables us, on the one hand, to obtain integral equations for the observable microscopic quantities and, on the other hand, to write the kernel of these equations in the form of a power series in the coupling constant. We use this method to derive the Maxwell equation and the radiative transfer equation and we consider the coherent generation of harmonics process.

1. WE shall call a collection of nuclei and electrons for which we take the Coulomb interaction between them into account a Coulomb system (subsystem). In many cases of interest the following estimate holds for the interaction of a Coulomb system with a transverse magnetic field:

$$\langle \mathcal{H}_{\text{Coul. s.}} \rangle \gg \langle \mathcal{H}_{\text{field}} \rangle \sim \langle \mathcal{H}_{\text{int}} \rangle, \tag{1}$$

This shows that it is not possible to use the traditional perturbation theory, but this estimate leads us to expect that some kind of perturbation theory could be developed to apply to a Coulomb system.

Estimates such as (1) are satisfied also in a number of other cases, for instance, for the interaction of a transverse electromagnetic field, which in turn interacts with a Coulomb subsystem, with the spin degrees of freedom of nuclei and unpaired electrons. In those cases we shall call the system which plays the role of the Coulomb system the heavy system and the system which plays the role of the transverse field the light system. In the last example the transverse field is a heavy subsystem.

As a rule the above-mentioned fact is taken into account indirectly when one writes down phenomenological equations which contain as unknowns only quantities characterizing the light subsystem (Maxwell equations in a medium,[1] the Bloch equation and its variations describing the motion of the spin moment[2]). The heavy subsystem is then taken into account through the phenomenological parameters which enter into the equations (dielectric constant, chemical shift, spin-spin interaction[2-4]).

The phenomenological equations describe the observed phenomena adequately, but sometimes (rather often) they turn out to be insufficient and it is very important (especially when the interaction considered is used to study the structure of the heavy system) to make manifest the microscopic nature of the phenomenological parameters which are introduced.

As a rule this problem is solved using traditional perturbation theory[5-8] which is incorrect as we noted above. All the same, the results obtained in first approximation are valid. In the following we discuss this problem.

In the present paper we use the example of the interaction of a transverse electromagnetic field with a Coulomb system to develop a method which enables us to avoid the difficulties of the traditional perturbation theory. Using it we can not only obtain correctly expressions for the dielectric susceptibility but also derive the Maxwell equations for the transverse field themselves. Moreover, it makes it possible to obtain equations which describe nonlinear and incoherent processes (radiative transfer equations in dispersive media[9-11]).

The main point of this method consists in expressing the averaged perturbation theory series for the quantities in which we are interested not simply in terms of the average values of products of operators but in terms of specially selected combinations of them, the semi-invariants. The following relation can be used for the definition of a semi-invariant:

$$\begin{aligned}\langle B_1 B_2 \dots B_n \rangle &= \langle B_1 B_2 \dots B_{n-1} \rangle S_1(B_n) \\ &+ \sum_{1 \leqslant l_i \leqslant n-1} \langle B_1 \dots B_{l_i-1} B_{l_i+1} \dots B_{n-1} \rangle S_2(B_{l_i}, B_n) \\ &+ \dots + S_n(B_1, B_2, \dots, B_n).\end{aligned} \tag{2}$$

It is clear that

$$\begin{aligned}S_1(B_1) &= \langle B_1 \rangle, \\ S_2(B_1, B_2) &= \langle B_1 B_2 \rangle - \langle B_1 \rangle \langle B_2 \rangle.\end{aligned} \tag{3}$$

The semi-invariants have important properties. In fact, according to the principle of correlation damping, formulated by Bogolyubov[12] the averages of operator products taken at sufficiently distant points in space must be expressible in terms of the products of the corresponding averages. The same can probably also be said of the averages of operator products taken at points which are sufficiently far away from each other in time. In fact, the semi-invariants are just such combinations of averages that if their arguments are spaced out (in space or time) they asymptotically tend to zero.

The series obtained which can most conveniently be described by using diagram methods allow a partial summation which leads to integral equations either for the quantities of interest to us or for some auxiliary quantities. The kernels of these equations turn out to

be series with a well-known explicit form in the semi-invariants of the Coulomb system and in the coupling constant. The physical meaning of the kernels is completely obvious.

We shall assume here that the state of the interacting systems considered by us at finite times can be obtained by the adiabatic switching on of the interaction between the systems which are non-interacting at $-\infty$. This enables us to average independently operators of the Coulomb system and of the electromagnetic field. As far as the averaging of the operators of the transverse electromagnetic field is concerned, we must make one remark. The phenomenological Maxwell equations are valid assuming the electromagnetic field to be weak. The definition of a weak field does not meet with difficulties in the classical case. We generalize it for the quantal case by formally assigning a small parameter to each vector potential operator which stands under the average value or normal product sign. We must thus first of all expand in terms of normal products and neglect terms containing under the normal product sign more than a given number of operators.

We show how to expand any product of field operators in terms of normal products.

If U_α is a unitary operator that adds to the vector potential A(j) an arbitrary function $\alpha(j)$ satisfying the D'Alembert equation and the transversality condition[13]

$$U_\alpha^+ A(j) U_\alpha = A(j) + \alpha(j) \tag{4}$$

and

$$\prod_{j=1}^{n} A(j) = \sum_{l=0,\,\{j\}}^{n} P(j_1, j_2, \ldots, j_l) N(A(j_1), A(j_2) \ldots A(j_l)), \tag{5}$$

then

$$\left\langle U_\alpha^+ \prod_{j=1}^{n} A(j) U_\alpha \right\rangle = \sum_{l=0,\,\{j\}}^{n} P(j_1, j_2, \ldots, j_l)\, \alpha(j_1)\, \alpha(j_2) \ldots \alpha(j_l). \tag{6}$$

All other operators encountered in the present paper will, like the operator A(j) depend on the space-time coordinates (r, t) and on the tensor index α. We shall give these arguments a numerical index and to simplify the notation we shall write n instead of $\mathbf{r}_n$, t_n, α_n. The brackets $\langle\ \rangle_0$ indicate averaging over the vacuum state of the free electromagnetic field.

It follows from (6) that $P(j_1, j_2, \ldots, j_l)$ is the same as the vacuum average of the product of all operators A(j) except $A(j_1), A(j_2), \ldots, A(j_l)$. However,

$$\left\langle \prod_{j=1}^{n} A(j) \right\rangle = \sum_{l=1}^{n-1} \left\langle \prod_{\substack{j=1 \\ j\neq l}}^{n-1} A(j) \right\rangle \langle A(l) A(n) \rangle_0. \tag{7}$$

In the general case one can show that the expansion

$$F = \langle F \rangle_0 + \sum_{n=1}^{\infty} \frac{1}{n!} \frac{\delta}{\delta\alpha(1)} \cdots \frac{\delta}{\delta\alpha(n)} \langle U_\alpha^+ F U_\alpha \rangle_0 |_{\alpha=0} N(A(1) \ldots A(n)) \tag{8}$$

is valid for any functional F.

The method can be generalized to the case when the systems at $-\infty$ are in a state of thermodynamic equilibrium and there exists some non-equilibrium additional term to the electromagnetic field. The switching on of this extra term can be performed formally in Glauber's P-representation.[13, 14]

2. It is convenient for the expansion of the average of any operator in terms of the semi-invariants of the Coulomb system to start from the equations of quantum electrodynamics written down in such a form that the operators of the Coulomb system occur in the interaction representation while the operators of the transverse electromagnetic field are in the Heisenberg representation. The Heisenberg and interaction representations are defined, however, in such a way that the operators in those representations would be the same as the same operators in the Schrödinger representation, not at time t = 0, as is usually done, but at time $t = t^*$ which in the following we shall let go to $-\infty$. We shall assume that this limiting process has already taken place.

It is clear that

$$B^H(0) = S^+(t_0) B(0) S(t_0), \tag{9}$$

where B(0) is an arbitrary operator at time t_0 in the interaction representation and S(t) satisfies the equation

$$\frac{\partial S(t)}{\partial t} = -\frac{i}{\hbar} \mathcal{H}_{\text{int}}(t) S(t) \tag{10}$$

and the boundary condition $S(-\infty) = 1$.

If $C = C(\tilde{t}_1, \tilde{t}_2, \ldots, \tilde{t}_n)$, the following differential relation will hold by virtue of (2):

$$\frac{\partial}{\partial t'} (S^+(t') C S(t')) = -\frac{i}{\hbar} S^+(t') [C, \mathcal{H}_{\text{int}}(t')] S(t'), \tag{11}$$

and integrating it from $-\infty$ to t we get the following integral relation:

$$S^+(t) C S(t) = C - \frac{i}{\hbar} \int_{-\infty}^{t} dt'\, S^+(t') [C, \mathcal{H}_{\text{int}}(t')] S(t'). \tag{12}$$

Putting $C = B(\tilde{t})$ and $t = t_0$, we get

$$B^H(0) = B(0) - \frac{i}{\hbar} \int_{-\infty}^{t_0} dt_1\, S^+(t_1) [B(0), \mathcal{H}_{\text{int}}(t_1)] S(t_1). \tag{13}$$

The interaction Hamiltonian is[15]

$$\mathcal{H}_{\text{int}}(t) = -\frac{1}{c} \int d\mathbf{r}\, \mathbf{A}(\mathbf{r}, t)\, \mathbf{j}(\mathbf{r}, t) + \frac{1}{2} \int d\mathbf{r}\, \mathbf{A}^2(\mathbf{r}, t) \Psi(\mathbf{r}, t), \tag{14}$$

where

$$\mathbf{j}(\mathbf{r}, t) = \frac{1}{2} \sum_a \frac{e_a}{m_a} (\delta(\mathbf{r} - \mathbf{r}_a(t))\, \mathbf{p}_a(t) + \mathbf{p}_a(t) \delta(\mathbf{r} - \mathbf{r}_a(t))), \tag{15}$$

$$\Psi(\mathbf{r}, t) = \sum_a \frac{e_a^2}{m_a c^2} \delta(\mathbf{r} - \mathbf{r}_a(t)), \tag{16}$$

$$\operatorname{div} \mathbf{A}(\mathbf{r}, t) = 0, \tag{17}$$

A is the vector potential satisfying the commutator relations (11.6) of [15]. The summation in (15) and (16) is over all particles of the medium. We have intentionally not included the spin term in (15).

We write out Eq. (13) for the operator A and evaluating the commutator under the integral sign we "split off" the operator A using the fact that $S^+(t)CA(t)S(t) = S^+(t)CS(t)A^H(t)$ for any operator C. We apply the relation (12) to $S^+(t)CS(t)$. Extending this transformation to infinity we get

$$A^H(0) = A(0) - \frac{1}{c} D_0(0, 1) j(1) + D_0(0, 1) \sum_{n=2}^{\infty} (1, 2, \ldots, n) A^H(n) \ldots A^H(2), \tag{18}$$

where

$$(1, 2) = -\frac{i}{c^2 \hbar} [j(1), j(2)] \theta(t_1 - t_2) + \Psi(1) \delta(1, 2), \tag{19}$$

$$(1,2,3)=\frac{i}{c\hbar}[(1,2),j(3)]\theta(t_2-t_3)+\frac{i}{2c\hbar}[j(1),\Psi(2)]\theta(t_1-t_2)\delta(2,3),$$
$$(1,2,\ldots,n)=\frac{i}{c\hbar}[(1,2,\ldots,n-1),j(n)]\theta(t_{n-1}-t_n) \quad (20)$$

$$-\frac{i}{2\hbar}[(1,2,\ldots,n-2),\Psi(n-1)]\theta(t_{n-2}-t_{n-1})\delta(n-1,n), \quad (21)$$

$$D_0(0,1)=-\frac{i}{\hbar}\theta(t_0-t_1)[A(0),A(1)]. \quad (22)$$

We note that taking the A^H operators to the right or to the left one can put Eq. (18) into different forms, one of which is such that each term of the sum would be a Hermitian operator.

In exactly the same way we have for any operator u(0) of the medium

$$u^H(0)=u(0)+\sum_{n=1}^{\infty}(0,1,\ldots,n)_u A^H(n)\ldots A^H(1), \quad (23)$$

where

$$(0,1)_u=\frac{i}{c\hbar}\theta(t_0-t_1)[u(0),j(1)], \quad (24)$$

$$(0,1,2)_u=\frac{i}{c\hbar}\theta(t_1-t_2)[(0,1)_u,j(2)]$$
$$-\frac{i}{2\hbar}\theta(t_0-t_1)\delta(1,2)[u(0),\Psi(1)], \quad (25)$$

$$(0,1,\ldots,n)_u=\frac{i}{c\hbar}\theta(t_{n-1}-t_n)[(0,\ldots,n-1)_u,j(n)]$$
$$-\frac{i}{2\hbar}\theta(t_{n-2}-t_{n-1})\delta(n-1,n)[(0,\ldots,n-2)_u,\Psi(n-1)]. \quad (26)$$

Equation (18) has a simple physical meaning: if we forget about the quantum-mechanical nature of the quantities occurring in it, the right-hand side is the sum of the unperturbed field, the field induced by free currents, and the field induced by induced currents.

The convenience of writing the equations of motion in the form (18), (23) is connected with the peculiarities of the operators (1, 2, ... , n) and $(1, 2, \ldots, n)_u$. The structure of these operators is such that when expanding the averages of their products in terms of semi-invariants only those terms remain which contain under the semi-invariant sign exactly one of the terms of each such operator. This, in turn, means that when expanding in terms of semi-invariants the above-mentioned operators can be considered as being primary ones on par with the local operators j and Ψ. For instance, the quantity

$$S_2((1,2,\ldots,n_1),(1,2,\ldots,n_2))$$
$$=\langle(1,2,\ldots,n_1)(1,2,\ldots,n_2)\rangle-\langle(1,2,\ldots,n_1)\rangle\langle(1,2,\ldots,n_2)\rangle$$

also tends to zero when the distance between any of its arguments increases.

3. The equations of motion (18) and (23) can now be used to expand the averages of any operators of interest to us in a series. These series are essentially series in the coupling constant. They differ from similar series which are obtained by iterating the equations of motion written in the traditional form in the grouping of the terms. In the new form the series can very easily be expressed in terms of the semi-invariants of the Coulomb system and expanded in terms of normal products of the operators of the free electromagnetic field. It is then convenient to use diagram techniques. The retarded Green function of the free electromagnetic field is represented by a solid line. Each term of this series obtained through iterating Eq. (18), except the

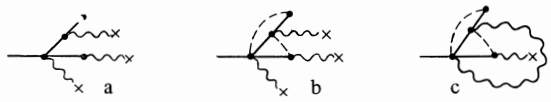

FIG. 1

first one, starts with a solid line which "ends up" in the operator (1, 2, ... , n) or $-c^{-1}j(1)$ depicted by a thick dot. The diagram can be broken off in this point and in that case the point corresponds to the operator $-c^{-1}j(1)$, or n − 1 lines may start from it to the right and the point then corresponds to the operator (1, 2, ... , n). To each end of a solid line abutting in the point we assign an argument 1, 2, 3, ... , n in counterclockwise order. If the operator A ends in the point we depict it by a wavy line with a cross at the other end. In Fig. 1a we have shown in diagram language the term

$$D_0(0,1)(1,2,3,4)D_0(4,5)(5,6,7)D_0(7,8)\left(-\frac{1}{c}j(8)\right)$$
$$\times D_0(3,9)(9,10)A(6)A(10)A(2).$$

We shall call such a diagram an operator diagram.

When writing down products of Heisenberg operators the order from top to bottom corresponds to the order from left to right in the analytical form.

Performing the average using the density matrix of the medium we express the averages of operator products in terms of semi-invariants in accordance with the rules defined by Eq. (2). In Fig. 1b we give the diagram corresponding to the term

$$D_0(0,1)S_2\left((1,2,3,4),-\frac{1}{c}j(8)\right)D_0(4,5)S_2((5,6,7),(9,10))$$
$$\times D_0(7,8)D_0(3,9)A(6)A(10)A(2).$$

We shall call such a diagram an operator diagram with respect to the electromagnetic field. The dotted line connects operators combined in the semi-invariant sign. The number of operators connected by it shows the order of the semi-invariant.

When expanding in a series in normal products of the operators of the electromagnetic field we must be led by the rules defined by Eqs. (5) and (7). A wavy line ending at both ends in operators of the medium corresponds to the quantity $\langle A(1)A(2)\rangle_0$. The operators of the electromagnetic field which in this case correspond to wavy lines with a cross at the end are taken under the normal product sign. In Fig. 1c we have given the diagram for

$$D_0(0,1)S_2\left((1,2,3,4),-\frac{1}{c}j(8)\right)D_0(4,5)S_2((5,6,7),(9,10))$$
$$\times D_0(7,8)D_0(3,9)\langle A(6)A(2)\rangle_0\langle A(10)\rangle.$$

4. We now apply the technique developed here to derive the linear Maxwell equations. We assume that there is no coherent radiation of the medium and that therefore the sum of all terms which do not contain the vector potential operators under the normal product sign vanish.

We must sum only those terms in the expansion of $\langle A^H(0)\rangle$ which contain under the normal product sign up to one A operator. We can write this sum in the form of Fig. 2 in which a block depicted by a circle is the sum

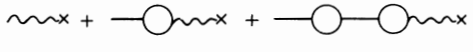

FIG. 2

FIG. 3

of all diagrams which have one incoming and one outgoing end which cannot be divided into a block of the same structure by cutting one D_0 line. This block we shall denote by $\alpha(1, 2)$. It is clear that in that case

$$\langle A^r(0)\rangle = \langle A(0)\rangle + D_0(0, 1)\alpha(1, 2)\langle A^r(2)\rangle. \qquad (27)$$

Equation (27) is just the linear Maxwell equation for the transverse electromagnetic field. Its Green function which we shall represent by a thick line (Fig. 3) and which we shall denote by D(0, 1) satisfies the equation

$$D(0, 1) = D_0(0, 1) + D_0(0, 2)\alpha(2, 3)D(3, 1) = \\ = D_0(0, 1) + D(0, 2)\alpha(2, 3)D_0(3, 1). \qquad (28)$$

The block $\alpha(1, 2)$ is defined by the series, the lowest terms of which are given in Fig. 4.

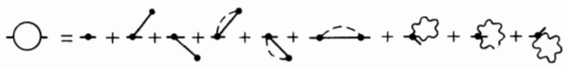

FIG. 4

It is now clear why the expressions for the transverse dielectric susceptibility evaluated in the first approximation of the usual perturbation theory are valid. Indeed, the correction to the average of the current, evaluated in first approximation, corresponds to the second term in Fig. 2 without the D_0 line. We can thus in this way obtain the first term in the expansion of Fig. 4 but it is impossible to obtain the other terms.

We note also that Eq. (27) turns out to be valid for any Fourier components of the transverse electromagnetic field and not only for the long-wavelength ones as in the case for the Maxwell equations for the total (transverse and longitudinal) field.

5. The coherent generation of harmonics is in lowest order described by those terms in the expansion for $\langle A^H(0)\rangle$ which contain two A operators under the normal product sign. These terms can be most conveniently summed by splitting off beforehand the block of diagrams whose lines are not connected by a single dotted line (Fig. 5). After this the contribution of all diagrams considered can be described as shown in Fig. 6, where the V_1 block has two incoming ends and one outgoing one and it is impossible to separate from it a block of the same structure by cutting one or two D lines. The α_2 block has two incoming and two outgoing ends and it is impossible to split off from it a block of the same structure by cutting two D lines or a block of the structure of v_1 by cutting one D line. The first terms of the expansions of v_1 and α_2 are given in Fig. 9.

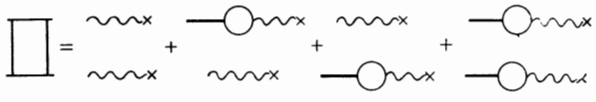

FIG. 5

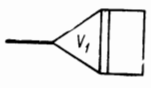

FIG. 6

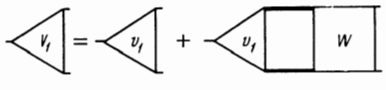

FIG. 7

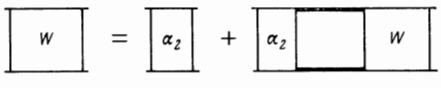

FIG. 8

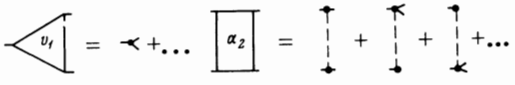

FIG. 9

We must note especially that the coherent generation of harmonics is described by additional terms in the average of the potential and not by non-linear Maxwell equations.

6. We now derive the radiative transfer equations which describe the propagation of incoherent radiation in a medium. If we drop in the expansion for $\langle A^H(0)A^H(0')\rangle$ the terms which do not contain the A operators under the normal product sign (it is easy to take them into account) and assume that the coherent component vanishes we need only sum terms containing under the normal product sign two A operators. The sum of these terms can conveniently be written in the form of Fig. 10. We can express the V_2 block in terms of v_2 (Fig. 11), where the v_2 block has two outgoing and one incoming ends and it is impossible to separate from it a block of the same structure by cutting one or two D lines.

If the original radiation contained a coherent component we should include in the sum of Fig. 10 the term of Fig. 12 which describes the scattering of coherent radiation. The last term in Fig. 10 clearly describes the same process but after the coherent generation of harmonics. If we neglect these terms we note easily that the sum of Fig. 10 satisfies the equation of Fig. 13. This is a linear Bethe–Salpeter type integral equation and it describes the radiative transfer processes and takes into account all possible light scattering effects. If the scattering is small and can be neglected there remains in the sum of Fig. 10 only the first term. It is clear that to evaluate it it is sufficient to know the Green function of Eq. (27) or, what amounts to the same, we can first of all solve Eq. (27) and afterwards average

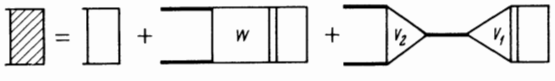

FIG. 10

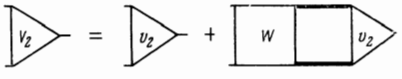

FIG. 11

FIG. 12

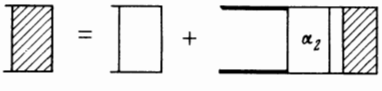

FIG. 13

the result obtained. In other words, under such conditions we can consider the equation

$$A^r(0) = A(0) + D_0(0, 1)\alpha(1, 2)A^r(2) \qquad (29)$$

as the equation for the operator of the dressed transverse electromagnetic field.

We note that we can obtain from the radiative transfer equations in the form of Fig. 13 equations for the Stokes vector parameter. It has four independent components which are functions of the wave vector, the coordinate and the frequency. This parameter is directly connected with the left-hand side of the equation of Fig. 13. The latter is symmetric under a permutation of the ends as follows from the symmetry under the same permutation of the first term on the right-hand side and the symmetry of the α_2 block under a simultaneous permutation of incoming and outgoing ends. By virtue of this symmetry and of the transversality condition the left-hand side of the equation of Fig. 13 has only four independent components which are functions of $\mathbf{r}_1, t_1$ and $\mathbf{r}_2, t_2$. If we change to the variables $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$, $\mathbf{R} = \frac{1}{2}(\mathbf{r}_1 + \mathbf{r}_2)$, and $t = t_1 - t_2$ (stationary process) and perform a Fourier transformation with respect to $\mathbf{r}$ and t the corresponding independent components which have the same number as the number of components of the Stokes parameter turn out to be functions of the same variables as the Stokes parameter. Further transformations of the equations are performed assuming that the properties of the medium and of the parameters we are looking for depend only weakly on R. We feel, however, that the equation of Fig. 13 and the quantities occurring in it have a more direct physical meaning than the Stokes parameters and the corresponding equations.

7. One can consider in similar fashion also other processes connected with the interaction of a transverse radiation with a medium. We have considered only the simplest ones. Probably one can consider the procedure proposed here as that form of perturbation theory which might be expected from the strong inequality sign in Eq. (1). It differs from the traditional perturbation theory in that series in the coupling constant can be constructed not for any quantity, but for some selected quantities. This procedure shows up those selected quantities and establishes their connection with other quantities.

The author expresses his gratitude to V. M. Andreev, L. V. Keldysh, and A. V. Lazarev for discussions of the results of this paper.

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Translated by D. ter Haar
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