COMPLEMENT A_{IV}

ELEMENTARY INTRODUCTION TO THE ELECTRIC DIPOLE HAMILTONIAN

We consider an ensemble of charged particles forming a localized system with spatial extension of the order of a. This is the case, for example, in atoms or molecules made up of electrons and nuclei in bound states whose spatial dimensions are of the order of a few Bohr radii. If one such system interacts with radiation with wavelength λ large with respect to a, it is legitimate to neglect the spatial variation of the electromagnetic field over the expanse of the system. This approximation, called the *long-wavelength approximation*, has been used in §B.3 of this chapter to get a simpler equivalent formulation of electrodynamics for a localized system of charges coupled to an external field. The corresponding transformation, the Göppert-Mayer transformation, has been presented as a change of Lagrangian or a gauge change. We again treat the same problem here ($\S A_{IV}.1$) by directly studying the unitary transformation which permits one to get the electric dipole Hamiltonian $\mathbf{E} \cdot \mathbf{r}$ by starting with the usual Hamiltonian $\mathbf{A} \cdot \mathbf{p}$ (*). Next we generalize the Göppert-Mayer transformation to the case where the electromagnetic field is treated, not as an external field, but as a quantized system with its proper dynamics ($\S A_{IV}.2$). Finally, we look at some possible extensions of the method used here ($\S A_{IV}.3$).

1. The Electric Dipole Hamiltonian for a Localized System of Charges Coupled to an External Field

a) The Unitary Transformation Suggested by the Long-Wavelength Approximation

Let $\mathbf{A}_e(\mathbf{r}, t)$ be the potential vector describing the external radiation (the scalar potential $U_e(\mathbf{r}, t)$ is assumed to be zero). The long-wavelength approximation involves neglecting the spatial variation of $\mathbf{A}_e(\mathbf{r}, t)$ in the Hamiltonian. Thus one can replace $\mathbf{A}_e(\mathbf{r}_\alpha, t)$ by $\mathbf{A}_e(\mathbf{R}, t)$ in the kinetic energy term, where $\mathbf{R}$ is a point taken in the interior of the system of charges. In the following, we take $\mathbf{R}$ as a fixed point, which we choose as the origin of the coordinates $\mathbf{R} = \mathbf{0}$, which amounts to ignoring all the displacements of the atom or molecule as a whole (see, however, the Remark in $\S A_{IV}.1.b$ below). Under these conditions, the Hamiltonian is

(*) Since we are not using here the Lagrangian formalism, reading this complement does not require knowledge of the ideas introduced in Chapters II and IV.

written

$$H(t) = \sum_{\alpha} \frac{1}{2\,m_\alpha} [\mathbf{p}_\alpha - q_\alpha\, \mathbf{A}_e(\mathbf{0}, t)]^2 + V_{\text{Coul}} \tag{1}$$

where V_{Coul} is the Coulomb energy of the system.

The simple form which the kinetic energy term takes in the long-wavelength approximation suggests the application of a unitary transformation $T(t)$ which translates each operator $\mathbf{p}_\alpha$ by an amount $q_\alpha \mathbf{A}_e(\mathbf{0}, t)$:

$$T(t)\, \mathbf{p}_\alpha\, T^+(t) = \mathbf{p}_\alpha + q_\alpha\, \mathbf{A}_e(\mathbf{0}, t)\,. \tag{2}$$

The translation operator $T(t)$ which effects this transformation is given by

$$\begin{aligned} T(t) &= \exp\left[-\frac{\mathrm{i}}{\hbar} \sum_{\alpha} q_\alpha\, \mathbf{r}_\alpha \cdot \mathbf{A}_e(\mathbf{0}, t) \right] \\ &= \exp\left[-\frac{\mathrm{i}}{\hbar}\, \mathbf{d} \cdot \mathbf{A}_e(\mathbf{0}, t) \right] \end{aligned} \tag{3}$$

where

$$\mathbf{d} = \sum_{\alpha} q_\alpha\, \mathbf{r}_\alpha \tag{4}$$

is the electric dipole moment of the charge distribution with respect to the origin. Equation (3) coincides with that found in Part B above from the Lagrangian formalism [see Equation (B.34)].

Remark

We have not included in (1) the interaction terms of the particle spin magnetic moments with the magnetic field of the external radiation. These terms are actually smaller than the interaction terms in $\mathbf{A}_e \cdot \mathbf{p}$ by a factor of the order of $\hbar k/p$ [see Equation (D.15), Chapter III], that is to say, of the order of a/λ, since $\hbar/p \sim a$. They are thus of the same order of magnitude as the interaction terms which have been neglected by replacing $\mathbf{A}_e(\mathbf{r}_\alpha, t)$ with $\mathbf{A}_e(\mathbf{0}, t)$.

b) The Transformed Hamiltonian

In this new representation, the temporal evolution of the transformed state vector $|\psi'(t)\rangle = T(t)|\psi(t)\rangle$ is governed by the Hamiltonian

$$H'(t) = T(t)\, H(t)\, T^+(t) + \mathrm{i}\hbar \left[\frac{\mathrm{d}T(t)}{\mathrm{d}t} \right] T^+(t)\,. \tag{5}$$

By using (1) and (2), we find

$$T(t)\, H(t)\, T^+(t) = \sum_{\alpha} \frac{\mathbf{p}_\alpha^2}{2\,m_\alpha} + V_{\text{Coul}} \tag{6}$$

and by using (3)

$$i\hbar\left[\frac{dT(t)}{dt}\right] T^{+}(t) = \mathbf{d} \cdot \dot{\mathbf{A}}_e(\mathbf{0}, t) = -\mathbf{d} \cdot \mathbf{E}_e(\mathbf{0}, t) \tag{7}$$

which gives finally

$$H'(t) = \sum_{\alpha} \frac{\mathbf{p}_\alpha^2}{2m_\alpha} + V_{\text{Coul}} - \mathbf{d} \cdot \mathbf{E}_e(\mathbf{0}, t)\,. \tag{8}$$

We have thus found that in the long-wavelength approximation, the interaction with the external field is simply described in this new representation by a coupling term between the dipole moment $\mathbf{d}$ of the atom and the external electric field evaluated at the position of the atom.

Remark

It is possible to take into account the global motion of the atom and to refer the positions of the charges q_α to a point $\mathbf{R}$ which, rather than being a fixed point, is taken at the center of mass of the atom (see Exercise 3). One then finds that if the total charge $Q = \sum_\alpha q_\alpha$ is zero, Equation (8) remains valid provided that one replaces $\mathbf{0}$ by $\mathbf{R}$ in $\mathbf{E}_e(\mathbf{0}, t)$. In contrast, if the system is an ion ($Q \neq 0$), new terms appear in the Hamiltonian $H'(t)$. They describe the coupling of the global motion of the ion to the external field $\mathbf{A}_e$. In all that follows in this complement, we only consider globally neutral systems:

$$Q = \sum_{\alpha} q_\alpha = 0\,. \tag{9}$$

c) The Velocity Operator in the New Representation

In the initial description, the velocity of the particle is represented by the operator

$$\mathbf{v}_\alpha = \frac{1}{m_\alpha}\left[\mathbf{p}_\alpha - q_\alpha \mathbf{A}_e(\mathbf{0}, t)\right] \tag{10}$$

while in the new description it is represented by

$$\mathbf{v}'_\alpha = T(t)\, \mathbf{v}_\alpha\, T^{+}(t) \tag{11.a}$$

which, using (2) and (10), is equal to

$$\mathbf{v}'_\alpha = \frac{\mathbf{p}_\alpha}{m_\alpha}\,. \tag{11.b}$$

Thus, in the new description we find a much simpler relation between the momentum and the velocity.

2. The Electric Dipole Hamiltonian for a Localized System of Charges Coupled to Quantized Radiation

We now consider the radiation field as a quantized system with its own dynamics.

a) The Unitary Transformation

If the coupling between particles and radiation mainly involves the modes whose wavelength is large with respect to a, we can, in a first approximation, neglect the contribution of the other modes. The operator

$$\mathbf{A}(\mathbf{r}_\alpha) = \sum_j \mathscr{A}_{\omega_j}[a_j\,\boldsymbol{\varepsilon}_j\,e^{i\mathbf{k}_j\cdot\mathbf{r}_\alpha} + a_j^+\,\boldsymbol{\varepsilon}_j\,e^{-i\mathbf{k}_j\cdot\mathbf{r}_\alpha}] \tag{12}$$

can then be replaced by $\mathbf{A}(\mathbf{0})$, since for all the modes taken into account $|\mathbf{k}_j\cdot\mathbf{r}_\alpha| \ll 1$. In this approximation, the Hamiltonian in the Coulomb gauge is written

$$H = \sum_\alpha \frac{1}{2m_\alpha}[\mathbf{p}_\alpha - q_\alpha\,\mathbf{A}(\mathbf{0})]^2 + V_{\text{Coul}} + \sum_j \hbar\omega_j\left(a_j^+\,a_j + \frac{1}{2}\right). \tag{13}$$

An argument similar to that presented in §A_{IV}.1 suggests that one apply to (13) the unitary transformation

$$T = \exp\left[-\frac{i}{\hbar}\sum_\alpha q_\alpha\,\mathbf{r}_\alpha\cdot\mathbf{A}(\mathbf{0})\right] = \exp\left[-\frac{i}{\hbar}\mathbf{d}\cdot\mathbf{A}(\mathbf{0})\right] \tag{14}$$

which, while resembling (3), differs from it. In (14), $\mathbf{A}(\mathbf{0})$ is a time-independent field operator, while $\mathbf{A}_e(\mathbf{0}, t)$ in (3) is a classical function of time. It is indeed interesting in what follows to reexpress (14) with the aid of the operators a_j and a_j^+ using the expansion (12) for $\mathbf{A}(\mathbf{0})$. We then get

$$T = \exp\left\{\sum_j(\lambda_j^*\,a_j - \lambda_j\,a_j^+)\right\} \tag{15}$$

with

$$\lambda_j = \frac{i}{\sqrt{2\,\varepsilon_0\,\hbar\omega_j\,L^3}}\,\boldsymbol{\varepsilon}_j\cdot\mathbf{d}\,. \tag{16}$$

b) Transformation of the Physical Variables

Consider first the operators relating to the particles. Since $\mathbf{r}_\alpha$ commutes with T, this operator represents the position of the particle in both descriptions. As for the velocity, since the operator $\mathbf{A}(\mathbf{0})$ acts like a

c-number with respect to the particles, we get in a fashion identical to (10) and (11)

$$\mathbf{v}'_\alpha = T\mathbf{v}_\alpha T^+ = \frac{\mathbf{p}_\alpha}{m_\alpha}. \tag{17}$$

We now examine the field operators. Note first that the quantities λ_j introduced in (16) are purely atomic operators commuting between themselves and can then be considered as numbers with respect to the radiation operators a_j and a_j^+. The operator T in the form (15) thus appears as a translation operator for a_j and a_j^+ [see (C.61) and (C.66) of Chapter III]:

$$\begin{cases} Ta_j T^+ = a_j + \lambda_j & (18.a) \\ Ta_j^+ T^+ = a_j^+ + \lambda_j^*. & (18.b) \end{cases}$$

We are going to use these relations to calculate the operator $\mathbf{E}'_\perp(\mathbf{r})$ describing the transverse field in the new representation. Starting from the expression for the transverse electric field in the Coulomb gauge,

$$\mathbf{E}_\perp(\mathbf{r}) = \mathrm{i} \sum_j \mathscr{E}_{\omega_j}[a_j \boldsymbol{\varepsilon}_j \mathrm{e}^{\mathrm{i}\mathbf{k}_j\cdot\mathbf{r}} - a_j^+ \boldsymbol{\varepsilon}_j \mathrm{e}^{-\mathrm{i}\mathbf{k}_j\cdot\mathbf{r}}] \tag{19}$$

we find

$$\begin{aligned} \mathbf{E}'_\perp(\mathbf{r}) &= T\mathbf{E}_\perp(\mathbf{r}) T^+ \\ &= \mathrm{i} \sum_j \mathscr{E}_{\omega_j}[(a_j + \lambda_j) \boldsymbol{\varepsilon}_j \mathrm{e}^{\mathrm{i}\mathbf{k}_j\cdot\mathbf{r}} - (a_j^+ + \lambda_j^*) \boldsymbol{\varepsilon}_j \mathrm{e}^{-\mathrm{i}\mathbf{k}_j\cdot\mathbf{r}}] \\ &= \mathbf{E}_\perp(\mathbf{r}) - \sum_{\mathbf{k}_j\boldsymbol{\varepsilon}_j} \left[\frac{1}{2\,\varepsilon_0 L^3} \boldsymbol{\varepsilon}_j(\boldsymbol{\varepsilon}_j \cdot \mathbf{d}) \mathrm{e}^{\mathrm{i}\mathbf{k}_j\cdot\mathbf{r}} + c.c. \right] \end{aligned} \tag{20}$$

where $\mathbf{E}_\perp(\mathbf{r})$ is the mathematical operator given in (19). The last term of (20) will be interpreted later. Consider finally the magnetic field. Since $\mathbf{A}(\mathbf{r})$ and $\mathbf{A}(\mathbf{r}')$ commute for all $\mathbf{r}$ and $\mathbf{r}'$ [see (A.13) of Chapter III], $\mathbf{B}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r})$ commutes also with $\mathbf{A}(\mathbf{r}')$. It follows that $\mathbf{A}(\mathbf{r})$ and $\mathbf{B}(\mathbf{r})$ commute with T. In particular, the magnetic field operator retains the same form in both representations:

$$\mathbf{B}'(\mathbf{r}) = T\mathbf{B}(\mathbf{r}) T^+ = \mathbf{B}(\mathbf{r}). \tag{21}$$

c) Polarization Density and Displacement

To interpret the last term of (20) physically and to identify the variable represented in the new representation by the mathematical operator $\mathbf{E}_\perp(\mathbf{r})$ given in (19), it is convenient to describe the localized system of charges

by a polarization density $\mathbf{P}(\mathbf{r})$, and then, starting from the electric field and the polarization density $\mathbf{P}(\mathbf{r})$, to introduce the displacement $\mathbf{D}(\mathbf{r})$. For a more complete discussion of this problem not limited to the lowest order in a/λ as here, the interested reader should refer to §C.1 of Chapter IV, which can be read independently of the rest of that chapter.

The charge density associated with the system of localized charges q_α is written in real space as

$$\rho(\mathbf{r}) = \sum_\alpha q_\alpha \, \delta(\mathbf{r} - \mathbf{r}_\alpha) \tag{22.a}$$

and in reciprocal space as

$$\rho(\mathbf{k}) = \left(\frac{1}{2\pi}\right)^{3/2} \sum_\alpha q_\alpha \, e^{-i\mathbf{k}\cdot\mathbf{r}_\alpha} . \tag{22.b}$$

Since the charges are localized near the origin ($|\mathbf{r}_\alpha| \lesssim a$) and we are assuming that the coupling with the radiation takes place substantially with the long-wavelength modes ($ka \ll 1$), it is reasonable to expand the exponential of (22.b) and to take only the first nonzero term (a lowest-order calculation in a/λ). One then gets using (9) and (4)

$$\rho(\mathbf{k}) = -\left(\frac{1}{2\pi}\right)^{3/2} i\mathbf{k} \cdot \mathbf{d} \tag{23.a}$$

and by Fourier transformation

$$\rho(\mathbf{r}) = -\nabla \cdot [\mathbf{d}\, \delta(\mathbf{r})] . \tag{23.b}$$

Equation (23.b) suggests one introduce the *polarization density*

$$\mathbf{P}(\mathbf{r}) = \mathbf{d}\, \delta(\mathbf{r}) \tag{24.a}$$

corresponding to a dipole $\mathbf{d}$ localized at $\mathbf{r} = \mathbf{0}$, as well as its spatial Fourier transform

$$\mathscr{P}(\mathbf{k}) = \left(\frac{1}{2\pi}\right)^{3/2} \mathbf{d} . \tag{24.b}$$

Equation (23.b) is then written

$$\rho(\mathbf{r}) = -\nabla \cdot \mathbf{P}(\mathbf{r}) . \tag{25}$$

The simple form of Equation (25) shows that if one introduces the displacement $\mathbf{D}(\mathbf{r})$ defined by

$$\mathbf{D}(\mathbf{r}) = \varepsilon_0 \mathbf{E}(\mathbf{r}) + \mathbf{P}(\mathbf{r}) , \tag{26}$$

where $\mathbf{E}(\mathbf{r})$ is the total electric field, then Maxwell's equation $\nabla \cdot \mathbf{E} = \rho/\varepsilon_0$ and Equation (25) imply

$$\nabla \cdot \mathbf{D}(\mathbf{r}) = 0 \tag{27.a}$$

which shows that $\mathbf{D}(\mathbf{r})$ is a transverse field. Equation (27.a) can be rewritten using (26):

$$\mathbf{D}(\mathbf{r}) = \mathbf{D}_\perp(\mathbf{r}) = \varepsilon_0 \mathbf{E}_\perp(\mathbf{r}) + \mathbf{P}_\perp(\mathbf{r}). \tag{27.b}$$

The importance of $\mathbf{D}(\mathbf{r})$ rests with the fact that outside the origin $\mathbf{P}(\mathbf{r})$ is zero [see (24.a)], with the result that, from (26), $\mathbf{D}(\mathbf{r})$ coincides with $\varepsilon_0 \mathbf{E}(\mathbf{r})$. Thus, $\mathbf{D}(\mathbf{r})$ is a *transverse field which, outside the system of charges, coincides with the total electric field* to within a factor ε_0. Since the electric field is purely retarded, it follows that the displacement outside the atom is a retarded transverse field.

Return now to Equation (20). By transforming the discrete sum of the last line into an integral we get

$$-\frac{1}{\varepsilon_0} \int d^3k \sum_{\varepsilon \perp \mathbf{k}} \frac{1}{(2\pi)^3} \varepsilon(\varepsilon \cdot \mathbf{d})\, e^{i\mathbf{k}.\mathbf{r}}. \tag{28}$$

Comparison with (24.b) shows that (28) is just the Fourier transform of $-\mathscr{P}_\perp(\mathbf{k})/\varepsilon_0$, so that Equation (20) can be written

$$\mathbf{E}'_\perp(\mathbf{r}) = \mathbf{E}_\perp(\mathbf{r}) - \frac{1}{\varepsilon_0} \mathbf{P}_\perp(\mathbf{r}). \tag{29}$$

Finally we calculate the operator $\mathbf{D}'(\mathbf{r})$ which represents the displacement in the new description. Using (27.b), $\mathbf{D}'(\mathbf{r})$ is written

$$\mathbf{D}'(\mathbf{r}) = T\mathbf{D}(\mathbf{r})\, T^+ = \varepsilon_0\, T\mathbf{E}_\perp(\mathbf{r})\, T^+ + T\mathbf{P}_\perp(\mathbf{r})\, T^+. \tag{30}$$

Using (19), (20), (29), and the fact that $\mathbf{P}_\perp(\mathbf{r})$ commutes with T, one then gets

$$\frac{1}{\varepsilon_0} \mathbf{D}'(\mathbf{r}) = i \sum_j \mathscr{E}_{\omega_j} [a_j \varepsilon_j e^{i\mathbf{k}_j.\mathbf{r}} - a_j^+ \varepsilon_j e^{-i\mathbf{k}_j.\mathbf{r}}]. \tag{31}$$

It appears then that the *same* mathematical operator, namely the linear combination of a_j and a_j^+ on the right in (31), describes two *different* physical variables, depending on the representation used: the transverse electric field in the initial representation, and the displacement (divided by ε_0) in the new one. The advantage of the latter representation is that the simple operator (31) describes a transverse field which outside the atom is purely retarded.

d) THE HAMILTONIAN IN THE NEW REPRESENTATION

The Hamiltonian H' in the new representation is given by

$$H' = THT^{+}. \tag{32}$$

Here we are going to find the T-transformed expression for H given in (13). The physical interpretation of the results will then be obtained by attributing to the operators appearing in the expression for H' the physical meaning which they have in the new representation. The transform of the first term of (13) is simply

$$\sum_{\alpha} \frac{\mathbf{p}_{\alpha}^2}{2\, m_{\alpha}} \tag{33}$$

and according to (17) represents the kinetic energy of the particles. The second term of (13) is an atomic operator which depends only on the positions $\mathbf{r}_{\alpha}$ of the particles and is therefore unchanged in the transformation. It remains to find the transform of the third term of (13), that is, the operator called H_R in the initial representation and which describes in that representation the energy of the transverse field. Using (18), we get

$$\begin{aligned} H_R' &= TH_R\, T^{+} = T \sum_j \hbar\omega_j \left(a_j^{+}\, a_j + \frac{1}{2} \right) T^{+} \\ &= \sum_j \hbar\omega_j \left[(a_j^{+} + \lambda_j^{*})(a_j + \lambda_j) + \frac{1}{2} \right] \\ &= H_R + \sum_j \hbar\omega_j (\lambda_j\, a_j^{+} + \lambda_j^{*}\, a_j) + \sum_j \hbar\omega_j\, \lambda_j^{*}\, \lambda_j . \end{aligned} \tag{34}$$

In addition to the operator H_R we get a linear operator in λ_j and λ_j^{*} and a bilinear operator in λ_j and λ_j^{*}. Consider first the linear term, which can be written according to (16) as

$$-\,\mathbf{d} \cdot \sum_j \mathrm{i} \sqrt{\frac{\hbar\omega_j}{2\, \varepsilon_0\, L^3}}\, (a_j\, \boldsymbol{\varepsilon}_j - a_j^{+}\, \boldsymbol{\varepsilon}_j) . \tag{35}$$

We get the scalar product of $\mathbf{d}$ (which is equal to $\mathbf{d}'$, since $\mathbf{r}_{\alpha}' = \mathbf{r}_{\alpha}$) with an operator coincident with $\mathbf{D}'(0)/\varepsilon_0$. The operator appearing in (35) must indeed be interpreted as the displacement (divided by ε_0), since we are studying the Hamiltonian in the new representation. Thus for the term of H_R' linear in a_j and a_j^{+}, we get

$$-\,\mathbf{d} \cdot \frac{\mathbf{D}'(\mathbf{0})}{\varepsilon_0} . \tag{36}$$

Consider finally the last term of (34). According to (16), it is equal to

$$\sum_j \hbar\omega_j \lambda_j^* \lambda_j = \sum_{\mathbf{k}_j \varepsilon_j} \frac{1}{2 \varepsilon_0 L^3} (\boldsymbol{\varepsilon}_j \cdot \mathbf{d})^2 . \tag{37}$$

This term depends only on the operator $\mathbf{d}$ (equal to $\mathbf{d}'$). Physically it represents a dipole self-energy of the system, denoted ε_{dip}. The expression (37) for ε_{dip} seems to diverge. It fact, one must limit the sum to the values of k_j for which the long-wavelength approximation is valid.

By regrouping the various terms, one gets the following expression for H':

$$H' = \sum_\alpha \frac{\mathbf{p}_\alpha^2}{2 m_\alpha} + V_{\text{Coul}} + \varepsilon_{\text{dip}} + \sum_j \hbar\omega_j \left(a_j^+ a_j + \frac{1}{2} \right) - \mathbf{d} \cdot \frac{\mathbf{D}'(\mathbf{0})}{\varepsilon_0} . \tag{38}$$

The structure of the new Hamiltonian H' is very simple. One has first a purely atomic Hamiltonian representing the sum of the kinetic energy, the Coulomb energy, and the dipole self-energy. Then one has a proper radiation Hamiltonian H_R, and finally, an electric dipole interaction Hamiltonian coupling the dipole moment of the system of charges to the displacement at the origin $\mathbf{0}$. The fact that the new interaction Hamiltonian is purely linear in the field and no longer has quadratic terms (such as H_{I2} examined in §D.1 of Chapter III) is an important advantage of this new description.

Remark

In the new representation, the energy of the transverse field is given by H_R' and differs from H_R by the last two terms in (34). Since the operators $\mathbf{D}'(\mathbf{r})/\varepsilon_0$ (31) and $\mathbf{B}'(\mathbf{r})$ (21) have the same mathematical form in the new description as $\mathbf{E}(\mathbf{r})$ and $\mathbf{B}(\mathbf{r})$ in the earlier one, we have

$$H_R = \frac{1}{2} \int \mathrm{d}^3 r \left[\frac{\mathbf{D}'^2(\mathbf{r})}{\varepsilon_0} + \frac{\mathbf{B}'^2(\mathbf{r})}{\mu_0} \right] \tag{39}$$

(where $\varepsilon_0 c^2$ is replaced by $1/\mu_0$).

3. Extensions

We are now going to examine two possible extensions of the treatment above.

a) The Case of Two Separated Systems of Charges

Consider two systems of charges $\mathscr{S}_A$ and $\mathscr{S}_B$ localized about well-separated points $\mathbf{R}_A$ and $\mathbf{R}_B$, each of the systems being neutral. The transfor-

mation T which generalizes (14) is

$$T = \exp\left\{ -\frac{i}{\hbar}\left[\mathbf{d}_A \cdot \mathbf{A}(\mathbf{R}_A) + \mathbf{d}_B \cdot \mathbf{A}(\mathbf{R}_B)\right] \right\} \tag{40}$$

$\mathbf{d}_A$ and $\mathbf{d}_B$ being the respective dipole moments.

The Hamiltonian in the Coulomb gauge describing this system is in the long-wavelength approximation equal to

$$\begin{aligned} H = & \sum_\alpha \frac{1}{2m_\alpha}\left[\mathbf{p}_\alpha - q_\alpha \mathbf{A}(\mathbf{R}_A)\right]^2 + \sum_\beta \frac{1}{2m_\beta}\left[\mathbf{p}_\beta - q_\beta \mathbf{A}(\mathbf{R}_\beta)\right]^2 \\ & + V_{\text{Coul}}^{AA} + V_{\text{Coul}}^{BB} + V_{\text{dip dip}}^{AB} \\ & + \sum_i \hbar\omega_i\left(a_i^+ a_i + \frac{1}{2}\right) \end{aligned} \tag{41}$$

where V_{Coul}^{AA} (V_{Coul}^{BB}) is the Coulomb energy of the system of charges $\mathscr{S}_A$ ($\mathscr{S}_B$), and $V_{\text{dip dip}}^{AB}$ is the electrostatic interaction energy between the dipoles $\mathbf{d}_A$ and $\mathbf{d}_B$ of the two systems of charges.

The Hamiltonian $H' = TH'T^+$ in the new representation is gotten with the aid of the transformation (40), and one finds it equal to

$$\begin{aligned} H' = & \sum_\alpha \frac{\mathbf{p}_\alpha^2}{2m_\alpha} + V_{\text{Coul}}^{AA} + \varepsilon_{\text{dip}}^A + \sum_\beta \frac{\mathbf{p}_\beta^2}{2m_\beta} + V_{\text{Coul}}^{BB} + \varepsilon_{\text{dip}}^B \\ & + H_R - \mathbf{d}'_A \cdot \frac{\mathbf{D}'(\mathbf{R}_A)}{\varepsilon_0} - \mathbf{d}'_B \cdot \frac{\mathbf{D}'(\mathbf{R}_B)}{\varepsilon_0}. \end{aligned} \tag{42}$$

The structure of the new Hamiltonian H' is very simple. We have first of all two purely atomic Hamiltonians for $\mathscr{S}_A$ and $\mathscr{S}_B$, representing in each case the sum of the kinetic energy, the Coulomb energy (inside $\mathscr{S}_A$ or $\mathscr{S}_B$), and the dipole self-energy. Then, we have a radiation Hamiltonian H_R, and finally, an electric dipole interaction Hamiltonian coupling the dipoles of $\mathscr{S}_A$ and $\mathscr{S}_B$ to the displacement at $\mathbf{R}_A$ and $\mathbf{R}_B$. The important point is that there are no longer instantaneous electrostatic interaction terms between the systems in (42). The term $V_{\text{dip dip}}^{AB}$ has in fact been compensated for by the dipole terms (37), which contain, besides the self-energy terms $\varepsilon_{\text{dip}}^A$ and $\varepsilon_{\text{dip}}^B$, cross terms simultaneously involving $\mathbf{d}_A$ and $\mathbf{d}_B$. The corresponding calculations are not detailed here, since they are treated again in the general case in Complement C_{IV}. One can however understand this point by noting that the new Hamiltonian contains the coupling between the electric dipole moment $\mathbf{d}'_A$ of $\mathscr{S}_A$ and the total displacement at $\mathbf{R}_A$, $\mathbf{D}'(\mathbf{R}_A)$, which includes in particular the displacement generated at $\mathbf{R}_A$ by $\mathscr{S}_B$. Now the displacement generated by

$\mathscr{S}_B$ outside $\mathscr{S}_B$ coincides with the total electric field generated by $\mathscr{S}_B$ outside $\mathscr{S}_B$. It follows that the coupling term $-\mathbf{d}'_A \cdot \mathbf{D}'(\mathbf{R}_A)/\varepsilon_0$ of (42) contains the interaction of $\mathscr{S}_A$ with the transverse electric field as well as the longitudinal field created by $\mathscr{S}_B$ at $\mathbf{R}_A$.

b) The Case of a Quantized Field Coupled to Classical Sources

In §$A_{IV}.1$ above we have examined the coupling of a system of quantized particles with a classical field. The opposite problem, which we now treat, is that of a quantized field interacting with classical currents. The Hamiltonian in Coulomb gauge for such a system has been given in Complement B_{III} [Equation (7)]:

$$H = H_R - \int d^3r\, \mathbf{j}_{cl}(\mathbf{r}, t) \cdot \mathbf{A}(\mathbf{r}) . \tag{43}$$

The current $\mathbf{j}_{cl}(\mathbf{r}, t)$ is produced by classical particles with charge q_α whose positions and velocities are described by the classical functions $\mathbf{r}_\alpha(t)$ and $\dot{\mathbf{r}}_\alpha(t)$, with a given time dependence

$$\mathbf{j}_{cl}(\mathbf{r}, t) = \sum_\alpha q_\alpha \dot{\mathbf{r}}_\alpha(t)\, \delta(\mathbf{r} - \mathbf{r}_\alpha(t)) . \tag{44}$$

If the charges creating the current are localized near the origin in a volume of dimension a and if one considers only the dynamics of the modes with wavelength long with respect to a, it is possible to simplify the interaction Hamiltonian

$$H_I = - \int d^3r\, \mathbf{j}_{cl}(\mathbf{r}, t) \cdot \mathbf{A}(\mathbf{r}) = - \sum_\alpha q_\alpha \dot{\mathbf{r}}_\alpha(t) \cdot \mathbf{A}(\mathbf{r}_\alpha) \tag{45}$$

by replacing $\mathbf{A}(\mathbf{r}_\alpha)$ with its value at the origin:

$$H_I \simeq - \sum_\alpha q_\alpha \dot{\mathbf{r}}_\alpha(t) \cdot \mathbf{A}(\mathbf{0}) = - \dot{\mathbf{d}}(t) \cdot \mathbf{A}(\mathbf{0}) \tag{46}$$

$\mathbf{d}(t)$ being the electric dipole moment of the charge distribution, equal to $\sum_\alpha q_\alpha \mathbf{r}_\alpha(t)$. We then apply to the Hamiltonian

$$H = H_R - \dot{\mathbf{d}}(t) \cdot \mathbf{A}(\mathbf{0}) \tag{47}$$

a unitary transformation similar to the transformations in §§$A_{IV}.1$ and $A_{IV}.2$ and aimed at giving rise to an electric dipole interaction term. Consider the transformation

$$T = \exp\left\{ - \frac{i}{\hbar} \mathbf{d}(t) \cdot \mathbf{A}(\mathbf{0}) \right\} \tag{48}$$

analogous to (3) and (14), but where $\mathbf{d}(t)$ is now a classical function

depending on time and $\mathbf{A}(\mathbf{0})$ an operator acting on the radiation variables. The transformed Hamiltonian is given by Equation (5). The calculation of TH_RT^+ is identical to that presented in (34) and gives

$$TH_R\,T^+ = H_R - \mathbf{d}(t) \cdot \frac{\mathbf{D}'(\mathbf{0})}{\varepsilon_0} + \varepsilon_{\text{dip}} \tag{49}$$

where $\mathbf{D}'(\mathbf{0})/\varepsilon_0$ corresponds to the operator of (31) evaluated at $\mathbf{r} = \mathbf{0}$ and where ε_{dip} is now a classical function of time gotten by replacing the operator $\mathbf{d}$ of (37) with the function $\mathbf{d}(t)$. Here TH_IT^+ is unchanged, since H_I commutes with T. Finally the term in $i\hbar(\partial T/\partial t)T^+$ is equal to $\dot{\mathbf{d}}(t) \cdot \mathbf{A}(0)$ and thus offsets H_I [see (46)]. The new Hamiltonian is then

$$H = H_R - \mathbf{d}(t) \cdot \frac{\mathbf{D}'(\mathbf{0})}{\varepsilon_0} + \varepsilon_{\text{dip}}\,. \tag{50}$$

The last term here is a classical function of time and can eventually be dropped.

Note that, just as in $\S A_{IV}.2$, $\mathbf{D}'(\mathbf{r})$ represents the displacement. The interaction between the quantized field and the classical sources is thus, in the new representation, proportional to the product of the classical electric dipole with the displacement at the point about which the dipole is localized.