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Gauge-invariant resolution of the controversy over length versus velocity forms of the interaction with electric dipole radiation

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The controversy over whether to use the length or the velocity form of the interaction of electric dipole radiation with atoms is resolved on the basis of gauge invariance. When the unperturbed Hamiltonian is chosen to be the atomic Hamiltonian, the condition that the probability amplitudes are gauge invariant implies that the interaction is of the length form, $-q\vec{E}\cdot\vec{r}$, not the velocity form, $-(q/mc)\vec{A}\cdot\vec{p}$. Hartree-Fock theory is examined and shown to be form invariant under local gauge transformations, i.e., gauge invariant. A comparison is made between the length and velocity forms of the interaction and oscillator strengths. If the true Hamiltonian is nonlocal, only the length form of the dipole oscillator strength is valid. If the true Hamiltonian is local, the length form of the dipole oscillator strength may be transformed to the velocity form.

I. INTRODUCTION

Ever since Chandrasekhar[1] showed that the oscillator strengths for electric dipole transitions in atoms calculated from the exact wave functions could be written in several apparently equivalent forms, there has been a controversy over which form to use for approximate calculations. Chandrasekhar[1] advocated the use of the velocity form for calculations using variationally determined wave functions. Others have shown that in many situations the length form is superior.[2,3] A rich lore has developed with many "rules of thumb" over when to use the length form and when to use the velocity form.[4] Some understanding of these "rules of thumb" has been given by Anderson and Weinhold[5] using qualitative considerations of upper and lower bounds.[6] However, the situation is still more of an art than a science, since there are many factors that must be taken into account in deciding which form to use and the decision cannot be made unambiguously.

On the other hand, Starace[7] has applied the principle of minimal electromagnetic coupling to various approximate Hamiltonians with nonlocal potentials. These Hamiltonians give different values for the dipole length and velocity oscillator strengths. He has come to the conclusion that a length form of the interaction should be used.[8] He presumably shows that the Hartree-Fock (HF) equations are not gauge invariant.[7,9,10] When the principle of minimal electromagnetic coupling is used to make the HF equations gauge invariant, an additional interaction with the electromagnetic field is induced.[11,12] This interaction converts the original "velocity" interaction into a "length" form of the interaction. Starace[7] claims that his length form is the only physically correct interaction for Hamiltonians with nonlocal potentials.

Starace's recommendation has been criticized for not telling whether the "length" or the "velocity" form of the oscillator strength is closer to the experimentally observed value for a particular transition.[13,14] Starace has responded[15] that the value of the velocity form is irrelevant, since "once one has started by choosing an approximation procedure (for nonlocal potentials)... only the length formula should be used."

In this paper we show that on grounds of gauge invariance it is indeed the *length* form of the *interaction* that should be used The gauge-invariant formulation of the interaction of radiation and matter[16] shows that in order to use the atomic Hamiltonian as the unperturbed Hamiltonian for *electric dipole transitions*, it is necessary to make a gauge transformation to the gauge in which the vector potential is negligible and the scalar potential is the electric dipole potential, $-\vec{E}\cdot\vec{r}$.[17] It is this "length" form of the interaction that is then treated by time-dependent perturbation theory, in contrast to another "length" form advocated by Starace[7] and Lin.[8]

Much confusion has arisen in the literature by associating the length form of the dipole oscillator strength with the length form of the interaction, and the velocity form of the dipole oscillator strength with the velocity form of the interaction. On the basis of gauge-invariance arguments we show that it is only the length form of the *interaction* which should be used. For a nonlocal Hamiltonian, this means that only the length form of the dipole oscillator strength is valid. However, if the true Hamiltonian is local, the length form of the dipole oscillator strength may be transformed to the velocity form. If a nonlocal model Hamiltonian, like the HF Hamiltonian, is used to evalu-

ate these expressions, different results are obtained. Which form gives better agreement with experiment is then a question of detailed computation. However, the velocity form of the dipole oscillator strength must be derived from the length form, which emerges naturally from the length form of the interaction. The length form of the dipole oscillator strength may, in this sense, be considered as more fundamental than the velocity form.

Contrary to several authors,[7,9,10] we also show that the HF equations are in fact gauge invariant. In order to remove the dependence of the HF equations on the electromagnetic field, the gauge transformation mentioned previously must be made which gives the length form of the electric dipole interaction as the perturbation.

In Sec. II the gauge invariance of many-body wave mechanics is discussed to review concepts and establish the notation. Then in Sec. III the gauge-invariant formulation of the interaction of electromagnetic radiation and matter is given, based on the gauge invariance of transition probability amplitudes. In Sec. IV the HF equations are shown to be gauge invariant. The nonlocal HF exhange potential is shown to lead nonuniquely to a velocity-dependent potential in Sec. V. When the principle of minimal electromagnetic coupling is used in this velocity-dependent potential, it is made gauge invariant. However, in the electric dipole approximation the dependence on the electromagnetic field cancels out. In Sec. VI the different length and velocity interactions are discussed and compared. The different forms of the oscillator strengths are discussed in Sec. VII. Finally, the conclusions are given in Sec. VIII.

II. GAUGE INVARIANCE OF THE MANY-BODY SCHRÖDINGER EQUATION

In order to provide some background and to establish the notation, the gauge invariance of the many-body Schrödinger equation is discussed. The form invariance of the many-body Schrödinger equation under local gauge transformations is a straightforward generalization of the treatment for a single particle.[18] It is this form invariance of the equation that we will call the "gauge invariance" of the equation. In order to clarify the basic ideas, we will use wave mechanics instead of the second-quantization formalism used by Lin.[9]

The Schrödinger equation for a system of N identical fermions in a *time-dependent* electromagnetic field characterized by the vector potential $\vec{A}$ and scalar potential A_0 is

$$H(\vec{A},A_0)\Psi = i\hbar\frac{\partial\Psi}{\partial t}\,. \tag{2.1}$$

The antisymmetric wave function $\Psi=\Psi(x_1,\ldots,x_N;t)$ depends on the coordinates x_i $(i=1,2,\ldots,N)$, where $x_i=(\vec{r}_i,\sigma_i)$ denote the spatial coordinates $\vec{r}_i$ and spin σ_i of particle i. The Hamiltonian $H(\vec{A},A_0)$ for fermions of mass m and charge q is

$$H(\vec{A},A_0)=\sum_{i=1}^{N}\left(\frac{1}{2m}\left[\vec{p}_i-\frac{q}{c}\vec{A}(\vec{r}_i,t)\right]^2+qA_0(\vec{r}_i,t)+V_1(x_i)\right)+\frac{1}{2}\sum_{i\neq j}^{N}V_2(x_i,x_j)\,, \tag{2.2}$$

where $V_1(x)$ is the static, external potential energy. For an atom, for example, the external potential energy is the Coulomb potential between an electron and the nucleus. The two-body interaction $V_2(x,y)$ between a particle at the point x and a particle at the point y is assumed to be symmetric, $V_2(x,y)=V_2(y,x)$. For an atom, the two-body potential is the Coulomb repulsion between the electrons.

A local gauge transformation of the first kind[19] can be made on the wave function

$$\Psi'(x_1,\ldots,x_N;t)=\exp\left(i\frac{q}{\hbar c}\sum_{1}^{N}\Lambda(\vec{r}_i,t)\right)\Psi(x_1,\ldots,x_N;t)\,, \tag{2.3}$$

where $\Lambda(\vec{r},t)$ is an arbitrary, differentiable, real function of space and time. The Schrödinger equation in Eq. (2.1) is form invariant under the gauge transformation in Eq. (2.3),

$$H(\vec{A}',A_0')\Psi'=i\hbar\frac{\partial\Psi'}{\partial t}\,. \tag{2.4}$$

The new vector potential $\vec{A}'$ is related to the old one $\vec{A}$ by

$$\vec{A}'=\vec{A}+\nabla\Lambda\,, \tag{2.5}$$

and the new scalar potential A_0' is related to the old one A_0 by

$$A_0'=A_0-\frac{\partial\Lambda}{\partial(ct)}\,, \tag{2.6}$$

which are the usual gauge transformations of the second kind[19] in electrodynamics. A comparison of Eq. (2.4) with Eq. (2.2) shows that the Schrödinger equation is form invariant under the local gauge transformation in Eq. (2.3). This "form invariance under local gauge transformations" is considered here to by synonymous with the "gauge invariance" of the Schrödinger equation.

III. GAUGE INVARIANCE OF PROBABILITY AMPLITUDES

A gauge-invariant formulation of the interaction of electromagnetic radiation with matter has been developed by Yang.[16] His approach is summarized

here, since the ideas are essential to an understanding of the resolution of the "length" versus "velocity" controversy. The transition probabilities must be independent of the choice of gauge. If the transition-probability amplitudes are independent of the gauge, the transition probabilities will be also. By choosing the basis functions in terms of which the true wave function is expanded in an appropriate way, the transition-probability amplitudes can be made invariant. It is commonly thought that the gauge can be chosen arbitrarily and that the unperturbed or basis determining Hamiltonian can also be chosen arbitrarily. Yang[16] showed that making the formulation gauge invariant requires the basis-determining Hamiltonian to be the quantum-mechanical energy operator. The energy operator is $H(\vec{A}, 0)$, i.e., the Hamiltonian without the scalar potential of the time-dependent field.[20] Yang[16] has shown that the energy operator obeys the correspondence principle. The time derivative of its expectation value is the expectation value of the quantum-mechanical power operator.

The eigenvalue problem for the energy operator is

$$H(\vec{A}, 0)\Psi_n = E_n\Psi_n , \tag{3.1}$$

where the energy eigenvalue E_n and the eigenstate Ψ_n in general depend on the time through the vector potential $\vec{A} = \vec{A}(\vec{r}, t)$. In order to develop a gauge-invariant formulation of the interaction of electromagnetic radiation with matter, Yang[16] expanded the true wave function in Ψ in terms of the eigenstates in Eq. (3.1),

$$\Psi = \sum_n c_n\Psi_n . \tag{3.2}$$

When this wave function is substituted into the Schrödinger equation in Eq. (2.1), we obtain

$$i\hbar\dot{c}_n - E_n c_n = \sum_m \langle\Psi_n| \left(\sum_1^N qA_0(\vec{r}_i, t) - i\hbar\frac{\partial}{\partial t}\right)\Psi_m\rangle\, c_m . \tag{3.3}$$

The time derivative appears on the right-hand side because the eigenstates Ψ_n depend on time through the vector potential $\vec{A}$ in Eq. (3.1). Under the local gauge transformation in Eq. (2.3) on the wave functions, the coefficients c_n in Eq. (3.2) are unchanged. The matrix elements in Eq. (3.3) are also gauge invariant,

$$\langle\Psi_n'| \left(\sum_1^N qA_0'(\vec{r}_i, t) - i\hbar\frac{\partial}{\partial t}\right)\Psi_m'\rangle = \langle\Psi_n| \left(\sum_1^N qA_0(\vec{r}_i, t) - i\hbar\frac{\partial}{\partial t}\right)\Psi_m\rangle , \tag{3.4}$$

under the local gauge transformation in Eq. (2.3). Therefore, Eq. (3.3) for the probability amplitudes is gauge invariant.

The case of electric dipole radiation is considered now in which (a) magnetic effects are negligible and (b) the electric field is slowly varying over the dimensions of the atom. Then the new gauge in Eq. (2.5) can be chosen such that the new vector potential vanishes,[17]

$$\vec{A}' = \vec{A} + \nabla\Lambda = 0 . \tag{3.5}$$

Of course, electromagnetic radiation requires both the electric and magnetic fields for its existence. The choice of gauge in Eq. (3.5) is applicable only to the description of the effect of the electromagnetic radiation on the atom in the electric dipole approximation. The effect of magnetic and higher-order electric multipoles on the atom is neglected. When Eq. (3.5) is solved for Λ, and substituted into Eq. (2.6) for the new scalar potential, we obtain

$$A_0'(\vec{r}, t) = -\vec{E}(0, t)\cdot\vec{r} , \tag{3.6}$$

where the electric field of the electromagnetic radiation $\vec{E}(0, t)$ is evaluated at the origin. In its effect on the atom in the electric dipole approximation, the field of the electromagnetic radiation can be described by a scalar potential only. For magnetic multipoles the vector potential is, of course, required. The electric field $\vec{E}(\vec{r}, t)$ must of course be computed from Maxwell's equations which involves the magnetic field $\vec{B}(\vec{r}, t)$. However, for the interaction of this electromagnetic radiation with the atom in the electric dipole approximation, only the electric field at the atom $\vec{E}(0, t)$ is required, and it is derivable from the scalar potential in Eq. (3.6).

With the choice of gauge given in Eqs. (3.5) and (3.6) the Hamiltonian in Eq. (2.2) becomes

$$H(\vec{A}', A_0') = H(0, 0) - \sum_1^N q\vec{E}(0, t)\cdot\vec{r}_i . \tag{3.7}$$

The Hamiltonian in the absence of the electromagnetic field is $H(0, 0) = H_0$. The eigenvalue problem for the energy operator in Eq. (3.1) in this gauge becomes

$$H_0\Phi_n = E_n\Phi_n , \tag{3.8}$$

where $H(\vec{A}', 0) = H(0, 0) = H_0$ is the atomic Hamiltonian in the absence of the electromagnetic field and $\Psi_n' = \Phi_n$ are its eigenfunctions with eigenvalues E_n. The states Φ_n are not time dependent because the atomic Hamiltonian H_0 is not time dependent.

With the choice of gauge in Eqs. (3.5) and (3.6), Eq. (3.3) reduces to

$$i\hbar\dot{c}_n - E_n c_n = -\sum_m \vec{E}(0, t)\cdot\langle\Phi_n| \sum_1^N q\vec{r}_i\,|\Phi_m\rangle\, c_m . \tag{3.9}$$

The transition probability amplitudes in the electric dipole approximation are thus calculated from the length formula.

The conclusion then is that *if eigenstates of the atomic Hamiltonian in Eq. (3.8) are to be used for the basis states, the electric dipole interaction in the "length" form in Eq. (3.7) is the proper perturbation.* This form of the Hamiltonian is commonly used in practice.[21] Since questions have been raised about whether to use the length or velocity form of the interaction in approximate calculations involving nonlocal potentials, Hartree-Fock theory is examined in Sec. IV.

IV. GAUGE INVARIANCE OF THE HARTREE-FOCK EQUATIONS

Equation (3.1) is an N-body "time-independent" Schrödinger equation. One way in which it can be solved approximately is to use Hartree-Fock theory,[22] in which the wave functions Ψ_n are approximated by Slater determinants, $\det\{\phi_i(x_j)\}$, where $\phi_i(x)$ is a single-particle function for the state i. The Rayleigh-Ritz energy variational principle can be applied to the ground-state expectation value of the energy operator $H(\vec{A}, 0)$ calculated with a Slater determinant as a trial wave function. This procedure leads to the Hartree-Fock equations.

The HF equations derived by such a procedure for N identical fermions in a time-dependent electromagnetic field are[23]

$$\frac{1}{2m}\left(\vec{p}-\frac{q\vec{A}}{c}\right)^2\phi_\alpha(x)+V_1(x)\phi_\alpha(x)+\int dy\, V_2(x,y)\rho_0(y,y)\phi_\alpha(x)-\int dy\, V_2(x,y)\rho_0(y,x)\phi_\alpha(y)=\epsilon_\alpha\phi_\alpha(x). \quad (4.1)$$

A particle in the single-particle state α has the wave function $\phi_\alpha(x)$ and eigenenergy ϵ_α. The integrals in Eq. (4.1) include both integration over the spatial coordinates and summations over the spin. The single-particle reduced density matrix $\rho_0(y,x)$ is defined as

$$\rho_0(y,x)=\sum_1^N \phi_i^*(y)\phi_i(x), \quad (4.2)$$

where the sum is over the first N occupied orbitals. The first integral in Eq. (4.1) is the direct potential due to all the other particles while the second integral is the exchange potential. Several authors[7,9,10] have said that the HF exchange potential is not gauge invariant, but it appears that they have not transformed the density matrix properly.

If a local gauge transformation of the first kind[19] is made on the orbitals $\phi_\alpha(x)$, where $x=(\vec{r},\sigma)$, the new orbital

$$\phi'_\alpha(x)=\exp[iq\Lambda(\vec{r},t)/\hbar c]\phi_\alpha(x) \quad (4.3)$$

is obtained, where $\Lambda(\vec{r},t)$ is an arbitrary, differentiable, real function of the space coordinate $\vec{r}$ and time t, but not of spin σ. For the sake of conciseness, we will write $\Lambda(\vec{r},t)=\Lambda(x)$ in Eq. (4.3) with the stipulation that it does not depend on the spin, but does depend on the time. Under the gauge transformation of the first kind in Eq. (4.3), the single-particle reduced density matrix in Eq. (4.2) transforms as

$$\rho'_0(y,x)=\exp\{i(q/\hbar c)[\Lambda(x)-\Lambda(y)]\}\rho_0(y,x). \quad (4.4)$$

If Eqs. (4.3) and (4.4) are solved for ϕ_α and ρ_0 in terms of ϕ'_α and ρ'_0, and substituted into Eq. (4.1), the equation becomes

$$\frac{1}{2m}\left(\vec{p}-\frac{q\vec{A}'}{c}\right)^2\phi'_\alpha(x)+V_1(x)\phi'_\alpha(x)+\int dy\, V_2(x,y)\rho'_0(y,y)\phi'_\alpha(x)-\int dy\, V_2(x,y)\rho'_0(y,x)\phi'_\alpha(y)=\epsilon_\alpha\phi'_\alpha(x), \quad (4.5)$$

where the new vector potential $\vec{A}'$ is related to the old one by Eq. (2.5). Equation (4.5) has the same form as Eq. (4.1). The HF equation is therefore form invariant under the local gauge transformation in Eq. (4.3), which is what we mean by the gauge invariance of a Schrödinger-like equation.

Thus, in spite of the HF exchange potential being nonlocal, the HF Hamiltonian for the system $H_{HF}(\vec{A},0)=\sum_i h_i(\vec{A},0)$, where $h_i(\vec{A},0)$ is the single-particle HF Hamiltonian, can serve as an approximation to the energy operator for the system in Eq. (3.1). The approximation may or may not be very good, but at least it retains the proper symmetry properties under gauge transformations.

On the other hand, Starace[7] has shown that a nonlocal potential is in general not form invariant under local gauge transformation[11] and must be made gauge invariant. His proof is valid for nonlocal potentials that do not depend on the wave functions. However, in the HF case, the exchange potential does depend on the orbitals through the density matrix in Eq. (4.2). In Sec. V we consider Starace's procedure.

V. VELOCITY DEPENDENCE OF THE HARTREE-FOCK EXCHANGE POTENTIAL

Starace[7] showed that in general a nonlocal potential is not gauge invariant. In order to make it gauge invariant, he rewrote it as a velocity-dependent (i.e., momentum-dependent) potential.[24] He then used the *principle of minimal electromag-*

netic coupling, in which the momentum operator $\vec{p}$ is replaced by $\vec{p}-q\vec{A}/c$, to make it gauge invariant.[11,12] An additional interaction with the electromagnetic field was thus induced. When an expansion was made, and only the first two terms retained, he found that the additional interaction with the electromagnetic field could be incorporated into a "length" form of the interaction.

Since the HF exchange potential is already gauge invariant, as shown in Sec. IV, it is not necessary to change it to obtain a gauge-invariant expression.[25] However, for the sake of illustrating the procedure used by Starace[7] and others,[12] we show in this section that it can be applied in a modified form to the HF exchange potential to obtain a velocity-dependent potential. The principle of minimal electromagnetic coupling is then applied to make it gauge invariant. If the electric dipole approximation can be made, in which the vector potential depends only on the time, it is shown that the additional dependence on the electromagnetic field which is induced just cancels out. We are thus left with the original velocity-dependent potential before the principle of minimal electromagnetic coupling was applied. This velocity-dependent potential is equal to the original nonlocal exchange potential, so the procedure has not changed the equations.

The HF exchange potential in Eq. (4.1) is a nonlocal potential,

$$[V_{ex}\phi_\alpha](\vec{r},\sigma)=\int d^3r'\,V_2(\vec{r},\vec{r}')\times\sum_1^N\phi_i^*(\vec{r}',\sigma')\phi_i(\vec{r},\sigma)\phi_\alpha(\vec{r}',\sigma'), \tag{5.1}$$

where summation on repeated spin indices is implied. Any nonlocal potential can be rewritten in terms of a velocity-dependent (i.e., momentum-dependent) potential.[24] However, for a potential that depends on the orbitals, the prescription for rewriting it in terms of the velocity is not without ambiguity. Starace[7] considered only a nonlocal potential in which the potential did not depend on the orbitals, although he applied his result to the HF exchange potential.

Using the translation operator, Starace[7] writes the orbital $\phi_\alpha(\vec{r}',\sigma')$ in terms of $\phi_\alpha(\vec{r},\sigma')$ as

$$\phi_\alpha(\vec{r}',\sigma')=\exp[i(\vec{r}'-\vec{r})\cdot\vec{p}/\hbar]\phi_\alpha(\vec{r},\sigma'), \tag{5.2}$$

where $\vec{p}=-i\hbar\partial/\partial\vec{r}$ is the momentum operator. The expansion of the exponential in Eq. (5.2) gives a Taylor series. From this expansion it is clear that the operator $\vec{p}$ does not operate on the expansion parameter $\vec{r}'-\vec{r}$.[26] The effect of the translation operator in Eq. (5.2) is to introduce a momentum dependence.[27]

A point that was apparently overlooked by Starace[7] and others[12] is that the HF exchange potential does not depend on the *local* density at $\vec{r}'$, since it involves $\phi_i(\vec{r},\sigma)$. The orbital $\phi_i(\vec{r},\sigma)$ can be written in terms of the orbital $\phi_i(\vec{r}',\sigma)$ by again applying the translation operator

$$\phi_i(\vec{r},\sigma)=\exp[i(\vec{r}-\vec{r}')\cdot\vec{p}'/\hbar]\phi_i(\vec{r}',\sigma), \tag{5.3}$$

where $\vec{p}'=-i\hbar\partial/\partial\vec{r}'$ is the momentum operator. This equation can also be used in Eq. (5.1). There is thus not a unique prescription for replacing the nonlocal exchange potential by a velocity-dependent potential, since Starace[7] did not use Eq. (5.3).

When both the expressions in Eq. (5.2) and (5.3) are used, the HF exchange potential in Eq. (5.1) becomes

$$[V_{ex}\phi_\alpha](\vec{r},\sigma)=\int d^3r'\,V_2(\vec{r},\vec{r}')\sum_{i=1}^N\phi_i^*(\vec{r}',\sigma')\exp\left(\frac{i(\vec{r}-\vec{r}')\cdot\vec{p}'}{\hbar}\right)\phi_i(\vec{r}',\sigma)\exp\left(\frac{i(\vec{r}'-\vec{r})\cdot\vec{p}}{\hbar}\right)\phi_\alpha(\vec{r},\sigma'). \tag{5.4}$$

In contrast to Eq. (5.1), this velocity-dependent potential is no longer form invariant under the local gauge transformation of Eq. (4.3). For example, Eq. (5.2) transforms under the local gauge transformation in Eq. (4.3) as

$$\phi'_\alpha(\vec{r}',\sigma)=\exp\{i(q/\hbar c)[\Lambda(\vec{r}',t)-\Lambda(\vec{r},t]\}\exp\{(i/\hbar)(\vec{r}'-\vec{r})\cdot[\vec{p}-q\nabla\Lambda(\vec{r},t)/c]\}\phi'_\alpha(\vec{r},\sigma'), \tag{5.5}$$

which is obviously not the same form as Eq. (5.2).

In order to make the velocity-dependent potential in Eq. (5.4) form invariant under local gauge transformations, the principle of minimal electromagnetic coupling can be used, in which $\vec{p}$ is replaced by $\vec{p}-q\vec{A}/c$.[28] Then the potential in Eq. (5.4) becomes

$$[V_{ex}(\vec{A})\phi_\alpha](\vec{r},\sigma)=\int d^3r'\,V_2(\vec{r},\vec{r}')\sum_{i=1}^N\phi_i^*(\vec{r}',\sigma')\exp\{(i/\hbar)(\vec{r}-\vec{r}')\cdot[\vec{p}'-q\vec{A}(\vec{r}',t)/c]\}\phi_i(\vec{r}',\sigma)\times\exp\{(i/\hbar)(\vec{r}'-\vec{r})\cdot[\vec{p}-q\vec{A}(\vec{r},t)/c]\}\phi_\alpha(\vec{r},\sigma'). \tag{5.6}$$

Equation (5.6) is now form invariant under the gauge transformation in Eq. (4.3) in the sense that

$$[V_{ex}(\vec{A}')\phi'_\alpha](\vec{r},\sigma)=\exp[iq\Lambda(\vec{r},t)/\hbar c][V_{ex}(\vec{A})\phi_\alpha](\vec{r},\sigma), \tag{5.7}$$

where the new vector potential $\vec{A}'$ is given by Eq. (2.5). Equation (5.6) shows that there is thus an additional interaction of the system with the electromagnetic field which is induced by this procedure.

However, in situations where (a) magnetic effects are negligible, and (b) the electric field is slowly varying over the dimensions of the atom, the spatial dependence of $\vec{A}$ can be neglected, and $\vec{A}(\vec{r},t) = \vec{A}(t)$.[29] Then Eq. (5.6) becomes

$$[V_{ex}(\vec{A})\phi_\alpha](\vec{r},\sigma) = \int d^3r'\, V(\vec{r},\vec{r}') \sum_{i=1}^{N} \phi_i^*(\vec{r}',\sigma') \exp\{(i/\hbar)(\vec{r}-\vec{r}')\cdot[\vec{p}' - q\vec{A}(t)/c]\}\phi_i(\vec{r}',\sigma) \times \exp\{(i/\hbar)(\vec{r}'-\vec{r})\cdot[\vec{p} - q\vec{A}(t)/c]\}\phi_\alpha(\vec{r},\sigma'). \quad (5.8)$$

Now since $\vec{p}'$ does not act on the expansion variable $\vec{r}-\vec{r}'$ in the first translation operator and $\vec{p}$ does not act on the expansion variable $\vec{r}'-\vec{r}$ in the second translation operator, the dependence on the $\vec{A}(t)$ cancels out. We then obtain in the dipole approximation

$$[V_{ex}(\vec{A})\phi_\alpha](\vec{r},\sigma) = [V_{ex}(0)\phi_\alpha](\vec{r},\sigma), \quad (5.9)$$

where the right-hand side is given in Eq. (5.4). Thus for the HF exchange potential in the electric dipole approximation, there is *no* additional dependence on the electromagnetic field which is induced by converting it in this way to a momentum-dependent, gauge-invariant potential.

It should be emphasized that this section only illustrates a modification of the procedure used by Starace[7] and others.[12] This procedure is not at all necessary because the exchange potential is already gauge invariant as shown in Sec. IV. In the atomic case there are no nonlocal or velocity-dependent potentials in nature, in contrast to the nuclear potential.[11]

VI. LENGTH AND VELOCITY FORMS OF THE INTERACTION

The conclusion arrived at by Starace[7] is that because of the nonlocal nature of the HF exchange potential, a "length" form of the interaction between electromagnetic radiation and matter should be used. As we saw in Sec. III, the "length" form of the interaction should indeed be used, but not in the sense of Starace[7] and Lin.[8] If the HF equations are not to involve the vector potential, the length form that should be used is the electric dipole interaction, $-q\vec{E}\cdot\vec{r}$.

Since the HF equations are gauge invariant, as shown in Sec. IV, any gauge can be used to calculate the effect of the electromagnetic field. However, the freedom to choose the gauge and the freedom to choose the unperturbed Hamiltonian are intimately related, and not independent as commonly supposed.[16,17] When an arbitrary vector potential $\vec{A}$ is used, the gauge invariant HF equations in Eq. (4.1) involve the vector potential.

The usual procedure[30] of expanding out the quadratic term in the Hamiltonian in Eq. (2.2) and treating the electromagnetic field by perturbation theory destroys the gauge invariance.[17] Expanding out Eq. (2.2), we obtain for the interaction

$$H_{int} = -\frac{q}{2mc}\sum_i [\vec{p}_i\cdot\vec{A}(\vec{r}_i,t) + \vec{A}(\vec{r}_i,t)\cdot\vec{p}_i] + \frac{q^2}{2mc^2}\sum_i \vec{A}(\vec{r}_i,t)^2 + \sum_i qA_0(\vec{r}_i,t). \quad (6.1)$$

The Coulomb gauge is usually used, where $\nabla\cdot\vec{A}=0$ and $A_0=0$. In the electric dipole approximation $\vec{A}$ depends only on the time, and the A^2 term can be removed by a unitary transformation.[31] Then Eq. (6.1) gives the usual "velocity" form of the interaction

$$H_{int} = -\frac{q}{mc}\sum_i \vec{A}(t)\cdot\vec{p}_i. \quad (6.2)$$

However, the expansion of the Hamiltonian *destroys* the manifest gauge invariance of the theory developed in Sec. III. When Eq. (6.2) is used as the perturbation the expansion coefficients of the wave function Ψ in terms of the atomic states Φ_n are gauge dependent, and are therefore not probability amplitudes.[16,17]

For many first- and second-order processes, the interaction in Eq. (6.2) gives the same result as the length form of the interaction in Eq. (3.7).[32,33] However, for nonresonant processes, the two interactions are not in general the same.[34] Lamb[35] recommended the "length" interaction in Eq. (3.7) as the proper interaction, because it agreed with his experiments. The interaction in Eq. (3.7) is manifestly gauge invariant because it explicitly involves the electric field $\vec{E}$. On the other hand, the "velocity" interaction in Eq. (6.2) involves the vector potential, and so it is not unique because a term $\nabla\Lambda$ can be added to $\vec{A}$ without changing the physical situation if Λ satisfies Laplace's equation.[35(a)]

Grant[36] has recommended that the velocity form of the interaction be given a privileged position. The velocity form of the interaction in Eq. (6.2) comes from the vector potential in the Coulomb gauge. The Coulomb gauge is one of the most convenient to use when quantizing the electromagnetic

field,[37] but the Lorentz gauge can also be used.[38] In any case, the combined electron-photon system must, of course, be gauge invariant, so obviously any gauge can be used even in the quantized field problem.[39] There thus does not seem to be any basis for recommending the velocity form of the interaction other than convention.

In the situation considered here, the electromagnetic field is classical and the electrons are described nonrelativistically. Any gauge for the electromagnetic field can be used in the Schrödinger equation in Eq. (2.1). As the discussion in Secs. II and III points out, it is not the Hamiltonian that is gauge invariant, but it is the energy operator.[16,17] For the probability amplitudes in Eq. (3.2) to be gauge invariant the eigenstates of the energy operator must be used. In the electric dipole approximation when the gauge is chosen such that the energy operator is equal to the atomic Hamiltonian, it is the length form of the interaction that is obtained. Although this is a specific choice of gauge, it is the only one for which the basis functions do not depend on the electromagnetic field. Grant[36] emphasizes that gauge invariance implies charge conservation in the Lagrangian formalism. He then shows that the HF theory does not satisfy the equation of continuity, which apparently means that the HF theory is not gauge invariant. As we have seen in Sec. IV, HF theory as formulated here is in fact gauge invariant. Grant and Starace[40] have attempted to reconcile their differences, but conclude that "it is clear that there is still much to be done before the last word can be said on the subject."

Because of the extra dependence on the electromagnetic field which was induced by the nonlocal potential, Starace[7] found the interaction to be given by the "length" form

$$H''_{\rm int} = -i\,\frac{q}{\hbar c}\left[H_{\rm HF}, \sum_i \vec{r}_i\right]\cdot\vec{A}, \tag{6.3}$$

where $H_{\rm HF}$ is the HF Hamiltonian for the system in the absence of the electromagnetic field. Because $H_{\rm HF}$ is nonlocal, Eq. (6.3) is not the same as the "velocity" form of the interaction. However, it is also not the same as the length form in Eq. (3.7). As we have seen in Sec. V, the procedure Starace[7] used in obtaining Eq. (6.3) did not include the additional velocity dependence in Eq. (5.3) required to make the exchange potential gauge invariant. Thus the interaction in Eq. (6.3) should not be used.

It is instructive to compare the matrix elements of the interactions in Eq. (6.2) and (6.3). In the Coulomb gauge, the electric field is

$$\vec{E} = -\frac{\partial\vec{A}}{\partial(ct)} = \left(\frac{i\omega}{c}\right)\vec{A}, \tag{6.4}$$

for a harmonically varying field $\vec{A}(t) = \vec{A}(0)\,e^{-i\omega t}$. The commutation relation,

$$\left[H_0, \sum_i \vec{r}_i\right] = -i\hbar\sum_i \frac{\vec{p}_i}{m}\,, \tag{6.5}$$

is valid only for a local potential. When it is used, the matrix elements of $H_{\rm int}$ in Eq. (6.2) with respect to the eigenstates of H_0 in Eq. (3.8) are

$$\langle\Phi_n|H_{\rm int}|\Phi_m\rangle = -q\left(\frac{\omega_{nm}}{\omega}\right)\vec{E}(t)\cdot\langle\Phi_n|\sum_i \vec{r}_i|\Phi_m\rangle\,, \tag{6.6}$$

where $\hbar\omega_{nm} = E_n - E_m$. The HF system Hamiltonian $H_{\rm HF}$ does not satisfy Eq. (6.5). However, the matrix elements of the interaction $H''_{\rm int}$ in Eq. (6.3) are

$$\langle\Phi_n^{\rm HF}|H''_{\rm int}|\Phi_m^{\rm HF}\rangle = -q\left(\frac{\omega_{nm}^{\rm HF}}{\omega}\right)\vec{E}(t)\cdot\langle\Phi_n^{\rm HF}|\sum_i \vec{r}_i|\Phi_m^{\rm HF}\rangle\,, \tag{6.7}$$

where $\hbar\omega_{nm}^{\rm HF} = E_n^{\rm HF} - E_m^{\rm HF}$ is the difference between the system energies calculated in the HF approximation, and $\Phi_n^{\rm HF}$ is the HF wave function for the system in the state n.

Thus, both Eqs. (6.6) and (6.7) have the same form.[41] If the condition of resonance $\omega_{nm} = \omega$ or $\omega_{nm}^{\rm HF} = \omega$ is used in Eqs. (6.6) or (6.7), respectively, they are equal to the corresponding matrix element of the interaction in Eq. (3.7). However, the resonance condition can be satisfied by only one matrix element. Unless two operators have all their matrix elements equal they are not the same operator. For nonresonant processes the interactions in Eqs. (6.2) and (6.3) give results different than the length interaction in Eq. (3.7).[34] In Sec. VII we explore the implications of the inequivalence of length and velocity forms of the interaction on the length and velocity forms of the dipole oscillator strengths.

VII. LENGTH AND VELOCITY FORMS OF OSCILLATOR STRENGTHS

A question that is related with whether to use the length or the velocity form of the interaction is the question of whether to use the length or velocity form of the dipole oscillator strengths. Hansen[42] has shown that to each form of the interaction Hamiltonian there corresponds a form of the dipole oscillator strength, although he says they are all equivalent. We have shown in Sec. III that if the atomic Hamiltonian is to be used as the unperturbed Hamiltonian, it is the length form of the interaction that must be used to insure a gauge invariant result. The length form of the interaction implies that it is the length form of the dipole os-

cillator strength that naturally occurs in Eq. (3.9) for the gauge-invariant probability amplitudes.

The "length" form for oscillator strengths for dipole transitions[43] is defined as

$$f_{nm}^{L}=\frac{2m}{3\hbar}\,\omega_{nm}\left|\langle\Phi_n|\sum_i\vec{r}_i|\Phi_m\rangle\right|^2, \tag{7.1}$$

where $\hbar\omega_{nm}=E_n-E_m$ is the energy difference between the eigenstates Φ_n and Φ_m of the atomic Hamiltonian H_0 in Eq. (3.8). For a *local* Hamiltonian, the commutation relation in Eq. (6.5) can be used to express the dipole length form in terms of the dipole "velocity" form[1]

$$f_{nm}^{V}=\frac{2}{3m\hbar}\,\omega_{nm}^{-1}\left|\langle\Phi_n|\sum_i\vec{p}_i|\Phi_m\rangle\right|^2. \tag{7.2}$$

A further application of the commutation relations in Eq. (6.5) gives the dipole "acceleration" form,[1]

$$f_{nm}^{A}=\frac{2}{3m\hbar}\,\omega_{nm}^{-3}\left|\langle\Psi_n|\sum_i\nabla_i V|\Psi_m\rangle\right|^2, \tag{7.3}$$

where V is the total potential energy. If the exact wave functions are given, then the different forms of the oscillator strengths in Eqs. (7.1)–(7.3) are all numerically equal,

$$f_{nm}^{L}=f_{nm}^{V}=f_{nm}^{A}, \tag{7.4}$$

for a local Hamiltonian H_0. This conclusion is also true if exact eigenstates of any local model Hamiltonian are used. It is universally agreed that the dipole acceleration form in Eq. (7.3) does not give reliable results because it gives a high weight to the region near the origin where approximate wave functions are not especially reliable.

If the exact Hamiltonian H_0 is *nonlocal* (i.e., velocity-dependent) as it is in nuclear physics,[11] then the commutation relation in Eq. (6.5) is not valid. In this case Eq. (7.4) is not valid, and the various expressions in Eqs. (7.1)–(7.3) are not equal. Because of our gauge-invariance arguments in Sec. III there is only *one* dipole oscillator strength, namely, the *length form* in Eq. (7.1). The other expressions in Eqs. (7.2) and (7.3) are based on interactions that violate the gauge invariance of the theory.

In the atomic case where the atomic Hamiltonian is *local*, the situation is more subtle. Although Eq. (3.9) with the length form of the interaction is still the correct equation to use, it may be transformed by using the commutation relation in Eq. (6.5) to give

$$i\hbar\dot{c}_n-E_nc_n=-\sum_{m\neq n}\vec{E}(0,t)\cdot\left(\frac{-i}{m\omega_{nm}}\right)\langle\Phi_n|\sum_1^N q\vec{p}_i|\Phi_m\rangle c_m. \tag{7.5}$$

Even though Eq. (7.5) involves the matrix elements of the "velocity operator" $\vec{p}/m$, it is not the same equation as we would have obtained using the velocity form of the interaction in Eq. (6.2). The matrix elements of the length and velocity forms of the interaction are related by Eq. (6.6).

The controversy over the length versus velocity form of the dipole oscillator strengths is now seen in a different perspective. The length form of the *interaction* in Eq. (3.7) is the one that should be used. However, for a *local* Hamiltonian H_0 Eq. (3.9) can be transformed into Eq. (7.5) which is an equivalent equation for the gauge-invariant probability amplitudes. For a nonlocal model Hamiltonian like the HF Hamiltonian, which in some sense approximates H_0, the length and velocity forms of the dipole oscillator strengths are not equal. The velocity form of the dipole oscillator strength would occur when Eq. (7.5) is solved, and the length form would occur when Eq. (3.9) is solved. It can be legitimately asked then whether the length form of the dipole oscillator strength in Eq. (7.1) or the velocity form in Eq. (7.2) gives a result closer to the exact value. This is a question that must be ultimately answered on the basis of detailed computation.

However, since Eq. (7.5) is derived from Eq. (3.9), which is based on the length form of the interaction, another point of view is also possible when nonlocal model Hamiltonians are used. The length form of the dipole oscillator strength can be considered to be the fundamental one, since it emerges directly from the correct form of the interaction. The deviation of the calculated length form of the dipole oscillator strength from the true value, if known, is a measure of the accuracy of the model Hamiltonian. If the velocity form of the dipole oscillator strength is also calculated, the deviation of it from the calculated length form is a measure of the nonlocality of the model Hamiltonian. This point of view emphasizes that it is the length form of the interaction which is fundamental, and which leads naturally to the length form of the dipole oscillator strength. The velocity form of the dipole oscillator strength is a derived result that is only valid when the true local Hamiltonian is used in the commutation relation of Eq. (6.5).

VIII. CONCLUSION

This paper has shown that on the basis of the gauge invariance of the interaction of electric dipole radiation with matter, the length form of the interaction, $-q\vec{E}\cdot\vec{r}$, should be used, and not the velocity form of the interaction, $-(q/m)\vec{A}\cdot\vec{p}$. Therefore, the length form of the dipole oscillator strength arises naturally in the solution of the

equations of time-dependent perturbation theory. If the Hamiltonian is nonlocal, as it is in nuclear physics,[11] the length form of the dipole oscillator strength is the only valid form.

If the Hamiltonian is local, as it is in atomic physics, the exact length form of the dipole oscillator strength may be transformed into the velocity form. In practice, exact wave functions are seldom available and it is necessary to use approximate wave functions that are eigenstates of some model Hamiltonian. If the model Hamiltonian is local there is no problem, since the length and velocity forms of the dipole oscillator strengths are still equal, although not equal to the exact oscillator strength. But if the model Hamiltonian is nonlocal, as is the HF Hamiltonian, the length and velocity forms of the dipole oscillator strengths are not equal.

Two alternatives then arise. The first is to calculate the length form of the dipole oscillator strength with the eigenstates of the nonlocal Hamiltonian. The second is to transform the length form of the oscillator strength to the velocity form with the true local Hamiltonian, and then calculate the matrix elements with eigenstates of the nonlocal model Hamiltonian. These two procedures lead to different results, and which gives an answer closest to the exact oscillator strength is a question that must be answered by detailed computations in a particular situation.

Since the length form of the interaction is the one implied by gauge-invariance arguments, the length form of the dipole oscillator strength is the form that occurs naturally. The velocity form must be obtained by a transformation based on the commutation relation in Eq. (6.5). Thus a valid point of view is to consider the deviation of the length form of the dipole oscillator strength from the true value, if known, as a measure of the accuracy of the theory. The deviation of the velocity form of the dipole oscillator strength from the length form is then taken to be a measure of the nonlocality of the model Hamiltonian.

This paper has not solved the problem for the case of a local Hamiltonian of which form of the dipole oscillator strength gives the best numerical result. It has shown on the basis of gauge invariance that it is the length form of the *interaction* that must be used. For a nonlocal Hamiltonian only the length form of the oscillator strength makes sense. For a local Hamiltonian the velocity form of the oscillator strength must be derived from the length form using an exact commutation relation. In this sense the length form of the dipole oscillator strength is more fundamental than the velocity form.

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[19]The terminology "gauge transformation of the first and second kind" goes back to W. Pauli [Rev. Mod. Phys. 13, 203 (1941)]. Unfortunately, this terminology is not universally used.
[20]Note that $H(\vec{A},0)$ is *not* the Hamiltonian in the *specific gauge* in which $A_0=0$. The scalar potential is still nonzero in general, and the total Hamiltonian $H(\vec{A},A_0)$ is related to the energy operator $H(\vec{A},0)$ by

$$H(\vec{A},A_0)=H(\vec{A},0)+\sum_i qA_0(\vec{r}_i,t).$$

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[23]The term $qA_0(\vec{r},t)$ is specifically excluded in Eq. (4.1). For a time-dependent electromagnetic field, the negative gradient of qA_0 is related to the electric force $q\vec{E}$ by

$$-\nabla qA_0=q\vec{E}+\frac{q}{c}\frac{\partial\vec{A}}{\partial t}.$$

Thus qA_0 in this case is not a true potential energy, since its negative gradient is not a true force.

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[29]Starace (Ref. 7) also limited himself to this case when he expanded the translation operator. He retained only the first two terms in the expansion

$$\exp[-i(q/\hbar c)(\vec{r}'-\vec{r})\cdot\vec{A}(t)]$$
$$=1-i(q/\hbar c)(\vec{r}'-\vec{r})\cdot\vec{A}(t)+\cdots.$$

The validity of such an approximation is open to question. Then he absorbed the term involving $\vec{A}$ with the "velocity" interaction $\vec{A}\cdot\vec{p}$ to get a "length" form of the interaction. When $\vec{A}=\vec{A}(t)$, then $\vec{B}=\nabla\times\vec{A}(t)=0$, so the magnetic field is neglected.

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$$H'_{\rm int}=-\frac{q}{mc}\sum_i\vec{A}'(\vec{r}_i,t)\cdot\vec{p}_i+\frac{q^2}{2mc^2}\sum_i\vec{A}'(\vec{r}_i,t)^2-\frac{q}{c}\sum_i\frac{\partial\Lambda(\vec{r}_i,t)}{\partial t}$$

where $\vec{A}'$ is given in Eq. (2.5).

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