

Equivalence of interaction Hamiltonians in the electric dipole approximation*

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Abstract. The interaction of an atomic system with an externally applied electromagnetic field can be treated in the electric dipole approximation by means of either the minimal coupling ($\mathbf{p} \cdot \mathbf{A}$) or direct coupling ($\mathbf{d} \cdot \mathbf{E}$) Hamiltonian. It is shown that both methods lead to identical and unambiguous predictions for observable quantities as long as the atomic wavefunctions are transformed when used in the minimal-coupling formulation. The physical meaning of kinetic momentum is used to show that the atomic states must be described by wavefunctions calculated in the absence of an electromagnetic field when using the $\mathbf{d} \cdot \mathbf{E}$ (but not the $\mathbf{p} \cdot \mathbf{A}$) form of the interaction Hamiltonian. When, however, observables are calculated using the common approximations of resonance atomic physics – the two-level approximation and the rotating-wave approximation – the two formulations can lead to measurably different results. This point is illustrated by calculating the induced polarization (and hence the refractive index) of an atomic system for the two exactly soluble cases of the harmonic oscillator and the hydrogen atom.

1. Introduction

There are two competing ways of describing the interaction of a quantum mechanical system with an electromagnetic radiation field, the characteristic wavelengths of which are assumed much greater than the dimensions of the system. One method involves replacing \mathbf{p} with $\mathbf{p} - (e/c)\mathbf{A}$ in the field-free Hamiltonian, and is referred to as the ‘minimal coupling’ procedure or as the $\mathbf{p} \cdot \mathbf{A}$ form of the interaction. The other method, which is conceptually somewhat simpler, involves introducing an interaction Hamiltonian of the form $\mathbf{d} \cdot \mathbf{E}$, and is referred to as the ‘direct coupling’ of atomic dipole transition moment \mathbf{d} to the applied electric field strength \mathbf{E} . Many authors refer to these formulations as the velocity gauge and the length gauge, respectively. The question of whether the two descriptions always lead to the same predictions for observable quantities has been addressed by many authors [1–10], but there has been considerable disagreement regarding the answer to this question. In particular, references [2, 3, 6, 7] state

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that there are no measurable differences between the predictions of the two descriptions, whereas references [1, 5, 8, 10, 11] state that such differences can exist.

We note that Aharanov and Au [11] have presented an eloquent argument that an additional phase factor has to be added to the atomic wavefunction when used to calculate the interaction in the $\mathbf{p} \cdot \mathbf{A}$ but not the $\mathbf{d} \cdot \mathbf{E}$ form of the interaction Hamiltonian. This line of reasoning suggests that the $\mathbf{d} \cdot \mathbf{E}$ form is the simpler to use in that correction factors need not be included when using this form. We also note that conclusions similar to those reached in the present paper have been presented by Leubner and Zoller [12] for the specific case of multiphoton ionization probabilities. It has also been pointed out [13] that direct numerical integration of the Schrödinger equation for strong-field ionization if performed with the $\mathbf{p} \cdot \mathbf{A}$ form yields consistent results only if the distinction between canonical and kinetic momentum is properly treated. In treating large-scale atomic structure calculations, Deb *et al.* [14] point out that results obtained using the two forms of the interaction Hamiltonian are numerically very similar, which is taken to be a self-consistency check of their results. Cormier and Lampropoulis [15] state that the $\mathbf{p} \cdot \mathbf{A}$ form is more convenient for calculations in strong-field ionization. However, Xie *et al.* [16] claim that the $\mathbf{d} \cdot \mathbf{E}$ form gives results that more closely agree with experiment than does the $\mathbf{p} \cdot \mathbf{A}$ form. Also, Kuan *et al.* [17] suggest that different forms of the interaction Hamiltonian lead to different predictions.

In this paper, we restrict our attention to the case in which the electromagnetic field is treated as an external, prescribed, nondynamic, c -number function. We believe that in this simple case there is no ambiguity, and that either description, if used in accordance with the principle of gauge invariance, leads to the same results. In section 2 of this paper, we demonstrate the general equivalence of these two descriptions, and point out the necessity of transforming the initial states of the system when describing the dynamics through the use of a gauge in which the vector potential \mathbf{A} is nonzero. In section 3, we illustrate these general results by applying them to a calculation of the refractive index of a collection of atoms. We show that while the exact results of either description lead to the same prediction for the refractive index, the commonly used approximations of resonance atomic physics, i.e., the two-level approximation and the rotating-wave approximation (RWA), lead to measurably large discrepancies between the predictions of the two descriptions. We suspect that some of the past confusion regarding the possible observable consequences of the choice of the interaction Hamiltonian has resulted from a failure to make a distinction between exact and approximate results. Since it is often necessary to make the two-level and/or rotating-wave approximations in practical problems in quantum optics, section 3 also presents a numerical comparison of the predictions based on these approximations with the exact results for two cases – the harmonic oscillator and the hydrogen atom – where exact results can be obtained.

2. General equivalence of the two formulations

Before turning to our main problem, let us consider the following simple example which illustrates the influence of the choice of gauge for a

problem in classical Hamiltonian mechanics. The Hamiltonian describing the dynamics of a particle of charge $e = -|e|$ in a constant, uniform magnetic field is given by

$$H = \frac{1}{2m} \left[\mathbf{p} - \frac{e}{c} \mathbf{A}(\mathbf{r}) \right]^2, \quad (2.1)$$

where, however, there is no unique choice for the vector potential $\mathbf{A}(\mathbf{r})$. Two (out of infinitely many) of the most commonly used forms are given by

$$\mathbf{A}_1(\mathbf{r}) = \frac{1}{2}(-By, Bx, 0), \quad (2.2)$$

and

$$\mathbf{A}_2(\mathbf{r}) = (-By, 0, 0), \quad (2.3)$$

for a magnetic field given by $\mathbf{B} = (0, 0, B)$. One can easily verify that $L_z = (\mathbf{r} \times \mathbf{p})_z$ commutes with the Hamiltonian (2.1) if (2.2) is substituted as the vector potential, but that it does not if (2.3) is used. The question then arises: is the z -component of the angular momentum a conserved quantity for a charged particle moving in a constant z -directed magnetic field? The paradox is easily resolved by deriving a formula for the kinetic momentum $\boldsymbol{\pi}$ from (2.1):

$$\boldsymbol{\pi} \equiv m\dot{\mathbf{r}} = m \frac{\partial H}{\partial \mathbf{p}} = \left[\mathbf{p} - \frac{e}{c} \mathbf{A}(\mathbf{r}) \right]. \quad (2.4)$$

Since velocity has a well-defined operational meaning and the vector potential can be chosen in different ways, we conclude that the canonical momentum \mathbf{p} is not a unique dynamical variable, and in fact depends upon the choice of gauge. Therefore, together with the two different vector potentials $\mathbf{A}_1(\mathbf{r})$ and $\mathbf{A}_2(\mathbf{r})$ we also have two different canonical momenta \mathbf{p}_1 and \mathbf{p}_2 and consequently two different canonical angular momentum L_{1z} and L_{2z} . Incidentally, neither of these coincides with the kinetic angular momentum $\mathbf{r} \times \boldsymbol{\pi}$. The distinction between different canonical momenta is well understood in classical mechanics, but is often forgotten in quantum mechanics because we are used to representing canonical variables in the coordinate-space Schrödinger representation, where canonical momentum is always represented by differentiation:

$$\hat{\mathbf{p}} \rightarrow -i\hbar \frac{\partial}{\partial \mathbf{r}}. \quad (2.5)$$

The primary intent of our paper is to consider the interaction of an atomic system with an external electromagnetic field. In the electric-dipole approximation and using the minimal-coupling formulation of the interaction, the evolution of the atomic system is governed by the Schrödinger equation

$$i\hbar \frac{\partial \phi(\mathbf{r}, t)}{\partial t} = \left\{ \frac{1}{2m} \left[\mathbf{p} - \frac{e}{c} \mathbf{A}(t) \right]^2 + V(\mathbf{r}) \right\} \phi(\mathbf{r}, t), \quad (2.6)$$

where $V(\mathbf{r})$ is the scalar potential and where the vector potential is restricted only by the requirement

$$\mathbf{E}(t) = -\frac{1}{c} \frac{\partial \mathbf{A}}{\partial t}, \quad (2.7)$$

i.e., that its time derivative must reproduce the applied electric field.

Now, making the substitution

$$\phi(\mathbf{r}, t) = e^{i(e/\hbar c)\mathbf{A}(t)\cdot\mathbf{r}}\psi(\mathbf{r}, t), \quad (2.8)$$

which was known already to Pauli [18] in the early 1940s, we obtain

$$i\hbar\frac{\partial\psi(\mathbf{r}, t)}{\partial t} = \left[\frac{1}{2m} \boldsymbol{\pi}^2 + V(\mathbf{r}) - e\mathbf{r} \cdot \mathbf{E}(t) \right] \psi(\mathbf{r}, t), \quad (2.9)$$

which constitutes another equally legitimate formulation of the same dynamical problem, and is governed by the Hamiltonian:

$$H = \frac{\boldsymbol{\pi}^2}{2m} - e\mathbf{r} \cdot \mathbf{E}(t) + V(\mathbf{r}). \quad (2.10)$$

This formulation is conceptually simpler than that of equation (2.6), because by (2.4) its canonical momentum coincides with the kinetic momentum of the charged particle. For this reason we chose $\boldsymbol{\pi}$ to denote the standard differential operator appearing in (2.9) and (2.10). Note, however, that going beyond the dipole approximation is simpler in the $\mathbf{p} \cdot \mathbf{A}$ formulation. To do so, one needs only include the position dependence of the vector potential. On the other hand, the analogous extension of the $\mathbf{d} \cdot \mathbf{E}$ formulation requires the use of, in principle, infinitely many multipole terms.

With the substitution (2.8) in mind, it is obvious that all physical quantities can be computed in either formulation in such a way that the results coincide. In the Heisenberg picture, for instance, the evolution of the expectation value of any gauge-independent operator \hat{O} can be computed using either formulation, since

$$\begin{aligned} \langle \hat{O}(t) \rangle &= \langle \phi(\mathbf{r}, 0) | U_{\mathbf{p}\cdot\mathbf{A}}^{-1}(t, 0) \hat{O} U_{\mathbf{p}\cdot\mathbf{A}}(t, 0) | \phi(\mathbf{r}, 0) \rangle \\ &= \langle \psi(\mathbf{r}, 0) | U_{\mathbf{d}\cdot\mathbf{E}}^{-1}(t, 0) \hat{O} U_{\mathbf{d}\cdot\mathbf{E}}(t, 0) | \psi(\mathbf{r}, 0) \rangle, \end{aligned} \quad (2.11)$$

where we have used obvious notation for evolution operators in the two formulations and have assumed that $\phi(\mathbf{r}, 0)$ and $\psi(\mathbf{r}, 0)$ are related by (2.8).

Within the context of the Heisenberg picture, the state of the system need be specified only at the initial time $t = 0$. The question then arises: supposing that at the initial time $t = 0$ the state of the system is known to be the ground state of the hydrogen atom, should we take the function $\psi(\mathbf{r}, t)$ of the $\mathbf{d} \cdot \mathbf{E}$ formulation or the function $\phi(\mathbf{r}, t)$ of $\mathbf{p} \cdot \mathbf{A}$ formulation to have the standard form

$$\psi_s(\mathbf{r}) = \frac{1}{\pi^{1/2} a_0^{3/2}} e^{-r/a_0} ? \quad (2.12)$$

First of all, this question does not arise in some physically interesting situations, namely those for which $\mathbf{A}(0) = 0$, as in this case the distinction between $\phi(\mathbf{r}, 0)$ and $\psi(\mathbf{r}, 0)$ obviously disappears. Thus, for the case of a scattering problem, i.e., one in which no field \mathbf{E} is present at the initial time, the situation is very simple. It is also worth stressing that since the state of the system need be specified only at the initial instant of time, one can always choose a gauge in which $\mathbf{A}(0) = 0$, even if the field \mathbf{E} is nonzero at $t = 0$, and thereby eliminate the problem.

This dilemma cannot always be eliminated in the Schrödinger picture, however. It is present, for instance, in the case in which transition amplitudes are to be computed between the two instants of time 0 and t , if $\mathbf{E}(0) \neq 0$ and $\mathbf{E}(t) \neq 0$, since in general we cannot choose $\mathbf{A}(t)$ to be zero at both of these times.

The question of whether $\phi(\mathbf{r}, 0)$ or $\psi(\mathbf{r}, 0)$ should be represented by $\psi_s(\mathbf{r})$ can be resolved by recalling the above-mentioned distinction between kinetic and canonical momentum, which exists whenever $\mathbf{A}(t) \neq 0$. Whereas $\psi(\mathbf{r}, t)$ and $\phi(\mathbf{r}, t)$ have identical probability distributions for the position, they have entirely different Fourier transforms and thus entirely different probability distributions for the canonical momenta. The correct choice of the initial wavefunction is the one which gives rise to the correct probability distribution for the kinetic momentum $\boldsymbol{\pi}$ which in the $\mathbf{p} \cdot \mathbf{A}$ formulation is equal to $\mathbf{p} - (e/c) \mathbf{A}$; and thus the correct choice of initial wavefunction depends on the choice of the vector potential \mathbf{A} . In order that the probability distribution for not depend on the arbitrary choice of the gauge of the vector potential, the standard wavefunction $\psi_s(\mathbf{r})$ of (2.12) must be used in conjunction with the $\mathbf{d} \cdot \mathbf{E}$ formulation, in which $A(t)$ is always zero, and this standard wavefunction transformed by (2.8) must be used in the $\mathbf{p} \cdot \mathbf{A}$ formulation. By way of illustration, if the standard wavefunction $\psi_s(\mathbf{r})$ were to be used in conjunction with the $\mathbf{p} \cdot \mathbf{A}$ formulation, the expectation value of the kinetic momentum for the ground state of the hydrogen atom, which is known to be zero, would be given by

$$\begin{aligned} \langle \boldsymbol{\pi} \rangle &= \langle \psi_s(\mathbf{r}) | \boldsymbol{\pi} | \psi_s(\mathbf{r}) \rangle = \langle \psi_s(\mathbf{r}) | \left[\mathbf{p} - \frac{e}{c} \mathbf{A}(t) \right] | \psi_s(\mathbf{r}) \rangle \\ &= -\frac{e}{c} A(t), \end{aligned} \tag{2.13}$$

which is explicitly nonzero and is obviously gauge dependent. It is necessary in the $\mathbf{p} \cdot \mathbf{A}$ formulation to transform the standard wavefunction $\psi_s(\mathbf{r})$ by (2.8), for which case the expectation value is given by

$$\begin{aligned} \langle \boldsymbol{\pi} \rangle &= \langle \psi_s(\mathbf{r}) e^{i(e/\hbar c)\mathbf{A}(t)\cdot\mathbf{r}} | \boldsymbol{\pi} | \psi_s(\mathbf{r}) e^{i(e/\hbar c)\mathbf{A}(t)\cdot\mathbf{r}} \rangle \\ &= \langle \psi_s(\mathbf{r}) | e^{-i(e/\hbar c)\mathbf{A}(t)\cdot\mathbf{r}} \left[\mathbf{p} - \frac{e}{c} \mathbf{A}(t) \right] e^{i(e/\hbar c)\mathbf{A}(t)\cdot\mathbf{r}} | \psi_s(\mathbf{r}) \rangle \\ &= \langle \psi_s(\mathbf{r}) | \mathbf{p} | \psi_s(\mathbf{r}) \rangle = \mathbf{0}. \end{aligned} \tag{2.14}$$

Once the result is established, it is obvious that consistently performed perturbative calculations will also yield the same results in both approaches. We shall illustrate this result by calculating the polarization of an atom subject to an external monochromatic field. The first-order perturbative formula for the time-evolution operator is

$$U(t, 0) \approx e^{-i\hbar^{-1}H_0t} \left[1 - i \int_0^t \left(e^{i\hbar^{-1}H_0\tau} H_I e^{-i\hbar^{-1}H_0\tau} \right) d\tau \right]. \tag{2.15}$$

Since the electron charge $e = -|e|$ constitutes the expansion coefficient, there are, for our situation, two different splittings of the Hamiltonian into free and interacting parts, which are given by

$$H'_o = \frac{\mathbf{p}^2}{2m} + V(\mathbf{r}); \quad H'_I = -\frac{e}{c} \mathbf{p} \cdot \mathbf{A}(t), \tag{2.16}$$

(we have neglected the \mathbf{A}^2 term in the lowest order) and

$$H''_o = \frac{\boldsymbol{\pi}^2}{2m} + V(\mathbf{r}); \quad H''_I = -e\mathbf{r} \cdot \mathbf{E}(t). \tag{2.17}$$

In the $\mathbf{p} \cdot \mathbf{A}$ formulation, the coupling constant e also appears in the description of the state, however, and to first order the transformation (2.8) reduces to

$$\phi(\mathbf{r}, t) = \left[1 + i \frac{e}{\hbar c} \mathbf{A}(t) \cdot \mathbf{r} \right] \psi(\mathbf{r}, t). \quad (2.18)$$

Let us now compute the evolution of the expectation value of the position operator \mathbf{r} if at time $t = 0$ the atomic system is in its ground state with wavefunction ψ_0 . Using the $\mathbf{d} \cdot \mathbf{E}$ formulation we obtain

$$\begin{aligned} & \langle \psi_0 | U_{\mathbf{d}\cdot\mathbf{E}}^{-1} \mathbf{r} U_{\mathbf{d}\cdot\mathbf{E}} | \psi_0 \rangle \\ &= - \frac{ie}{\hbar} \left\langle \psi_0 \left| \left[\int_0^t d\tau e^{i\hbar^{-1}H'_o\tau} \mathbf{r} \cdot \mathbf{E}(\tau) e^{-i\hbar^{-1}H'_o\tau}, e^{i\hbar^{-1}H'_ot} \mathbf{r} e^{-i\hbar^{-1}H'_ot} \right] \right| \psi_0 \right\rangle. \end{aligned} \quad (2.19)$$

If instead we use the $\mathbf{p} \cdot \mathbf{A}$ formulation we obtain

$$\begin{aligned} & \langle \phi_0 | U_{\mathbf{p}\cdot\mathbf{A}}^{-1} \mathbf{r} U_{\mathbf{p}\cdot\mathbf{A}} | \phi_0 \rangle \\ &= - \frac{ie}{m\hbar c} \left\langle \psi_0 \left| \left[\int_0^t d\tau e^{i\hbar^{-1}H'_o\tau} \mathbf{p} \cdot \mathbf{A}(\tau) e^{-i\hbar^{-1}H'_o\tau}, e^{i\hbar^{-1}H'_ot} \mathbf{r} e^{-i\hbar^{-1}H'_ot} \right] \right| \psi_0 \right\rangle \\ &+ \frac{ie}{\hbar c} \left\langle \psi_0 \left| \left[\mathbf{r} \cdot \mathbf{A}, e^{i\hbar^{-1}H'_ot} \mathbf{r} e^{-i\hbar^{-1}H'_ot} \right] \right| \psi_0 \right\rangle. \end{aligned} \quad (2.20)$$

The last term in this expression comes from the transformation of the states. We shall now show that (2.20) is equal to (2.19). The standard identity

$$\mathbf{p} = im\hbar^{-1} [\mathbf{r}, H'_o], \quad (2.21)$$

is substituted into (2.20) and the expression is then transformed using

$$i\hbar^{-1} e^{i\hbar^{-1}H'_ot} [\mathbf{r}, H'_o] e^{-i\hbar^{-1}H'_ot} = \frac{d}{dt} \left(e^{i\hbar^{-1}H'_ot} \mathbf{r} e^{-i\hbar^{-1}H'_ot} \right). \quad (2.22)$$

If the resulting expression is integrated by parts with respect to τ , the boundary terms are found to cancel the last term in (2.20), and thus this final form of (2.20) coincides with (2.19).

If the atomic states had not been transformed using Eq. (2.8), the second term on the right-hand side of (2.20) would appear as a discrepancy between the predictions of the $\mathbf{p} \cdot \mathbf{A}$ and $\mathbf{d} \cdot \mathbf{E}$ formulations. This term does not generally vanish and for the case of a harmonic oscillator of resonance frequency ω_0 can be evaluated as

$$\frac{i\hbar}{m\omega} \mathbf{A}(0) \sin \omega_0 t. \quad (2.23)$$

Unlike the extremely small differences between the predictions of the two formulations for the case of a quantized dynamical field, [5, 19, 20], this artificial discrepancy can be made arbitrarily large simply by adding a huge constant to the vector potential. It should be noted, however, that this discrepancy has Fourier components only at the transition frequencies of the atomic system and not at the frequency of the applied field. This discrepancy thus appears only in the transient response of the atomic system; the steady state response can be calculated correctly even if the atomic states are not properly transformed.

3. Refractive index of an atomic vapor

The refractive index of an atomic vapor, sufficiently dilute that local-field effects can be neglected, is given by [21]

$$n(\omega) = 1 + 2\pi N \alpha(\omega), \tag{3.1}$$

where N is the number density of atoms and $\alpha(\omega)$ is the atomic polarizability, which is defined in terms of the Fourier component of the induced atomic dipole moment at the frequency of the applied field as

$$\mathbf{d}(\omega) = e\langle \mathbf{r}(\omega) \rangle = \alpha(\omega)\hat{\boldsymbol{\epsilon}}E_0, \tag{3.2}$$

where it has been assumed that the applied optical field is of the form

$$\mathbf{E}(t) = \hat{\boldsymbol{\epsilon}}E_0e^{-i\omega t} + \hat{\boldsymbol{\epsilon}}^*E_0^*e^{i\omega t}. \tag{3.3}$$

An expression for $\langle \mathbf{r}(\omega) \rangle$, using the $\mathbf{d} \cdot \mathbf{E}$ formulation, can be obtained from (2.17) by inserting a complete set of states $\sum_n |\psi_n\rangle\langle\psi_n|$ between the two expressions that appear in the commutator, giving the result

$$\langle \mathbf{r}(\omega) \rangle = \frac{eE_0}{\hbar} \sum_n \left(\frac{\langle \psi_0 | \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_0 \rangle}{\omega_{n0} - \omega} + \frac{\langle \psi_0 | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \mathbf{r} | \psi_0 \rangle}{\omega_{n0} + \omega} \right). \tag{3.4}$$

(This expression can also be derived by the more conventional Schrödinger-picture method, as is done in reference [22].) The polarizability as derived using the $\mathbf{d} \cdot \mathbf{E}$ formulation is thus given by

$$\alpha(\omega) = e \hat{\boldsymbol{\epsilon}}^* \cdot \langle \mathbf{r}(\omega) \rangle / E_0,$$

or

$$\alpha_{\mathbf{d}\cdot\mathbf{E}}(\omega) = \frac{e^2}{\hbar} \sum_n \left(\frac{\langle \psi_0 | \hat{\boldsymbol{\epsilon}}^* \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_0 \rangle}{\omega_{n0} - \omega} + \frac{\langle \psi_0 | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}}^* \cdot \mathbf{r} | \psi_0 \rangle}{\omega_{n0} + \omega} \right). \tag{3.5}$$

If this calculation is now repeated using the $\mathbf{p} \cdot \mathbf{A}$ formulation, either from equation (2.20) or by using the same technique as in reference [22], we obtain

$$\alpha_{\mathbf{p}\cdot\mathbf{A}}(\omega) = \frac{e^2}{\hbar} \sum_n \left[\frac{\omega_n}{\omega} \left(\frac{\langle \psi_0 | \hat{\boldsymbol{\epsilon}}^* \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_0 \rangle}{\omega_{n0} - \omega} - \frac{\langle \psi_0 | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}}^* \cdot \mathbf{r} | \psi_0 \rangle}{\omega_{n0} + \omega} \right) \right]. \tag{3.6}$$

The equality of expressions (3.5) and (3.6), which must follow from the general principles outlined in the last section, can be explicitly verified by noting the difference between them is of the form

$$\frac{e^2}{\hbar\omega} \sum_n \langle \psi_0 | \hat{\boldsymbol{\epsilon}}^* \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_0 \rangle - \langle \psi_0 | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{r} | \psi_n \rangle \langle \psi_n | \hat{\boldsymbol{\epsilon}}^* \cdot \mathbf{r} | \psi_0 \rangle. \tag{3.7}$$

Since the unperturbed eigenstates $|\psi_n\rangle$ must form a complete set, we can replace the operator $\sum_n |\psi_n\rangle\langle\psi_n|$ by unity, and expression (3.7) is seen to be identically zero. The procedure we have used to deduce this result is similar to that described by Dirac [23].

Equation (3.1) together with equation (3.5) or (3.6) determines the refractive index of the gas so long as the field frequency ω is not too close to any resonance frequency ω_{n0} . For an isotropic medium the resulting expression for $n(\omega)$ can be simplified by expressing the dipole transition moments in terms of the oscillator strength f_{n0} defined by

$$f_{n0} = \frac{2m\langle\psi_0|\mathbf{r}|\psi_n\rangle \cdot \langle\psi_n|\mathbf{r}|\psi_0\rangle\omega_{n0}}{3\hbar}, \quad (3.8)$$

and becomes

$$n(\omega) = 1 + \frac{2\pi Ne^2}{m} \sum_n \frac{f_{n0}}{2\omega_{n0}} \left(\frac{1}{\omega_{n0} - \omega} + \frac{1}{\omega_{n0} + \omega} \right). \quad (3.9)$$

We now follow historical precedent and re-express this result in terms of the dimensionless Kramers-Heisenberg matrix element [23] $M(\omega)$ as

$$n(\omega) = 1 + \frac{2\pi Ne^2}{m\omega^2} M(\omega), \quad (3.10)$$

where $M(\omega)$ can be expressed in any of the three algebraically equivalent forms

$$M_1(\omega) = \sum_n \frac{f_{n0}}{2} \frac{\omega^2}{\omega_{n0}} \left(\frac{1}{\omega_{n0} - \omega} + \frac{1}{\omega_{n0} + \omega} \right), \quad (3.11)$$

$$M_2(\omega) = \sum_n \frac{f_{n0}}{2} \omega \left(\frac{1}{\omega_{n0} - \omega} - \frac{1}{\omega_{n0} + \omega} \right), \quad (3.12)$$

$$M_3(\omega) = -1 + \sum_n \frac{f_{n0}}{2} \omega_{n0} \left(\frac{1}{\omega_{n0} - \omega} + \frac{1}{\omega_{n0} + \omega} \right). \quad (3.13)$$

While these three expressions are exactly equal, commonly employed approximate forms [24] of these expressions are not equal. The rotating-wave approximation (RWA) entails ignoring the perturbation caused by the negative-frequency part of the electromagnetic field in calculating the positive-frequency part of the atomic response. If $\langle\mathbf{r}(\omega)\rangle$ is calculated in the RWA using the $\mathbf{d} \cdot \mathbf{E}$ form of the interaction Hamiltonian, one is led to an expression for the refractive index involving $M_1(\omega)$ with the antiresonant term [i.e., the term containing $1/(\omega_{n0} + \omega)$] absent. However, if $\langle\mathbf{r}(\omega)\rangle$ is calculated in the RWA using the $\mathbf{p} \cdot \mathbf{A}$ form of the interaction Hamiltonian, one obtains an expression involving $M_2(\omega)$, with the antiresonant term again missing. Finally the refractive index can be obtained by treating the interaction as a scattering problem [25]. If this calculation is performed in the RWA using the $\mathbf{p} \cdot \mathbf{A}$ form of the interaction Hamiltonian, one obtains an expression for the refractive index involving $M_3(\omega)$ with the antiresonant term and the term -1 absent. (In this case the term -1 results from the $\mathbf{A} \cdot \mathbf{A}$ interaction). We thus conclude that there is not a unique RWA but rather (at least) three such approximations. Another common approximation involves assuming that if the applied frequency is sufficiently close to one of the resonant frequencies, it is permissible to replace the infinite sums in equations (3.11) by the single term involving that frequency; this is called the two-level approximation.

We now investigate numerically the size of the errors introduced by making the rotating-wave and/or the two-level approximations. We consider first the case of a three-dimensional harmonic oscillation of resonance frequency ω_0 , which is known

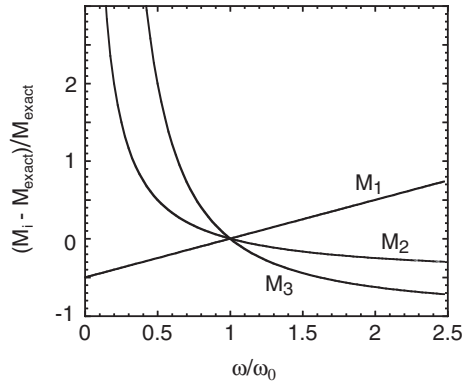


Figure 1. Relative difference between several forms of the Kramers-Heisenberg matrix element $M_i(\omega)$ in the rotating wave approximation and the exact form of $M(\omega)$ for a harmonic oscillator of resonance frequency ω_0 . Note that all of the forms agree at the resonance frequency, but that significant differences can occur far from resonance.

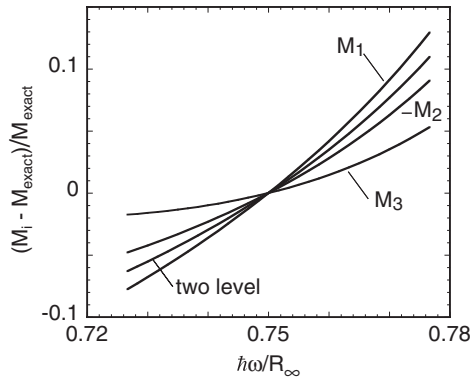


Figure 2. Relative difference between several approximate forms of the Kramers-Heisenberg matrix element $M(\omega)$ and the exact result for frequencies near the Lyman α line of hydrogen.

to be in its ground state. For this system the electric-dipole selection rules require that the principal quantum number change by unity, implying that for this case the two-level approximation is exact. The three forms of $M(\omega)$ in the RWA are compared with the exact result in figure 1. For the range $0 < \omega < 2.5\omega_0$, $M_3(\omega)$ in the RWA consistently gives the worst results. The relative errors vanish at the exact resonance. Below resonance, the approximate form of $M_1(\omega)$ gives the better agreement whereas above resonance the approximate form of $M_2(\omega)$ gives the better agreement. In all three cases, the discrepancies are comparable with the magnitude of the nonresonant background itself.

The other case for which an exact analytic expression for $M(\omega)$ is available is the hydrogen atom. Gavrilá [26] has shown that $M(\omega)$ can be expressed analytically in terms of the Gauss hypergeometric function ${}_2F_1$. We have numerically evaluated this exact expression for $M(\omega)$ and have compared these results with those of equations (3.11) in the two-level approximation and in the RWA. This comparison is shown in figure 2 for the Lyman α ($n = 1 \rightarrow n = 2$) transition of hydrogen. The oscillator strength of this transition is 0.4162. Thus, the

two-level approximation can be expected to be valid only close to resonance, and we have therefore compared the various approximations with theory only close to resonance. For this case, the two forms of the $\mathbf{p} \cdot \mathbf{A}$ interaction in the RWA give the best agreement, the $\mathbf{d} \cdot \mathbf{E}$ form in the RWA gives the worst agreement, and the full two-level approximation lies intermediately between. The actual values of the discrepancies change only slowly with ω near the resonance frequency. At exact resonance these discrepancies are given as follows [27]:

$$\begin{aligned} M \text{ (two-level)} - M \text{ (exact)} &= -0.500, \\ M_1 \text{ (two-level, RWA)} - M \text{ (exact)} &= -0.604, \\ M_2 \text{ (two-level, RWA)} - M \text{ (exact)} &= -0.396, \\ M_3 \text{ (two-level, RWA)} - M \text{ (exact)} &= -0.188. \end{aligned}$$

Again, the discrepancy between the various approximate values is comparable with the magnitude of the nonresonant contribution to M .

We wish to point out that the differences in these predictions, which are a consequence of the approximations involved and not of the choice of the interaction Hamiltonian, are in fact measurable. Consider the measurement of the refractive index of an atomic vapor of number density $N = 10^{16} \text{ cm}^{-3}$ at a frequency 1 cm^{-1} away from a strong optical transition of oscillator strength unity. Assuming that the resonance frequency corresponds to $20,000 \text{ cm}^{-1}$, the Kramers-Heisenberg matrix element will be of order 10^4 , and the difference of the refractive index from unity will be of order 0.01. The various approximate forms of the Kramers-Heisenberg matrix element will thus lead to variations in the refractive index of order 10^{-6} , which are easily measured using the techniques of classical interferometry.

4. Conclusion

In summary, we have shown that the two commonly used forms of the interaction Hamiltonian in the electric dipole approximation yield identical results so long as the atomic wavefunction is properly transformed when using the minimal coupling formulation. Nonetheless, when these formulations are used within the context of the common approximations of resonance quantum optics, the rotating wave approximation and the two-level approximation, they lead to different predictions for measurable quantities. We have also seen that for the case of the calculation of the refractive index of a collection of atoms, these differences are large enough to be measured by standard laboratory techniques.

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