Solvable approximate model for the harmonic radiation from atoms subjected to oscillatory electric fields

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We develop a nonperturbative model for the response of atomic electrons to oscillating external electric fields, valid in the physical regime where the central electric force is well described by an inverse square law and where the driven oscillations preserve the principal quantum number. We use the model to calculate the odd-harmonic electric-dipole radiation from a single atom driven by a laser field. The exact time-dependent eigensolutions of the model illustrate Zel'dovich's concept of quasi-energy for sinusoidally time-dependent Hamiltonians. The validity and applicability of the model and suggested improvements are briefly discussed.

1. INTRODUCTION

The behavior of atoms in oscillatory electric fields has been studied extensively[1–3] in recent years. The complexity of the interacting system, coupled with the necessity for nonperturbative methods, effectively precludes any nonnumeric approach if accuracy is essential. Yet in such situations analytic approximations, even of inherently limited accuracy (such as one-dimensional models) can be of value in affording qualitative insights.

Our purpose in the present paper is to develop a comprehensible analytic model intended to afford some understanding of harmonic radiation produced by the nonlinear response of driven electrons in a Coulomb field. The exact solvability of our model is obtained at the cost of completely neglecting multiphoton ionization. We believe, however, that there are physical regimes in which our model is realistic. The effects that we find certainly exist even if they must compete with ionization processes that predominate in other physical regimes.

The Hamiltonian for the problem is time dependent and, being sinusoidal in the steady state, it obeys the symmetry

$$H\left(t+\frac{2\pi}{\omega}\right)=H(t), \tag{1.1}$$

where ω is the circular frequency of the incident laser field. For a time-dependent Hamiltonian, the familiar concept of characteristic energies (and of energy eigenstates) loses its meaning. Because the problem is nonperturbative, the artifice of using the characteristic energies of a time-independent zeroth-order Hamiltonian H_0 as a basis fails. If we have the time-displacement symmetry [Eq. (1.1)] for our Hamiltonian, however, we can use the concept of quasi-energy, which appeared in early works by Shirley[4] and, independently, by Zel'dovich.[5] (This concept is implicit in the Floquet theorem for differential equations). Quasi-energies, like the analog of quasi-momentum for a spatially periodic crystalline lattice, are conserved only modulo ($\hbar\omega$); thus the radiation of harmonics $N\hbar\omega$ from a sinusoidally time-dependent system in the nonperturbative regime is both easily explicable and characteristic. In particular, the quasi-energy concept readily shows that the problem of the nonexistence of suitable levels for the sequential absorption of, say, 19 quanta to produce the 19th harmonic is really no problem at all, only an indication of the inapplicability[6] of perturbative techniques and perturbation-based concepts. One of the purposes of the present paper is to show explicitly, in an exactly solvable model, the usefulness and naturalness of the concept of quasi-energy in understanding harmonic production nonperturbatively.

The actual physical problem of an atom in a strong laser field is complicated, and there is little hope for a comprehensive, physically understandable, nonnumeric solution. To isolate the relevant phenomena in a solvable way we must make approximations.

In Section 2 we use a schematic model approximating the complicated many-electron distribution in the atom by one (possibly more) spinless electron in an outer (unfilled) shell, moving nonrelativistically under the influence of a pure Coulombic field. This is in itself a major simplification, but the problem (the time-dependent Coulomb–Stark effect) is still intractable, and we must approximate even further.

To motivate our approximation, consider the classical (one-electron) Coulomb problem in an oscillating electric-dipole field. If the electron's Kepler-orbit frequency is not near the frequency of the dipole field, then the energy transfer averaged over the electron period is small. Moreover, the dipole moment of the atom, averaged over a period, remains oriented along the direction of the semimajor axis of the elliptical orbit. Translating these classical considerations into quantum-mechanical terms, we find that the approximation to be made is that the principal quantum number (N) is preserved under the perturbation. Making this further approximation, we arrive at a nontrivial time-dependent model Hamiltonian that is completely solvable. It is one of a small number of completely solvable, time-dependent model Hamiltonians and should provide an excellent basis for a perturbation theory when we use more exact Hamiltonians of atoms in strong oscillatory fields—a further development that we defer for the present.

In Section 3 we use the model to calculate the harmonic radiation produced by these laser-driven electron oscillations of a single atom. We calculate the cross sections for both polarized and unpolarized measurements and find the

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result that the harmonic radiation is polarized perpendicularly to the driving radiation. (In Section 4 we discuss why this possibly surprising result is actually physically reasonable.)

In Section 4 we present our conclusions and discuss the validity and applicability of the approximations that we have found necessary to achieve this comprehensible model.

2. THE MODEL AND ITS SOLUTIONS

The Hamiltonian for an electron in the Coulomb field of a nucleus with effective charge Ze is

$$H_0 = \frac{p^2}{2m} - \frac{Ze^2}{r}. \tag{2.1}$$

It is well known that the symmetry of the Coulomb field[7] implies that the (dimensionless) vector operators, the angular momentum operator

$$\mathbf{L} = \mathbf{r} \times \mathbf{p}/\hbar, \tag{2.2}$$

and the Laplace–Lenz–Runge operator

$$\mathbf{A} = (2mc\alpha Z)^{-1}(\mathbf{L} \times \mathbf{p} - \mathbf{p} \times \mathbf{L}) + \hat{r}, \tag{2.3}$$

or equivalently—as proves much more convenient—the renormalized operator

$$\mathbf{K} = (mc\alpha Z)(-2mH_0)^{-1/2}\mathbf{A} \tag{2.4}$$

each commute with the Hamiltonian H_0,

$$[H_0, \mathbf{L}] = [H_0, \mathbf{A}] = [H_0, \mathbf{K}] = 0, \tag{2.5}$$

and are therefore conserved (constant) in time. We remark that the Laplace–Lenz–Runge operator lies in the plane of the orbit and points along the major axis of the elliptical orbit in the classical Kepler problem.

The problem that we wish to solve adds the external electromagnetic (laser) driving field to the Hamiltonian H_0 so that in the dipole approximation the total Hamiltonian becomes

$$H = H_0 + e\mathbf{r} \cdot \mathbf{E}_0 \cos \omega t. \tag{2.6}$$

The Hamiltonian given in Eq. (2.6) is the Hamiltonian describing the time-dependent (spinless) Stark effect in a pure Coulomb field. Like the time-independent Stark effect, this Hamiltonian is separable in parabolic coordinates, but this is not of much use because the behavior at infinity of a dipole-electric field is singular (in the static case, all characteristic energies lie in the continuum). Physically, we regard this poor behavior at infinity as a flaw resulting from an overidealization of the perturbing electric field. Because of the degeneracy of the different angular-momentum states ($L = 0, 1, \ldots, N-1$) belonging to an N shell, the Stark effect is very effective in mixing angular momenta (as in, for example, the Stark-mixing phenomenon in meson capture[8]). We will find that a similar effect is important in radiation phenomena.

To obtain a solvable model we must approximate the Hamiltonian in Eq. (2.6), and to this end we recall that, classically, the vector $\mathbf{r}$ when averaged over a cycle (transversing the orbit) necessarily points along the major axis. This is along the direction given by the Laplace–Lenz–Runge vector, which is constant in time. Thus we expect that

$$(\mathbf{r})_{\text{averaged}} = (\text{numerical constant}) \cdot \mathbf{K}. \tag{2.7}$$

If we express this approximation in quantum-mechanical terms, we arrive at the famous approximation used by Pauli[9] in his discussion of the mixed Stark–Zeeman effect. That is,

$$\mathbf{r} = \frac{3}{2} a_0\left(\frac{N}{Z}\right)\mathbf{K} + i[\mathbf{S}, H_0], \tag{2.8}$$

where[4]

$$\mathbf{S} = m\hbar(\alpha Zmc)^{-2}(N^2\mathbf{T} + \mathbf{T}^\dagger N^2), \tag{2.9a}$$

$$\mathbf{T} = \hbar\mathbf{L} \times \mathbf{r} + \mathbf{r} \cdot \mathbf{rp}; \tag{2.9b}$$

the dagger indicates a Hermitian conjugate.

Clearly, if we take diagonal matrix elements (diagonal in the principal quantum number N) for the operator $\mathbf{r}$, the commutator term in Eq. (2.8) drops out. Thus Pauli's approximation (which is quantum-mechanically equivalent to averaging over orbit cycles) is to replace the operator $\mathbf{r}$ by its diagonal component only:

$$\mathbf{r} \to \frac{3}{2} a_0\left(\frac{N}{Z}\right)\mathbf{K}. \tag{2.10}$$

In other words, we approximate such that $\mathbf{r}$ causes transitions between states of differing angular momentum (and parity), but it cannot cause transitions between different N shells (that is, differing energy eigenvalues). At first glance this approximation seems to forbid radiation. (Where does the emitted energy come from if the transitions are between states of the same energy only?) However, we will see shortly that this is not true.

With these restrictions, the Hamiltonian of Eq. (2.6) can be replaced by the model Hamiltonian

$$H_{\text{model}} \equiv H_0 + \frac{3}{2} a_0\left(\frac{N}{Z}\right) e\mathbf{E}_0 \cdot \mathbf{K} \cos \omega t. \tag{2.11}$$

The time-dependent model Hamiltonian given in Eq. (2.11) is exactly solvable, as we will now show in detail.

Let us first discuss the conventional symmetries of Eq. (2.11). The Coulomb part of the Hamiltonian, H_0 of Eq. (2.1), has the two symmetry generators $\mathbf{L}$ and $\mathbf{K}$, which, as mentioned above, are constants of the motion [Eq. (2.5)]. These two vector operators obey the commutation relations

$$[L_i, L_j] = iL_k, \tag{2.12a}$$

$$[L_i, K_j] = iK_k, \tag{2.12b}$$

and

$$[K_i, K_j] = iL_k, \tag{2.12c}$$

where (i, j, k) is a cyclic permutation of $(1, 2, 3)$.

These commutation relations admit, as a constant of the motion, the principal quantum number operator[7] given by

$$N_{\text{op}}{}^2 \equiv \mathbf{L}^2 + \mathbf{K}^2 + 1, \tag{2.13}$$

which has the eigenvalues

$$N_{\text{op}}{}^2 \to N^2, \qquad \text{with } N = 1, 2, \ldots. \tag{2.14}$$

[For clarity, we will distinguish operators from numbers

(eigenvalues) by the subscript op. Strictly speaking, the symbol N in Eqs. (2.8) and (2.11) and expression (2.10) should be replaced by $N = (N_{op}{}^2)^{1/2}$.] The Hamiltonian H_0 may itself be expressed in terms of the principal quantum number operator:

$$H_0 = -\frac{1}{2}\frac{mc^2(\alpha Z)^2}{N_{op}{}^2}. \tag{2.15}$$

For the Coulomb Hamiltonian H_0 there are two possible choices for the remaining commuting constants of the motion:

(1) Spherical basis:

$$N_{op}{}^2 \to N^2, \qquad N = 1, 2, \ldots; \tag{2.16a}$$

$$L_{op}{}^2 \to L(L+1), \qquad L = 0, 1, \ldots, N-1; \tag{2.16b}$$

$$(L_3)_{op} \to M, \qquad -L \le M \le L \ (M = \text{integer}). \tag{2.16c}$$

The orthonormal eigenket vectors in this basis are denoted by $|NLM\rangle$.

(2) Parabolic basis:

$$N_{op}{}^2 \to N^2, \qquad N = 1, 2, \ldots; \tag{2.17a}$$

$$\frac{1}{2}(L_3 + K_3)_{op} \to \mu, \qquad -\left(\frac{N-1}{2}\right) \le \mu \le \left(\frac{N-1}{2}\right), \tag{2.17b}$$

$$\frac{1}{2}(L_3 - K_3)_{op} \to \nu, \qquad -\left(\frac{N-1}{2}\right) \le \nu \le \left(\frac{N-1}{2}\right), \tag{2.17c}$$

where μ and ν are each half-integer or integer depending on whether $N-1$ is odd or even, respectively. The orthonormal eigenkets of the parabolic basis will be denoted by $|N\mu\nu\rangle$.

Turning now to the model Hamiltonian of Eq. (2.11), we see that the principal quantum number operator $N_{op}{}^2$ is a constant of the motion because

$$[N_{op}{}^2, \mathbf{K}] = 0, \tag{2.18}$$

which follows from the commutation relations.

However, we see at once that $L_{op}{}^2$ is no longer a constant of the motion because $[\mathbf{L}_{op}{}^2, \mathbf{K}] \neq 0$. Thus the spherical basis is not an eigenbasis for the model Hamiltonian.

If we choose the incident laser field to define the $\hat{z}$ axis, that is,

$$\mathbf{E}_0 \| \hat{z}, \tag{2.19}$$

then we see that both $(K_3)_{op}$ and $(L_3)_{op}$ commute with the model Hamiltonian [Eq. (2.11)]. It follows that the parabolic basis is an eigenbasis that is well adapted to the Hamiltonian of Eq. (2.11).

In quantum mechanics there are relatively few exactly solvable model problems, and of these few even fewer are time dependent. Perturbation theory reflects this fact and is almost exclusively based on assuming a time-independent solvable Hamiltonian. Because we intend to treat the generation of harmonic radiation nonperturbatively by using the time-dependent model Hamiltonian of Eq. (2.11), it is useful to discuss some of the unfamiliar features of such a system briefly.[5]

The first point to note is that the Hamiltonian in Eq. (2.11) has the symmetry

$$H_{\text{model}}\left(t + \frac{2\pi}{\omega}\right) = H_{\text{model}}(t) \tag{2.20}$$

because the interaction is sinusoidal in time. It follows from the time-dependent Schrödinger equation

$$H_{\text{model}}(t)\psi(\mathbf{r}, t) = i\hbar\frac{\partial\psi}{\partial t}(\mathbf{r}, t) \tag{2.21}$$

that there exist solutions ψ such that $\psi(t + 2\pi/\omega)$ differs from $\psi(t)$ by at most a phase factor. That is, there exist solutions such that

$$\psi\left(t + \frac{2\pi}{\omega}\right) = e^{-i\varphi}\psi(t). \tag{2.22}$$

In analogy to the time-independent case, we relate the phase φ to a quasi-energy:

$$\hbar\varphi = (E)\left(\frac{2\pi}{\omega}\right). \tag{2.23}$$

This situation is in exact analogy to that of a crystal lattice for which the spatial periodicity results in a replacement of the concept of momentum by that of quasi-momentum.[10]

Let us identify the states by the associated quasi-energy, that is, we define

$$\psi_{E_i}(\mathbf{r}, t) \equiv \exp(-iE_i t/\hbar)\Phi_{E_i}(\mathbf{r}, t), \tag{2.24}$$

where

$$\Phi_{E_i}\left(\mathbf{r}, t + \frac{2\pi}{\omega}\right) = \Phi_{E_i}(\mathbf{r}, t). \tag{2.25a}$$

It follows from Eqs. (2.23) and (2.24) that the quasi-energies E_i are well defined only mod $\hbar\omega$. In a transition from a state of quasi-energy E_{initial} to a state of quasi-energy E_{final}, the radiation has the circular frequency

$$\omega_{\text{emitted}} = \hbar^{-1}(E_{\text{initial}} - E_{\text{final}}) + N\omega, \tag{2.25b}$$

where

$$N = 0, \pm 1, \ldots. \tag{2.25c}$$

In particular, *harmonics of the driving radiation can be emitted in transitions between states of the same quasi-energy*. Radiation involving transitions between states of different quasi-energy are, in general, Raman-type spectra.

It is a general property that states of different quasi-energy are orthogonal, as can be shown directly from the Schrödinger equation, Eq. (2.21).

Let us proceed now to determine the quasi-energy eigensolutions of the model Hamiltonian explicitly. This is not difficult when the results developed above are used for the eigenenergies of H_0 [Eq. (2.15)] and for the constants of the motion [expressions (2.17)]. Because the Hamiltonian [Eq. (2.11)] is time dependent, the Schrödinger equation [Eq. (2.21)] implies that we have time-dependent eigenkets, which we denote by $|N\mu\nu; t\rangle$. Putting these equations together shows that the time-dependent eigenkets obey the differential equation

$$i\hbar \frac{d}{dt}|N\mu\nu; t\rangle = H_{\text{model}}|N\mu\nu; t\rangle$$
$$= \left\{-\left[\frac{mc^2(\alpha Z)^2}{2N^2}\right] + \frac{3}{2}\frac{a_0 N}{Z} eE_0 (\mu - \nu)\cos \omega t\right\}|N\mu\nu; t\rangle. \quad (2.26)$$

[The change from a partial to a total time derivative in Eq. (2.26) results from the fact that N, μ, and ν are independent of time so that only the explicit t is time dependent. The derivative is therefore necessarily as written.]

It is not difficult to integrate Eq. (2.26) and to obtain the time-dependent eigenkets. We find that

$$|N\mu\nu; t\rangle = \exp[-if(t)]|N\mu\nu\rangle, \quad (2.27a)$$

where

$$f(t) \equiv -\frac{1}{2}\frac{mc^2}{\hbar}\left(\frac{\alpha Z}{N}\right)^2 t + \frac{3}{2}\frac{ea_0 NE_0}{Z}(\mu - \nu)\frac{\sin \omega t}{\hbar\omega}. \quad (2.27b)$$

{The time-independent eigenkets $|N\mu\nu\rangle$ in Eq. (2.27a) are eigenkets of the Coulomb Hamiltonian [Eq. (2.1)] in the parabolic basis [Eqs. (2.17)].} Note that Eq. (2.27b) exhibits an exact result for the dynamical Stark effect in this model.

To complete the determination of the quasi-energies and the associated quasi-energy states, we use the defining Eq. (2.24). It follows that the quasi-energies E_i are exactly the original unperturbed Coulomb energies:

$$E_i \equiv E_N = -\frac{1}{2}mc^2\left(\frac{\alpha Z}{N}\right)^2. \quad (2.28)$$

The quasi-energy time-dependent eigenfunctions, denoted by $\Phi_{E_i}(\mathbf{r}, t)$ in Eq. (2.25a), now have the explicit form (noting that $E_i = E_N$ as above)

$$\Phi_{E_i}(\mathbf{r}, t) = \exp\left[\frac{3}{2} i \frac{ea_0 NE_0}{Z}(\mu - \nu)\frac{\sin \omega t}{\hbar\omega}\right]\langle \mathbf{r}|N\mu\nu\rangle, \quad (2.29)$$

with $\langle \mathbf{r}|N\mu\nu\rangle$ denoting the parabolic time-independent Coulomb eigenfunctions in configuration coordinates.

Note that these quasi-energy eigenfunctions in Eq. (2.29) are invariant under a time displacement: $t \to t + 2\pi/\omega$. Note, too, that the orthogonality of the quasi-energy eigenfunctions in Eq. (2.29) is now manifestly true from the orthogonality of the parabolic functions $\langle \mathbf{r}|N\mu\nu\rangle$.

It is important to note that the set of all linearly independent eigensolutions with different quasi-energies form a complete set of functions at any given instant of time. This can be seen from the explicit form of the set of quasi-energy eigensolutions $\{\Phi_{E_i}(\mathbf{r}, t)\}$ [Eq. (2.29)]. At any instant of time the time-dependent coefficients have fixed values, and the completeness of the parabolic basis set $\{\langle \mathbf{r}|N\mu\nu\rangle\}$ then guarantees the desired result. [Zel'dovich remarks (Ref. 5, p. 1008, footnote) that an electron in a Coulomb field with the dipole interaction of Eq. (2.6) does not have a discrete quasi-energy eigenspectrum. This is not in contradiction to our results, as our approximation for the operator $\mathbf{r}$, expression (2.10), removes the singular behavior at spatial infinity of the interaction.] Let us note that this property of orthogonality and completeness shows that an arbitrary exact solution of the Hamiltonian, Eq. (2.11), will have constant expansion coefficients when expanded on the complete set of quasi-energy eigenstates.

3. HARMONIC RADIATION FROM DRIVEN ELECTRON OSCILLATIONS

In this section we will develop the harmonic radiation from the driven electron oscillations described by the time-dependent model Hamiltonian, Eq. (2.11). Let us sketch, in words, the standard perturbation approach for radiative transitions before we proceed to give the details of this calculation. Briefly, we adjoin to the model Hamiltonian the electromagnetic free-field Hamiltonian and the matter–field interaction Hamiltonian. Using the matter–field interaction Hamiltonian, we calculate, perturbatively but field-theoretically, the transition operator between Fock states of the electromagnetic field. This yields the effective transition operator[11] for the vector potential in interaction with the charged material system:

$\mathcal{O}^\alpha \equiv$ emission operator

$$= c\left[\frac{(n_{k,\alpha} + 1)\hbar}{2\omega V}\right]^{1/2} \epsilon^\alpha \exp[-i(\mathbf{k}\cdot\mathbf{r} - \omega t)], \quad (3.1)$$

where $\epsilon^\alpha \exp[-i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$ is the operator describing the emission of a photon $(\mathbf{k}, \omega)$ with four-vector polarization ϵ^α, V is the quantization volume, and $n_{k,\alpha}$ is the relevant occupation number of the photon field (we put $n_{k,\alpha} = 0$ because we consider spontaneous emission). The effective interaction Hamiltonian to be added to Eq. (2.11) is

$$H_{\text{int}} = j_\alpha \mathcal{O}^\alpha, \quad (3.2)$$

where j_α is the four-current of the electron.

For transitions between sharp angular-momentum states, we develop the plane-wave states $\epsilon^\alpha \exp(-i\mathbf{k}\cdot\mathbf{r})$ into electromagnetic multipoles[7]; this is particularly simple for electric dipoles. Noting that the time factor $e^{i\omega t}$ implies a Fourier decomposition of the effective current in Eq. (3.2), we may summarize the calculation for electric-dipole radiation, using Eq. (3.2), as simply the calculation of the Fourier components of the time-dependent matrix elements of the electric-dipole operator $e\mathbf{r}$, properly normalized. Once again we use Pauli's approximation, expression (2.10), which greatly simplifies the calculation.

The calculation of driven electric-dipole radiation is then, in effect, the calculation of the time-dependent matrix elements of the Laplace–Runge–Lenz vector operator $\mathbf{K}$. There are two equivalent ways that we may carry out this calculation: one uses either time-dependent eigenstates (as described in Section 2) and the time-independent operator $\mathbf{K}$ or, equivalently, vice versa. We shall use the latter, making use of the explicit form, Eqs. (2.27).

Because both $\mathbf{L}$ and $\mathbf{K}$ commute with H_0, we see that only the interaction in H_{model} induces a time dependence in $\mathbf{L}$ or $\mathbf{K}$. We find that

$$\mathbf{K}(t) \equiv \mathcal{A}^{-1}(t)\mathbf{K}\mathcal{A}(t), \qquad \mathbf{L}(t) \equiv \mathcal{A}^{-1}(t)\mathbf{L}\mathcal{A}(t), \quad (3.3a)$$

where

$$\mathcal{A}(t) \equiv \exp\left[\frac{-i}{\hbar}\int_0^t H_{\text{model}}(t')dt'\right]. \quad (3.3b)$$

The Hamiltonian H_0 drops out of the calculation, as noted above, and, using the integral given in Eq. (2.27b), with $(\mu - \nu)E_0 \to \mathbf{K}\cdot\mathbf{E}_0$), we find that[7]

$$\mathbf{K}(t) = [\cos\Gamma(t)]\hat{E}_0 \times [\mathbf{K}(0) \times \hat{E}_0] + [\sin\Gamma(t)](\hat{E}_0 \times \mathbf{K}(0)] + \hat{E}_0[\hat{E}_0 \cdot \mathbf{K}(0)], \tag{3.4a}$$

$$\mathbf{L}(t) = [\cos\Gamma(t)]\{\hat{E}_0 \times [\mathbf{L}(0) \times \hat{E}_0]\} + [\sin\Gamma(t)][\hat{E}_0 \times \mathbf{K}(0)] + \hat{E}_0[\hat{E}_0 \cdot \mathbf{L}(0)], \tag{3.4b}$$

where

$$\Gamma(t) \equiv \frac{3N}{2Z}\left(\frac{ea_0E_0}{\hbar\omega}\right)\sin\omega t. \tag{3.4c}$$

We note that the kinematical constraint $\mathbf{L} \cdot \mathbf{K} = 0$ is valid because direct calculation from Eqs. (3.4) shows that

$$\mathbf{L}(t) \cdot \mathbf{K}(t) = 0. \tag{3.5}$$

Thus the time-dependent electric-dipole operator $\mathbf{d}(t) \equiv e\mathbf{r}(t)$ in Pauli's approximation becomes

$$\mathbf{d}_{\text{model}}(t) = \left(\frac{3N}{2Z}\right)(ea_0)\mathbf{K}(t). \tag{3.6}$$

We see at once from Eqs. (3.4) and (3.6) that the dipole moment is not harmonic in time but is frequency modulated, which will lead to copious harmonic production.

To proceed further, the standard theory sketched above calculates the Fourier component of the effective dipole moment [Eq. (3.6)] at the harmonic frequency $M\omega$. Because purely harmonic radiation does not involve any change in the characteristic quasi-energies [see Eqs. (2.25)], we see that we need only calculate the Fourier transform of the term $\sin\Gamma(t)$. [For transitions between states of the same parity, the matrix elements of the dipole operator, Eq. (3.6), necessarily reduce to the $\sin\Gamma(t)$ term only. We can easily check that this is in agreement with results found in the perturbation theory limit.] Using the relation[12]

$$\sin[z(\sin\theta)] = 2\sum_{k=0}^{\infty} J_{2k+1}(z)\sin[(2k+1)\theta], \tag{3.7}$$

we find that the Fourier components of the effective dipole operator, Eq. (3.6), are given by

$$\mathbf{d}(M\omega) = \frac{3i}{2}\left(\frac{Nea_0}{Z}\right)J_M\left(\frac{3}{2}\frac{Nea_0E_0}{Z\hbar\omega}\right)\hat{E}_0 \times \mathbf{L}(0), \tag{3.8}$$

Here M is an odd integer (positive). That only odd harmonics occur for electric-dipole radiation accords with the general result for atomic systems (see Ref. 1, p. 94).

Let us now calculate the magnitude of the electric-dipole radiation and then the cross section for harmonic production. Classically the radiated power[7] is given by

$$\text{Radiated power for } M\text{th harmonic} = \frac{1}{2}\left(\frac{M\omega}{c}\right)^2\left[\frac{\mu_0}{\epsilon_0}\right]^{1/2}\sum_m |n_{1,m}|^2, \tag{3.9}$$

where $n_{1,m}$ is the mth spherical component of the electric dipole multipole moment, which (in the present calculation) is given by

$$n_{1,m} = (6\pi)^{-1/2}\left(\frac{3M\omega}{2}\right)\left(\frac{ea_0N}{Z}\right)J_M\left(\frac{3}{2}\frac{ea_0E_0N}{\hbar\omega Z}\right)[\hat{E}_0 \times \mathbf{L}(0)]_m \tag{3.10}$$

$$\equiv \mathcal{C}[\hat{E}_0 \times \mathbf{L}(0)]_m, \tag{3.11}$$

where we have collected all numerical factors in the constant $\mathcal{C}$ for ease of writing. The quantal calculation replaces the sum $\sum(\ldots)$ by a sum over final states and an average over initial states of the quantal matrix elements, that is,

$$\sum_m |n_{1,m}|^2 = |\mathcal{C}|^2(N^2)^{-1}\sum_{\substack{\mu,\nu \\ \mu',\nu' \\ m}} |\langle N\mu'\nu'|(\hat{E}_0 \times \mathbf{L})_m|N\mu\nu\rangle|^2 \tag{3.12}$$

$$= \tfrac{1}{3}|\mathcal{C}|^2(N^2-1). \tag{3.13}$$

To calculate the cross section, $\sigma(M)$, we use the definition

$$\text{cross section for } M\text{th harmonic} \equiv \sigma(M) \equiv \frac{\text{Radiated power}/\hbar M\omega}{\text{Energy flux incident}/\hbar\omega}. \tag{3.14}$$

Introducing the results of Eqs. (3.9)–(3.13) in Eq. (3.14) and evaluating the incident flux for a field strength $\mathbf{E}_0$, we find for the cross section for radiating the Mth harmonic that

$$\sigma(M) = (6\pi)(a_0)^2M^3\frac{N^2(N^2-1)}{Z^2}\left(\frac{\omega a_0}{c}\right)^4\left(\frac{e}{4\pi\epsilon_0 a_0{}^2E_0}\right)^2 \times\left[J_M\left(\frac{3}{2}\frac{eE_0a_0N}{Z\hbar\omega}\right)\right]^2 \quad (\text{with } M = \text{odd integer}). \tag{3.15}$$

In Eq. (3.15) we note the following:

1. For the principal quantum number $N = 1$, the angular momentum vanishes, and Eq. (3.15) reduces properly to zero.
2. The only dimensional factor in Eq. (3.15) is $(a_0)^2$, the square of the Bohr radius, as one would expect *a priori*.

We will discuss the characteristic features of Eq. (3.15) in Section 4.

To conclude this section, let us now calculate the polarization of the emitted harmonics. The transition operator in Eq. (3.8) shows that the matrix elements for linearly polarized incident radiation ($\hat{\epsilon}_{\text{inc}} \| \hat{E}_0$) and measurements of linearly polarized harmonic radiation ($\hat{\epsilon}_{\text{out}}$) depend on angles as

$$\text{Matrix element} \propto \hat{\epsilon}_{\text{out}} \times \hat{\epsilon}_{\text{inc}} \cdot \mathbf{L}(0). \tag{3.16}$$

It follows that the angular distribution W for this case is given by

$$W \propto (\hat{\epsilon}_{\text{inc}} \times \hat{\epsilon}_{\text{out}})^2. \tag{3.17}$$

For unpolarized incident radiation, the angular distribution becomes

$$W(\theta') \propto \tfrac{1}{2}(1 + \cos^2\theta'), \tag{3.18}$$

where $\cos\theta' = \hat{\epsilon}_{\text{out}} \cdot \hat{k}_{\text{inc}}$.

In the opposite case, i.e., linearly polarized incident radia-

tion and unpolarized measurements, we obtain the angular distribution:

$$W(\theta'') \propto \tfrac{1}{2}(1 + \cos^2\theta''), \tag{3.19}$$

with $\cos\theta'' \equiv \hat{\epsilon}_{\text{in}} \cdot \hat{k}_{\text{out}}$.

Finally, when neither polarization is measured we obtain the result

$$W(\theta) \propto \tfrac{1}{4}(2 + \sin^2\theta), \tag{3.20}$$

with $\cos\theta \equiv \hat{k}_{\text{inc}} \cdot \hat{k}_{\text{out}}$.

It is useful to recall that the angular distribution for Thomson scattering is given by

$$W(\theta)|_{\text{Thomson}} \propto \tfrac{1}{4}(1 + \cos^2\theta); \tag{3.21}$$

thus the result given in relation (3.20) is unexpected and counterintuitive. (We discuss why this result is actually physically reasonable in Subsection 4.C.)

4. CONCLUDING REMARKS

A. Dependence on Harmonic Number (M)

If we examine Eq. (3.15) to determine the parametric dependence of the cross section on the order of the harmonic, we see that the cross section for the Mth harmonic has the behavior

$$\sigma(M) \propto M^3[J_M(z)]^2, \tag{4.1}$$

where

$$z \equiv \frac{3}{2}\left(\frac{ea_0NE_0}{Z\hbar\omega}\right). \tag{4.2}$$

The dependence M^3 is easily recognized as arising from the typical frequency dependence of dipole radiation, and the Bessel function factor is typical of the frequency-modulation process.

The parameter that determines the various regimes characterizing harmonic production is denoted by z in Eq. (4.2) and can be seen to be, in essence, the work done on an electron in moving it through a typical atomic distance (Na_0/Z), as compared with the quantum energy of the incident photon field. There are three regimes:

1. $z \ll M$: In this case the Bessel function is limited by the centrifugal barrier and has the behavior[12]

$$J_M(z) \to (\tfrac{1}{2}z)^M/\Gamma(M+1) \tag{4.3}$$

such that the cross section for the Mth harmonic $\to (E_0)^{2M}$. We recognize this as the weak-field perturbation limit with its attendant sequential (matrix element $\propto |\mathbf{E}_0|$) interactions.

2. $z \approx M$: This is the turning-point regime[12] for the Bessel function (where it achieves a maximum before going into the oscillatory regime). We have

$$J_M(M) \sim \left(\frac{2}{9M}\right)^{1/3}\frac{1}{\Gamma(2/3)}. \tag{4.4}$$

3. $z \gg M$: This is the oscillatory region[12] for which we have

$$J_M(z) \sim \left(\frac{2}{\pi z}\right)^{1/2}\cos\left(z - \frac{2M+1}{4}\pi\right). \tag{4.5}$$

The typical behavior of a generic cross section ($z \approx 10$ or so) as M increases from 1 through the odd integers is then a region of small sizes (varying randomly in appearance), then an increase to a maximum at $M \sim z$, followed by an exponential decline for large M. Note, in particular, that *lower harmonics are not necessarily more copiously produced than higher harmonics in the transition region.*[6]

This general behavior of the cross section for harmonic production should be more or less typical and valid when the relevant parameters of N and Z are taken to have effective values characterizing a given experiment.

It is interesting to note that the dimensionless parameter z [Eq. (4.2)], which determines the various regimes discussed above, is essentially the same parameter that governs a different experimental situation, the percentage of atoms ionized by strong incident electromagnetic radiation, originally calculated by Keldysh[13] and extended by others.[14–16]

The Keldysh parameter

$$\gamma \equiv \omega(2mI)^{1/2}/eE_0 \tag{4.6}$$

(where I is the ionization energy) distinguishes the regime of strong (~100%) ionization from the regime of exponentially small ionization. The turnover takes place at $\gamma \approx 1$.

If we use the Coulomb energy $[\tfrac{1}{2}mc^2(\alpha Z)^2/N^2]$ for the ionization energy, then the parameters z and γ are related by

$$z = 3/2\gamma. \tag{4.7}$$

Qualitatively, then, we see that the same characteristic parameter typifies the very different processes of ionization and coherent harmonic production. This *a posteriori* agreement indicates that although our approximations are quite severe, some of the essential physics is qualitatively preserved. As Delone and Krainov remark (Ref. 1, p. 75), the Keldysh calculation itself involves drastic approximations, but the entire body of experimental data on nonlinear ionization of atoms (they assert) confirms the general conclusions derived from the theory.

B. Comparison with Experiment

Although the experiments of Rhodes and McPherson *et al.*[3] were the proximate cause for the present investigation, and although comparison of our result, Eq. (3.15), to their experiment agrees in order of magnitude and exponential falloff, we still cannot claim an explanation for their results. The reason is that the density of the gas irradiated in the Rhodes *et al.* experiment, although small enough to exclude the dominance of collisional processes, is still too large to consider single-atom processes. There are several hundred atoms within a cubic wavelength so that interference effects will predominate in a way that is exceedingly complicated to extricate. Similarly, the observed polarization effects cannot be expected to bear any resemblance to the polarizations calculated in Section 3.

C. Polarization of the Harmonics

The harmonics radiated from the driven atom were found (in Section 3) to be polarized perpendicular to the polarization of the driving radiation. This is a counterintuitive result. It is not so strange, however, if we consider the time evolution of the Laplace–Runge–Lenz vector of an atom in a static electric field. If we take the static-field form of Eq. (2.11), we have

$$H = H_0 + (3/2)a_0(N/Z)e\mathbf{E}_0 \cdot \mathbf{K}, \tag{4.8}$$

and from Eqs. (2.4), (2.8), (2.9), and (2.12), it can be shown[17] that the classical equation of motion is

$$\frac{d}{dt}(\mathbf{L} \pm \mathbf{K}) = -\{\mathbf{L} \pm \mathbf{K}, H\}$$

$$= \pm\omega \times (\mathbf{L} \pm \mathbf{K}) + \{\{e\mathbf{E}_0 \cdot \mathbf{S}, \mathbf{L} \pm \mathbf{K}\}, H_0\}, \tag{4.9}$$

where

$$\omega = (3/2)a_0(N/Z)e\mathbf{E}_0, \tag{4.10}$$

is the classical frequency and we use Poisson brackets. The last term in Eq. (4.9) can be neglected for nearly closed orbits, with the result that both **L** and **K** respond orthogonally to **E**.[17] Thus, if we consider orbital averages, the equations of motion simplify to

$$\frac{d\mathbf{L}}{dt} = \omega \times \mathbf{K} \tag{4.11a}$$

and

$$\frac{d\mathbf{K}}{dt} = \omega \times \mathbf{L}. \tag{4.11b}$$

Most importantly, **K**, which (in the Pauli approximation) is in the direction of the electric-dipole moment of the atom, responds orthogonally to $\mathbf{E}_0$, which qualitatively explains the effect that is found. It would be interesting to verify this predicted polarization effect experimentally, and an experiment on Rydberg atoms (or perhaps even excitons) with microwave radiation might be feasible.

D. Improved Treatment

It will hardly have escaped the reader that if indeed $\Delta N \neq 0$ effects cannot be neglected, then the nonperturbative results obtained in Sections 2 and 3 can be used as a basis for perturbatively including $\Delta N \neq 0$ transitions. We believe this is a feasible next step to improve the treatment of harmonic production, but we have not done so at this stage.

Let us conclude by remarking that our primary aim has been to show that the approximate Hamiltonian given by Eq. (2.11) admits of an exact solution useful in discussing both the physically important concept of quasi-energy and the discussion of a nontrivial model for harmonic production. It would be unrealistic to hope for too much accuracy from such a solvable model, but it should certainly be useful as a basis for more accurate further approximations.

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