Quantum Theory of Spontaneous Emission by Real Moving Atoms

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We outline the solution of a fundamental problem in quantum theory which has hitherto lacked a proper solution, namely, finding the requisite quantum theoretical framework guaranteeing that the calculated inverse spontaneous emission rate of a moving atom, as a composite system of charged particles interacting with the Maxwell field, is slowed down exactly as in time dilation.

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It has recently been shown that a neutral atom bearing an electric dipole moment moving in an external magnetic field can accumulate a quantum phase [1–5]. A moving dipole may, under suitable conditions, exhibit a detectable Aharonov-Bohm phase shift [6] and the rotational motion of a Bose-Einstein condensate in a vortex state can induce a magnetic monopole [7,8] distribution or an electric charge distribution [8]. These and other effects associated with atomic motion continue to receive considerable attention and especially so with the advent of atom optics [9,10] and laser cooling and trapping [11]. At first sight, it would appear that the requisite theory for the description of phenomena involving moving atoms could be constructed as a straightforward extension of nonrelativistic quantum optics by incorporating the translational motion of the atomic center of mass.

In fact, the need to incorporate the center of mass motion in quantum optics theory had necessitated a reappraisal of the corresponding nonrelativistic quantum electrodynamical theory where investigations sought to elucidate how the division of the motion into center of mass and internal motions is affected by the presence of the interaction with electromagnetic fields [12–15]. One of the main outcomes of these investigations was the emphasis on the role of the Röntgen interaction [16] energy term which couples the electric dipole moment to an effective electric field involving the center of mass velocity and the magnetic field of the light.

Wilkens [17] was the first to question the role of the center of mass motion in the process of spontaneous emission. In particular, he pointed out that a theory which excludes the Röntgen interaction would lead to spurious velocity-dependent effects when evaluating the spontaneous decay rate of an excited electric dipole moving freely in electromagnetic vacuum. Wilkens extended his work to include the Röntgen interaction and evaluated the scattering rate into a given solid angle in a given direction, deducing that this was free of any spurious velocity dependences [18]. He did not, however, proceed to ascertain whether the calculated *total* spontaneous emission rate based on his approach would be consistent with the requirements of special relativity. As we show here, the incorporation of the Röntgen interaction in the theory, albeit important, is not sufficient on its own to guarantee the emergence of a total spontaneous emission rate which conforms with relativity.

More recently, Barton and Calogeracos [19] highlighted the absence in the literature of a proper treatment of the quantum theory of spontaneous emission of atoms moving in a classically assigned trajectory. This is so even for the simplest case of a uniformly moving atom. As far as the authors are aware, a workable theoretical framework of the problem in which a real atom, as a composite structure, interacts with the full (vector) Maxwell field is hitherto unknown, and it is our purpose here to furnish such a framework.

The model of a real atom we consider here involves two oppositely charged particles of charges $e_1 = -e_2 = e$ and finite masses m_1 and m_2. In the center of mass frame (atomic frame), we denote the position vectors of the two particles by $\mathbf{q}_1'$ and $\mathbf{q}_2'$, and the electromagnetic scalar and vector potentials as $\phi'(\mathbf{r}')$ and $\mathbf{A}'(\mathbf{r}')$, respectively. The Lagrangian density for the electromagnetic field including the interaction with the two charged particles is

$$\mathcal{L}' = \frac{\epsilon_0}{2}\{[\dot{\mathbf{A}}'(\mathbf{r}') + \nabla'\phi'(\mathbf{r}')]^2 - c^2[\nabla' \times \mathbf{A}'(\mathbf{r}')]^2\} + \mathbf{J}'(\mathbf{r}')\cdot\mathbf{A}'(\mathbf{r}') - \rho'(\mathbf{r}')\phi'(\mathbf{r}'), \tag{1}$$

where the electric field is $\mathbf{E}'(\mathbf{r}') = -[\dot{\mathbf{A}}'(\mathbf{r}') + \nabla'\phi'(\mathbf{r}')]$, the magnetic field is $\mathbf{B}'(\mathbf{r}') = \nabla' \times \mathbf{A}'(\mathbf{r})$, and the current and charge densities of the particles are given, respectively, by $\mathbf{J}'(\mathbf{r}') = e[\dot{\mathbf{q}}_1'\delta(\mathbf{r}' - \mathbf{q}_1') - \dot{\mathbf{q}}_2'\delta(\mathbf{r}' - \mathbf{q}_2')]$ and $\rho'(\mathbf{r}') = e[\delta(\mathbf{r}' - \mathbf{q}_1') - \delta(\mathbf{r}' - \mathbf{q}_2')]$. The notation is such that the atomic frame (the rest frame) is referred to as S' and all quantities relative to this frame are primed. The laboratory frame is the unprimed frame and will be referred to as S, relative to which the atomic center of mass moves at velocity $\dot{\mathbf{R}}$, and all quantities relative to S are unprimed.

The Lagrangian density in Eq. (1) can be recast in terms of the primed center of mass coordinates, defined by $\mathbf{R}' = (m_1\mathbf{q}_1' + m_2\mathbf{q}_2')/M$ and $\mathbf{q}' = \mathbf{q}_1' - \mathbf{q}_2'$, where $M = m_1 + m_2$ is the atomic mass. Note that in the primed frame, or rest frame, we must have $\dot{\mathbf{R}}' = 0$, *by definition.* This allows us to carry out a Power-Zienau-Woolley gauge transformation [20] and straightforwardly obtain the new Lagrangian density,

$$\mathcal{L}' = \frac{\epsilon_0}{2}\{[\dot{\mathbf{A}}'(\mathbf{r}') + \boldsymbol{\nabla}'\phi'(\mathbf{r}')]^2 - c^2[\boldsymbol{\nabla}' \times \mathbf{A}'(\mathbf{r}')]^2\} - \mathcal{P}'(\mathbf{r}') \cdot [\dot{\mathbf{A}}'(\mathbf{r}') + \boldsymbol{\nabla}'\phi'(\mathbf{r}')] + \mathcal{M}'(\mathbf{r}') \cdot [\boldsymbol{\nabla}' \times \mathbf{A}'(\mathbf{r}')], \tag{2}$$

where the polarization and magnetization vectors are expressed as full multipolar series in closed analytical forms,

$$\mathcal{P}'(\mathbf{r}') = \sum_{i=1,2} e_i \int_0^1 d\lambda(\mathbf{q}'_{\mathbf{i}} - \mathbf{R}')\, \delta[\mathbf{r}' - \mathbf{R}' - \lambda(\mathbf{q}'_{\mathbf{i}} - \mathbf{R}')], \tag{3}$$

$$\mathcal{M}'(\mathbf{r}') = \sum_{i=1,2} e_i \int_0^1 d\lambda\lambda(\mathbf{q}'_{\mathbf{i}} - \mathbf{R}') \times \dot{\mathbf{q}}'_{\mathbf{i}}\, \delta[\mathbf{r}' - \mathbf{R}' - \lambda(\mathbf{q}'_{\mathbf{i}} - \mathbf{R}')]. \tag{4}$$

The Lagrangian density in Eq. (2) has a manifestly covariant form, viz.,

$$\mathcal{L}' = -\frac{\epsilon_0}{4} F'^{\mu\nu} F'_{\mu\nu} - \frac{1}{2} G'^{\mu\nu} F'_{\mu\nu}, \tag{5}$$

where in the primed S' frame $F'_{\mu\nu}$ is the well-known electromagnetic field four-tensor [21] and $G'_{\mu\nu}$ is the polarization field four-tensor [22]. Formally, $G'^{\mu\nu}$ has the same form as $F'^{\mu\nu}$ but with the substitutions $\mathbf{E}' \rightarrow \mathcal{P}'$ and $c\mathbf{B}' \rightarrow -\mathcal{M}'/c$.

Lorentz invariance allows us to write the Lagrangian density in the unprimed (laboratory) frame S exactly as in Eq. (5), or Eq. (2), simply by removing the primes. The total Lagrangian in the unprimed frame can now be written by adding the familiar relativistic Lagrangian contributions from the two particles as follows:

$$L = -m_1 c^2/\gamma(\dot{q}_1) - m_2 c^2/\gamma(\dot{q}_2) + \int d^3\mathbf{r}\left[\frac{\epsilon_0}{2}[E^2(\mathbf{r}) - c^2 B^2(\mathbf{r})] + \mathcal{P}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}) + \mathcal{M}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r})\right], \tag{6}$$

where $\gamma(\dot{q}) = (1 - \dot{q}^2/c^2)^{-1/2}$; the electric and magnetic fields are given by $\mathbf{E}(\mathbf{r}) = -\dot{\mathbf{A}}(\mathbf{r}) - \boldsymbol{\nabla}\phi(\mathbf{r})$ and $\mathbf{B}(\mathbf{r}) = \boldsymbol{\nabla} \times \mathbf{A}(\mathbf{r})$. It is important to bear in mind that the unprimed polarization and magnetization fields $\mathcal{P}(\mathbf{r})$ and $\mathcal{M}(\mathbf{r})$ appearing in Eq. (6) are *not* those in Eqs. (3) and (4). The primed polarization and magnetization fields are rest properties and the unprimed ones are related to them by relativistic connection rules involving a Lorentz transformation of the polarization field four-tensor $G_{\mu\nu}$ [22]. The interaction Lagrangian density [the last two terms in Eq. (6)] can thus be rewritten in terms of the primed polarization and magnetization by direct substitution as follows:

$$\mathcal{L}_{\text{int}} = \left\{\mathcal{P}'_{\|}(\mathbf{r}') + \gamma[\mathcal{P}'_{\perp}(\mathbf{r}') + \frac{1}{c^2}\dot{\mathbf{R}} \times \mathcal{M}'(\mathbf{r}')]\right\} \cdot \mathbf{E}(\mathbf{r}) + \{\mathcal{M}'_{\|}(\mathbf{r}') + \gamma[\mathcal{M}'_{\perp}(\mathbf{r}') - \dot{\mathbf{R}} \times \mathcal{P}'(\mathbf{r}')]\} \cdot \mathbf{B}(\mathbf{r}), \tag{7}$$

where the subscript $\|$ ($\perp$) denotes the vector projection parallel (perpendicular) to $\dot{\mathbf{R}}$ and $\gamma = \gamma(\dot{R})$. The term in Eq. (7) involving the velocity, electric polarization, and the magnetic $\mathbf{B}$ field is identified as the Röntgen Lagrangian interaction term [12–16], while the term involving the velocity, magnetization, and the electric field is identified as the Aharonov-Casher term [8,23].

Having expressed the interaction Lagrangian in terms of the known rest atomic properties $\mathcal{P}'$ and $\mathcal{M}'$, we now turn to the particle Lagrangian terms [given by the first two terms in Eq. (6)] and seek to express their sum in terms of the unprimed center of mass and relative velocities, $\dot{\mathbf{R}}$ and $\dot{\mathbf{q}}$, respectively, using the relations $\dot{\mathbf{q}}_1 = \dot{\mathbf{R}} + (m_2/M)\dot{\mathbf{q}}$ and $\dot{\mathbf{q}}_2 = \dot{\mathbf{R}} - (m_1/M)\dot{\mathbf{q}}$. Concentrating on the unprimed (S) frame, we now make use of the fact that the internal dynamics of the atom are not affected by relativistic considerations other than through $\dot{\mathbf{R}}$ (i.e., the motion of the electron around the nucleus is essentially nonrelativistic). We may then expand the sum of the particle Lagrangian terms up to terms quadratic in $\dot{\mathbf{q}}$ to obtain

$$-[m_1 c^2/\gamma(\dot{q}_1) + m_2 c^2/\gamma(\dot{q}_2)] = -Mc^2/\gamma + \tfrac{1}{2}\gamma\mu(\dot{q}_\perp^2 + \gamma^2 \dot{q}_\|^2), \tag{8}$$

where $\mu = m_1 m_2/M$ is the reduced mass. Note that in the equality of Eq. (8) the dependence on $\dot{R}$ is exact. In other words, all terms containing $\dot{R}$ have been retained. We now take the step of identifying the terms containing $\dot{q}$ as pertaining to the internal dynamics, in which context $\dot{R}$ has to be treated as a parameter.

After making use of Eqs. (7) and (8), the new Lagrangian emerging from Eq. (6) now becomes the starting point of the canonical procedure with $\mathbf{R}$ and $\mathbf{q}$ as the canonical variables for the atom and $\mathbf{A}(\mathbf{r})$ and $\phi(\mathbf{r})$ for the fields. The canonical momenta are $\mathbf{P}$ (conjugate to $\mathbf{R}$), $\mathbf{p}$ (conjugate to $\mathbf{q}$), and $\boldsymbol{\Pi}(\mathbf{r})$, which is identified as $-\mathbf{D}(\mathbf{r})$, the electric displacement field, is the momentum conjugate to $\mathbf{A}(\mathbf{r})$, while the momentum conjugate to ϕ is zero. Since we are concerned with the process of spontaneous emission by an atom characterized by an electric dipole moment, we may drop all interactions involving magnetic multipoles, but retain all those involving electric polarization, including the Röntgen term. The final Hamiltonian emerging from the canonical procedure can be written as a sum of three terms as follows:

$$H = H_a^0 + H_f^0 + H_{\text{int}}, \tag{9}$$

where

$$H_a^0 = \sqrt{M^2c^4 + P^2c^2} + \frac{1}{2\gamma\mu}\left(p_\perp^2 + \frac{p_\parallel^2}{\gamma^2}\right) + U(q), \quad (10)$$

$$H_f^0 = \int d^3\mathbf{r}\frac{\epsilon_0}{2}\left[\frac{1}{\epsilon_0^2}\Pi^2(\mathbf{r}) + c^2B^2(\mathbf{r})\right], \quad (11)$$

$$H_{\text{int}} = \int d^3\mathbf{r}\left\{\frac{1}{\epsilon_0}[\mathcal{P}'_\parallel(\mathbf{r}') + \gamma\mathcal{P}'_\perp(\mathbf{r}')]\cdot\mathbf{\Pi}(\mathbf{r}) + \frac{\mathbf{P}}{2M}\cdot[\mathcal{P}'(\mathbf{r}')\times\mathbf{B}(\mathbf{r})] + [\mathcal{P}'(\mathbf{r}')\times\mathbf{B}(\mathbf{r})]\cdot\frac{\mathbf{P}}{2M}\right\}. \quad (12)$$

The potential $U(q)$ in Eq. (10) is the interparticle Coulomb potential in the unprimed (laboratory) frame. In the primed frame (rest frame), the interparticle Coulomb potential, denoted as $U'(q')$, arises in the multipolar formulation from an integral term containing the square of the irrotational part $\mathcal{P}'^L$ of the polarization field, together with infinite Coulomb self-energies:

$$\frac{1}{2\epsilon_0}\int d^3\mathbf{r}'\{\mathcal{P}'^L\}^2 = U'(q') + \text{infinite Coulomb self-energies.} \quad (13)$$

On disregarding the infinite Coulomb self-energies, one then transforms the interparticle Coulomb energy $U'(q')$ to obtain $U(q)$, the interaction in the unprimed frame. The simplest and most direct route is by following the force transformation argument [24] to obtain

$$U(q) = \frac{U'(q')}{\gamma} = -\frac{e^2}{4\pi\epsilon_0\gamma q'}. \quad (14)$$

The expression for H_f^0 given in Eq. (11) is the familiar unperturbed field Hamiltonian which can be quantized following the standard methods of quantization for a free field in the laboratory frame. Finally, H_{int}, given in Eq. (12) is a new form of interaction Hamiltonian which couples the rest polarization field to electromagnetic fields in the laboratory frame.

The eigenfunctions of the atomic Hamiltonian H_a^0 are products of two parts: $\Psi(\mathbf{R}) = e^{i\mathbf{K}.\mathbf{R}}$, which satisfies the Klein-Gordon equation,

$$(-\hbar^2\nabla^2 + M^2c^2)\Psi(\mathbf{R}) = \left(\frac{E_K}{c}\right)^2\Psi(\mathbf{R}), \quad (15)$$

with $E_K = \sqrt{M^2c^4 + \hbar^2c^2K^2}$, and $\psi(\mathbf{q})$, which satisfies the Schrödinger equation,

$$\left[-\frac{\hbar^2}{2\gamma\mu}\left(\frac{\partial^2}{\partial q_x^2} + \frac{\partial^2}{\partial q_y^2} + \frac{1}{\gamma^2}\frac{\partial^2}{\partial q_z^2}\right) - \frac{e^2}{4\pi\epsilon_0\gamma q'}\right]\psi(\mathbf{q}) = \epsilon\psi(\mathbf{q}), \quad (16)$$

where, without loss of generality, we have taken the direction of the velocity $\dot{\mathbf{R}}$ to be the z direction. The total energy is given by $E = E_K + \epsilon \equiv \sqrt{M^2c^4 + \hbar^2c^2K^2} + \epsilon \equiv \gamma Mc^2 + \epsilon$ with $\hbar\mathbf{K} = \gamma M\dot{\mathbf{R}}$. Upon making the substitution $q_z' = \gamma q_z$ in Eq. (16), we obtain a Schrödinger equation governing the internal states of a hydrogenic atom in the atomic frame (rest frame) S' such that $\gamma\epsilon = \epsilon'$, where ϵ' are the internal eigenenergies in the rest frame S'.

We are now in a position to consider the energy and momentum conservation accompanying the process of spontaneous emission of a photon described in the unprimed (laboratory) frame as having wave vector $\mathbf{k}$ and frequency ω when the internal energy of the atom changes from ϵ_i to ϵ_f. Conservation of momentum requires that we have $\mathbf{K}_f = \mathbf{K}_i - \mathbf{k}$, where $\mathbf{K}_i = \gamma M\dot{\mathbf{R}}/\hbar$ is the initial center of mass wave vector, and $\mathbf{K}_f$ is the final wave vector in the laboratory frame. Conservation of energy, on the other hand, demands that we have

$$\omega = \frac{1}{\hbar}(\epsilon_i - \epsilon_f + E_{K_i} - E_{K_f}) \simeq \frac{1}{\hbar\gamma}(\epsilon_i' - \epsilon_f') + \frac{\hbar}{\gamma M}(\mathbf{K}_i\cdot\mathbf{k}) = \frac{\omega_0'}{\gamma} + \dot{\mathbf{R}}\cdot\mathbf{k}, \quad (17)$$

where $\hbar\omega_0'$ is the energy level difference in the primed (rest) frame S', and we have ignored the second order recoil energy. Note that Eq. (17) is equivalent to a Doppler shift in the photon frequency.

Two cases in the calculation of the spontaneous emission rate will have to be considered relative to the laboratory (unprimed) frame S, namely, (i) when the dipole moment vector is parallel to the velocity vector and (ii) when the dipole moment vector is perpendicular to the velocity vector. If these two calculations yield exactly the same result, then the spontaneous emission is deemed to be isotropic, i.e., free from angular dependence. Imposing the electric dipole approximation, $\mathcal{P}'(\mathbf{r}') = \mathbf{d}'\delta(\mathbf{r}' - \mathbf{R}')$, where $\mathbf{d}' = e\mathbf{q}'$, we obtain for the transition matrix element squared, with H_{int} as given in Eq. (12) as the interaction,

$$|ME|^2 = d'^2\left|\frac{\mathbf{E}_\parallel(\mathbf{R})}{\gamma} + \left[\mathbf{E}_\perp(\mathbf{R}) - \frac{1}{2}\left(2\dot{\mathbf{R}} - \frac{\hbar\mathbf{k}}{\gamma M}\right)\times\mathbf{B}(\mathbf{R})\right]\right|^2, \quad (18)$$

where only transverse (i.e., divergence-free) electromagnetic fields are involved and we have written $\mathbf{E}$ instead of $-\mathbf{\Pi}/\epsilon_0$, anticipating free-space quantization [25]. The photon momentum is much smaller than the particle momentum and may be ignored.

The free-space normalized electromagnetic fields can be obtained straightforwardly, remembering that we should identify two orthogonal wave polarizations. We choose the z direction, i.e., along $\dot{\mathbf{R}}$, as the axis along which there will be either a magnetic field (i.e., transverse electric or TE, $\xi = 1$) or an electric field (i.e., transverse magnetic or TM, $\xi = 2$). We can then write for a given wave vector $\mathbf{k} = (k, \theta, \phi)$ the following electric and magnetic fields in a normalization volume V:

$$\mathbf{E}_{\mathbf{k}}^{\mathrm{TM}}(\mathbf{r}) = \left(\frac{\hbar\omega}{2V\epsilon_0}\right)^{1/2}[\cos(\theta)\cos(\phi)\hat{\mathbf{x}} + \cos(\theta)\sin(\phi)\hat{\mathbf{y}} - \sin(\theta)\hat{\mathbf{z}}]e^{i(\mathbf{k}.\mathbf{r}-\omega t)} = c\mathbf{B}_{\mathbf{k}}^{\mathrm{TE}}(\mathbf{r}),$$

$$\mathbf{B}_{\mathbf{k}}^{\mathrm{TM}}(\mathbf{r}) = \left(\frac{\hbar\omega}{2V\epsilon_0 c^2}\right)^{1/2}[\sin(\phi)\hat{\mathbf{x}} - \cos(\phi)\hat{\mathbf{y}}]e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} = \frac{\mathbf{E}_{\mathbf{k}}^{\mathrm{TE}}(\mathbf{r})}{c}. \tag{19}$$

Turning finally to the Fermi golden rule formula in the unprimed (laboratory) frame S, we find that the spontaneous emission rate can be written as

$$\Gamma = \frac{2\pi}{\hbar^2}\sum_{\mathbf{k},\xi}|ME|^2\delta[\omega - \omega_0'/\gamma - \dot{R}k\cos(\theta)] = \frac{V}{4\pi^2\hbar^2}\sum_{\xi}\int_0^{\pi}d\theta\int_0^{2\pi}d\phi\,\frac{\omega_0'^3|ME|^2}{\gamma^3c^3[1-\dot{R}\cos(\theta)/c]^4}. \tag{20}$$

Substituting for the matrix element squared from Eq. (18) and the electric and magnetic fields from Eq. (19), we find that the spontaneous emission rate for a dipole parallel and perpendicular to the velocity in the laboratory frame S are given, respectively, by

$$\Gamma_{\parallel} = \Gamma_0'\int_{-1}^{1}dx\,\frac{3(1-x^2)}{4\gamma^5(1-\dot{R}x/c)^4} = \frac{\Gamma_0'}{\gamma}, \tag{21}$$

$$\Gamma_{\perp} = \Gamma_0'\int_{-1}^{1}dx\,\frac{3[(1+\dot{R}^2/c^2)(1+x^2)-4\dot{R}x/c]}{8\gamma^3(1-\dot{R}x/c)^4} = \frac{\Gamma_0'}{\gamma}, \tag{22}$$

where $\Gamma_0' = d'^2\omega_0'^3/(3\pi\epsilon_0\hbar c^3)$ is the free-space rate of spontaneous emission of the atom in the atomic rest frame S'. It is seen that there is no angular dependence, i.e., the rate of spontaneous emission is isotropic, and it does indeed vary like a relativistic clock.

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