Spurious velocity dependence of free-space spontaneous emission

Martin Wilkens
Optical Sciences Center, University of Arizona, Tucson, Arizona 85721
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It is shown that the standard calculation of the free-space spontaneous-emission rate leads to a spurious velocity dependence if the atomic motion is included nonrelativistically. The unphysical terms are of first order in v/c, akin to the nonrelativistic Doppler shift.

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Nonrelativistic quantum electrodynamics (QED) is the well-established framework which underlies quantum optics [1], cavity QED [2], atom optics [3], and laser cooling [4]. Traditionally, QED describes the dynamics of the electronic degrees of freedom of an atom and the electromagnetic field, but in more recent applications, the translational degrees of freedom of the atomic center-of-mass motion have been included to account for momentum conservation in light-matter interaction.

One of the most prominent textbook examples of QED is spontaneous emission, which results from the inevitable interaction of the atom with the quantized electromagnetic field vacuum. It has recently attracted renewed attention in the context of laser cooling and atom optics, where an atomic master equation has been derived from first-principle calculations, starting from a nonrelativistic Hamiltonian model which also incorporates the atomic translational degrees of freedom [5, 6]. The atomic master equation is obtained by adiabatically eliminating the fluorescence modes in a Born-Markoff approximation. In the final equations, the impact of these modes is manifest through directionally dependent transition rates which account for electronic transitions as well as transitions between different momentum states of the atom.

In this article we demonstrate that these rates acquire velocity dependencies which violate the Galilean invariance principle. This should come as no surprise, since nonrelativistic QED couples two radically different symmetry groups: the Galilei group which governs the center-of-mass motion of the atom, and the Lorentz group which governs the evolution of the electromagnetic field. What is somewhat disturbing, however, is that the unphysical terms are formally of the same order as the trustworthy linear Doppler shift.

It appears that this particular flaw of nonrelativistic QED has been left unnoticed so far, possibly due to the relative smallness of the spurious terms and their intimate connection to the Doppler effect. In fact, in most of the published derivations, the atomic-rate equations are simplified at a final stage and as a byproduct, the unphysical velocity dependencies disappears [5]. However, in at least one such derivation [6], the unphysical modifications are taken seriously to some extent, although no experimental significance seems to have been attached to them so far. It is quite clear, however, that the simultaneous appearance of artificial v/c terms and of important v/c corrections invites for confusion, in particular in more complicated situations, where our *a priori* knowledge of what is physical, and what is not, may be limited.

We proceed by first evoking a standard Wigner-Weisskopf approximation to rederive the rate of spontaneous emission of an excited two-level atom which travels freely in the electromagnetic vacuum. We then show that the unphysical corrections to the partial rates associated with emission in a given direction are of first order in the atomic velocity, but the explicit form of the corrections depends on whether the calculations are carried out in the dE scheme or in the pA scheme.

In nonrelativistic QED, the interaction of a two-level atom and the electromagnetic field, taking into account the atomic motion, is described by the standard Hamiltonian [7]

$$H = H_A + H_F + H_{A\text{-}F}, \tag{1}$$

which is a sum of the atomic Hamiltonian

$$H_A = \frac{\hat{\mathbf{p}}^2}{2M} + \hbar\omega_A \sigma_+ \sigma_-, \tag{2}$$

the free-field Hamiltonian

$$H_F = \sum_j \hbar\omega_j a_j^\dagger a_j, \tag{3}$$

and the atom-field interaction in the dipole and rotating-wave approximation

$$H_{A\text{-}F} = -\wp \cdot \mathbf{E}^+(\hat{\mathbf{x}})\sigma_+ + \text{H.c.} \tag{4}$$

Here $\hat{\mathbf{p}}$ and $\hat{\mathbf{x}}$ are the atomic center-of-mass momentum and position operators, whose components obey canonical commutation relations, $\sigma_\pm$ denote the atomic level raising and lowering operators where $\{\sigma_+, \sigma_-\} = 1$ with $\sigma_\pm^2 = 0$, a_j and $a_j^\dagger$ denote the photon annihilation and creation operators of the jth mode which obey $[a_i, a_j^\dagger] = \delta_{ij}$, $\mathbf{E}^+(\mathbf{x})$ is the positive-frequency part of the electrodynamic field operator, ω_A is the Bohr transition frequency, and $\wp$ is the dipole transition matrix element.

We consider a $\Delta m = 0$ transition in free space. In this case it is convenient to expand the electric field in terms of linearly polarized plane waves with periodic boundary conditions

$$\mathbf{E}^+(\mathbf{x}) = i\sum_{j,\lambda}\sqrt{\frac{\hbar\omega_j}{2\epsilon_0 V}}\mathbf{e}_{j\lambda}e^{i\mathbf{k}_j\cdot\mathbf{x}}a_{j\lambda}, \quad (5)$$

where $\mathbf{k}_j = 2\pi\mathbf{j}/L$, $\mathbf{j} = (j_x, j_y, j_z)$ is a vector of integers, $\lambda = 1, 2$ enumerates the polarization directions, and $L^3 \equiv V$ is the quantization volume. The transversality of the mode functions is guaranteed for mutually orthogonal propagation and polarization vectors. This determines the polarizations up to an arbitrary rotation around the direction of propagation $\mathbf{n}_j \equiv c\mathbf{k}_j/\omega_j$. We may use this freedom to choose $\mathbf{e}_{j2}$ orthogonal to the atomic dipole transition vector $\wp$. The scalar product (4) of the remaining polarization vector $\mathbf{e}_{j1}$ and the transition dipole element may then be expressed in terms of the angle θ_j between the wave vector $\mathbf{k}_j$ and $\wp$, i.e., $\wp\mathbf{e}_{1j} = \wp\sin\theta_j$. With these conventions, the atom-field interaction assumes the form

$$H_{A\text{-}F} = -i\sum_j g_j\left[e^{i\mathbf{k}_j\cdot\hat{\mathbf{x}}}\sigma_+ a_j - a_j^\dagger\sigma_- e^{-i\mathbf{k}_j\cdot\hat{\mathbf{x}}}\right], \quad (6)$$

with a coupling constant

$$g_j = \wp\sqrt{\frac{\hbar\omega_j}{2\epsilon_0 V}}\sin\theta_j. \quad (7)$$

Here, the momentum-shift operator $e^{-i\mathbf{k}_j\cdot\hat{\mathbf{x}}}$ accounts for momentum conservation in the atom-field interaction.

To derive the rate of spontaneous emssion we consider an initially excited atom which travels with momentum $\mathbf{p}$ through the electromagnetic vacuum. Due to the interaction (6), this atom will eventually end up in its ground state, traveling with some momentum $\mathbf{p} - \hbar\mathbf{k}_j$, where $\mathbf{k}_j$ is the wave vector of the emitted photon. Summing the corresponding rates over all possible final states where the photon is emitted in a given direction $\mathbf{n}$, one obtains to lowest order in the atom-field interaction for the rate of spontaneous emission in this direction (Fermi's golden rule)

$$d\gamma_{\mathbf{v}}(\mathbf{n}) = \frac{2\pi}{\hbar}\sum_j |g_j|^2\delta\left\{\hbar\omega_j + \frac{(\mathbf{p}-\hbar\mathbf{k}_j)^2}{2M} - \left[\hbar\omega_A + \frac{\mathbf{p}^2}{2M}\right]\right\}\delta(\mathbf{n}-\mathbf{n}_j), \quad (8)$$

where the argument of the first δ-distribution accounts for energy conservation.

Expanding this argument, we observe that the only contributions to the sum (8) stem from frequencies which obey

$$\omega_j = \omega_A + \omega_j\frac{\mathbf{n}_j\cdot\mathbf{v}}{c} - \frac{\hbar\omega_j^2}{2Mc^2}. \quad (9)$$

Here, the second term on the right-hand side accounts for the nonrelativistic Doppler shift and the third term accounts for the nonrelativistic recoil shift.

To proceed, we evaluate the sum (8) in the continuum limit $\sum_j \to \frac{V}{(2\pi c)^3}\int\omega^2 d\omega\, d^2\mathbf{n}'$, where $d^2\mathbf{n}'$ is the infinitesimal solid angle in direction $\mathbf{n}'$ of spontaneous emission. We also express the $\sin^2\theta_j$ factor of $|g_j|^2$ in terms of the dipole radiation pattern

$$\Phi(\mathbf{n}) = \frac{3}{8\pi}\left[1 - \frac{\wp\cdot\mathbf{n}}{\wp^2}\right], \quad (10)$$

which is normalized to $\int d^2\mathbf{n}\,\Phi(\mathbf{n}) = 1$. Finally, we assume that the atom is infinitely heavy, $M \to \infty$, which allows us to drop the recoil term in Eq. (9). With these conventions we obtain for the differential rate of spontaneous emission in the $\mathbf{n}$ direction

$$d\gamma_{\mathbf{v}}(\mathbf{n}) = \frac{1}{4\pi\epsilon_0}\frac{4\wp^2}{3\hbar c^3} \times\left\{\int_0^\infty d\omega\,\omega^3\delta\left[\omega - \omega_A - \omega\frac{\mathbf{n}\cdot\mathbf{v}}{c}\right]\right\}\Phi(\mathbf{n})d^2\mathbf{n}. \quad (11)$$

The remaining integration is easily performed and the result is

$$d\gamma_{\mathbf{v}}(\mathbf{n}) = \gamma_0\left[1 - \frac{\mathbf{n}\cdot\mathbf{v}}{c}\right]^{-4}\Phi(\mathbf{n})d^2\mathbf{n}, \quad (12)$$

where

$$\gamma_0 = \frac{1}{4\pi\epsilon_0}\frac{4\wp^2\omega_A^3}{3\hbar c^3} \quad (13)$$

is the rate of spontaneous emission for an atom at rest.

At this stage it is worthwhile to recall that in a Galilean theory, the differential rate of spontaneous emission in direction $\mathbf{n}$ can only depend on the direction of the dipole transition vector $\wp$ or other spatial directions related to the internal structure of the atom, but not on the center-of-mass velocity $\mathbf{v}$. The simple reason behind this is that time increments (i.e., rates) and directions (i.e., the dipole pattern) are invariant under Galilean boosts. [In a relativistic theory, in contrast, rates like γ transform like the inverse proper time, $\gamma_v = \gamma_0\sqrt{1 - v^2/c^2}$].

The calculated differential rate (12) violates Galilean invariance (as well as relativistic transformation properties). Even worse, the "correction" to that rate is of relative order v/c, which may be interpreted as a nonrelativistic effect akin to the non-relativistic Doppler effect. This interpretation is clearly unacceptable, and the v/c dependence of $d\gamma$ must be rejected.

Sometimes this is overlooked. The reason may be that the Doppler shift in Eq. (11) rests certainly on solid ground: the frequency picked by the δ function is nothing but the atomic transition frequency of the moving atom as measured in the laboratory frame. But the product of the transition element ($\sim\omega$) and the mode density ($\sim\omega^2$), and the trustworthy Doppler effect somehow conspire to produce the physically unacceptable result of Eq. (12). The formal reason is that the integrand in Eq. (11) reflects neither Galilei invariance nor the proper transformation properties of a fully relativistic theory, as the Hamiltonian (1) couples the Galelei group of the atomic motion with the Lorentz group of the field evolution.

It is also interesting to note that the degree of the misleading velocity dependence in Eq. (12) depends on whether the calculations are carried out in the dE or in the $p{\times}A$ scheme. Using the pA scheme, the transition matrix element varies $\sim \omega^{-1}$ and the power in Eq. (12) becomes -2 instead of -4. This is yet another indication that the velocity dependence of $d\gamma$ is unphysical and must be rejected.

In conclusion, we have demonstrated that the standard calculation of spontaneous-emission rates in the framework of nonrelativistic QED leads to a spurious v/c dependence. In contrast to the v/c dependence of the linear Doppler shift, which results from the proper invariance of spatio-temporal phases under Galilean boosts, the v/c dependence of the spontaneous-emission rate has to be rejected *a posteriori* because it violates Galilean invariance properties. This indicates that first-principle calculations in nonrelativistic QED are at risk of producing unphysical results already to order v/c if atomic motion is included. In particular, in more complicated situations, such as in the presence of external laser fields or inside cavities, the nature of velocity dependence of the atomic response may be opaque and not easily traced back to either a physical or an artificial source. It is then preferable—and indeed common practice—to use a master equation, where spontaneous emission is already included in a Galilean invariant form, rather than to rely on first-principles calculations. This eliminates at least one source of artificial velocity dependencies in the calculation of the atomic response.

Of course, the ultimate cure of this deficiency may be expected from a relativistically covariant description of the atomic motion, both internal and external, and the interaction with the electromagnetic field. Despite its innocent appearance, this program is, however, highly nontrivial [8]. If taken literally, it requires us to set up a covariant QED formulation of a (at least) two-body bound-state problem (light electron plus the rest of the atom). Alternatively, one could try to set up a covariant description of an effective two-level atom, thereby avoiding the notorious bound-state problem [9]. This program is perhaps more promising, but it is still in a very early stage. It also faces difficulties (the appearance of "antiatoms," for example) which are somewhat annoying considering the simplicity of its nonrelativistic counterpart. And finally, it might be a little bit out of proportion to set up such a program for just the purpose of deciding the existence or nonexistence of certain v/c corrections in nonrelativistic quantum optics. But then again, the program is certainly worthwhile pursuing, in particular if one plans to found the emerging field of atom optics on a basis more solid than the current standard model.

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