## Harmonic generation: Quantum-electrodynamical theory of the harmonic photon-number spectrum

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Simulations of harmonic generation in the single-atom limit usually describe the harmonic radiation as proportional to the squared magnitude of the Fourier transform (FT) of the expectation value of either the dipole moment or the dipole acceleration. However, no derivation of an explicit single-atom representation of the typical experimental measurement (i.e., the number of harmonic photons produced versus the frequency of the harmonic photon-number spectrum" is derived, which is proportional to the double FT of the time-autocorrelation function of the dipole velocity of the whole system. Within the long-wavelength approximation the time-correlation function can be recast as the FT of the squared magnitude of the expectation value of the dipole velocity of a single representative atom.

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The phenomenon of harmonic generation (HG), in which low-frequency electromagnetic radiation is converted, through the interaction of a laser pulse with a material system, to higher frequencies that are integer multiples (harmonics) of the low frequency, has been under investigation since the mid 1960s. L'Huillier et al. [1] lucidly describe the experimental measurement of HG in gases. An intense laser pulse impinges on a quantity of gas synchronously released at the focus of the laser through a pulse jet. The radiation emitted in the forward direction is dispersed on a grazing incidence grating and the separate frequencies are detected by photomultipliers. The number of photons produced at each frequency can be estimated within an order of magnitude. For our purpose the essential result of the experiment is a plot of the number of photons produced at frequency  $\omega$ (along the axis of propagation of the laser pulse) against  $\omega$ . To be precise, we reserve the rather awkward term "harmonic photon-number spectrum" (HPNS) for such a plot.

The theoretical treatment of HG is enormously complicated by the necessity of describing the evolution of a compound multidimensional system comprising charged particles coupled to modes of the radiation field. Brabec and Krausz [2] outline two basic, complementary approaches. On the one hand, one can describe the system at the macroscopic scale by means of Maxwell's equations, which account for the coupling of multiple processes (e.g., ionization, HG, absorption, propagation, and phase matching) that have a profound influence on (currently feasible) experimental measurements. On the other hand, in the hypothetical experiment in which the density of the gas approaches zero one can describe the system at the microscopic scale as a single atom interacting with an external field associated with the laser pulse. As Brabec and Krausz remark, although a detailed comparison of theory with experiment calls for a macroscopic theory including propagation effects, many important features of experiment can be understood on the basis of the

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Pioneers in the single-atom simulation of HG described the harmonic radiation in terms of the expectation value of the component of the dipole moment along the direction of polarization (e) of the laser pulse [i.e.,  $\langle \mathbf{d}(t) \cdot \mathbf{e} \rangle$  $\equiv \langle \Psi(t) | \mathbf{d} \cdot \mathbf{e} | \Psi(t) \rangle$ , where  $\mathbf{d} \equiv e\mathbf{r}$ ,  $\mathbf{r}$  is the position of the electron relative to the nucleus, and *e* is its charge  $\begin{bmatrix} 3-5 \end{bmatrix}$ . Kulander and Shore [3] state that "the square of the Fourier transform (FT) of  $\langle d(t) \rangle [\langle \mathbf{d}(t) \cdot \mathbf{e} \rangle]$  gives the distribution of frequencies radiated by the atom and so a plot of this quantity displays the fluorescence spectrum." In their paper on HG in a one-dimensional model, Eberly et al. [4] say "the coherently scattered light power is...proportional to the square of  $D(\omega)$ ," where  $D(\omega)$  is the FT of  $\langle \mathbf{d}(t) \cdot \mathbf{e} \rangle$ . The purely classical study of HG in the hydrogen atom by Bandarage *et al.* [5] took "the spectrum of light emitted by the forced dipole" to be "straightforwardly obtained from its power spectrum:  $D(\omega) = \lim_{T \to \infty} T^{-1} |\int_0^T \mu_z(t) e^{i\omega t} dt|^2$ ."

Sundaram and Milonni [6] stated that the spectrum actually should be proportional to the (double) FT of the timeautocorrelation function of the dipole moment. They further argued that the use of the expectation value, instead of the time-correlation function, may be justified in case the atoms in the region of interaction between the laser pulse and the gas (interaction region) are "uncorrelated." They also remarked in passing that the total power radiated by the driven atom is proportional to the expectation value of the square of the dipole acceleration. Focusing on this comment, Burnett et al. [7] questioned the use of the dipole moment in calculating the spectrum. Based on a comparison of spectra computed from expectation values of the dipole moment and the dipole acceleration, they recommended that the "acceleration form" be used. Since that time some researchers have followed their advice [8-13], while others have adhered to the dipole-moment form [14–17]. Comparisons of spectra based on both dipole-moment and dipole-acceleration forms [12,18], as well as on dipole-moment, dipole-velocity, and dipole-acceleration forms [19], have been reported. To the author's knowledge, no derivation of an explicit single-atom

representation of the harmonic photon-number spectrum has appeared, despite the large literature devoted to the singleatom simulation of HG.

The principal purpose of this article is to present a derivation of the formula for the HPNS. It begins with the Hamiltonian

$$H(t) = \sum_{\alpha} \frac{1}{2m_{\alpha}} [\mathbf{p}_{\alpha} - q_{\alpha} \mathbf{A}(\mathbf{r}_{\alpha}) - q_{\alpha} \mathbf{A}_{\text{ext}}(\mathbf{r}_{\alpha}, t)]^{2} + V_{c}$$
$$+ \sum_{i} \hbar \omega_{i} (a_{i}^{\dagger} a_{i} + 1/2)$$
(1)

that governs the interaction of the laser pulse with the material system and which is derivable from the standard Lagrangian in the Coulomb (velocity) gauge [20]. (We adhere to the notation of Ref. [20].) In Eq. (1)  $\alpha$  labels particles of mass  $m_{\alpha}$ , charge  $q_{\alpha}$ , position  $\mathbf{r}_{\alpha}$ , and canonical momentum  $\mathbf{p}_{\alpha}$ ; *i* labels discrete box-normalized plane-wave (normal) modes of the radiation field, which are characterized by propagation vector  $\mathbf{k}_i$ , frequency  $\omega_i = ck_i$  (where *c* is the speed of light) and polarization unit vector  $\mathbf{e}_i$ ;  $a_i^{\dagger}$  and  $a_i$ , respectively, represent the creation and annihilation operators for the (imaginary) harmonic oscillator associated with mode *i*.  $\mathbf{A}(\mathbf{r}_{\alpha})$ , the vector potential at the position of particle  $\alpha$  due to the motion of the particles, can be written as

$$\mathbf{A}(\mathbf{r}_{\alpha}) = \sum_{i} \left( \hbar/2\varepsilon_{0}\omega_{i}L^{3} \right)^{1/2} [a_{i}e^{i\mathbf{k}_{i}\cdot\mathbf{r}_{\alpha}} + a_{i}^{\dagger}e^{-i\mathbf{k}_{i}\cdot\mathbf{r}_{\alpha}}]\mathbf{e}_{i}, \quad (2)$$

where  $\hbar$  is the modified Planck constant,  $\varepsilon_0$  is the electric permittivity of the vacuum, and  $L^3$  is the normalization volume;  $\mathbf{A}_{\text{ext}}(\mathbf{r}, t)$  is the prescribed (external) vector potential characterizing the laser pulse. (We assume that  $\mathbf{A}_{\text{ext}}$  has a finite duration and vanishes before an initial instant, which we take to be t=0 for notational convenience. We also assume that the scalar potential of the external field is zero.) In Eq. (1)  $V_c$  stands for the Coulombic potential energy, given explicitly by

$$V_{\rm c} = \frac{1}{4\pi\varepsilon_0} \sum_{\alpha} \sum_{\beta > \alpha} \frac{q_{\alpha}q_{\beta}}{|\mathbf{r}_{\alpha} - \mathbf{r}_{\beta}|}.$$
 (3)

The commutators associated with the canonical variables of the particles and modes of the field assume the usual form

$$[r_{\alpha i}, p_{\beta j}] = i\hbar \,\delta_{\alpha\beta}\delta_{ij}, \quad [a_i, a_j^{\dagger}] = \delta_{ij}, \tag{4}$$

where the indices i and j in the first of Eqs. (4) refer to Cartesian components.

The theoretical quantity that we wish to calculate in terms of the microscopic properties of the system, and which we suppose corresponds to the "harmonic photon-number spectrum," is the expected (mean) population of photons in a field mode as a function of the variables (in particular,  $\omega_i$ ) that specify the mode. Thus, the mean population of mode *i* can be expressed as

$$\bar{n}_i(t) = \langle n_i(t) \rangle = \operatorname{Tr}\{\rho(0)n_i(t)\},\tag{5}$$

where  $\rho(0)$  is the initial density operator and  $n_i(t)$  is the operator in Heisenberg's picture corresponding to the occupation number of mode *i*, which is

$$n_i = a_i^{\dagger} a_i \tag{6}$$

in Schrödinger's picture at the initial instant (t=0). We assume that  $\rho(0)$  can be expressed as a direct product of density operators  $\rho_{\text{field}}(0)$  and  $\rho_{\text{part}}(0)$  specifying the initial, generally mixed, states of the field and the particles.

Rather than compute  $n_i(t)$  directly, we first compute  $a_i(t)$  and then multiply that by the Hermitian adjoint. Hence, using Eqs. (1)–(4), we obtain Heisenberg's equation of motion for  $a_i(t)$ ,

$$\dot{a}_{i}(t) = (i\hbar)^{-1} [a_{i}, H(t)] = -i\omega_{i}a_{i}(t) + i(2\varepsilon_{0}\hbar\omega_{i}L^{3})^{-1/2}\Delta_{i}(t),$$
(7)

where

$$\Delta_i(t) \equiv \frac{1}{2} \sum_{\alpha} q_{\alpha} [\dot{\mathbf{r}}_{\alpha}(t) e^{-i\mathbf{k}_i \cdot \mathbf{r}_{\alpha}(t)} + e^{-i\mathbf{k}_i \cdot \mathbf{r}_{\alpha}(t)} \dot{\mathbf{r}}_{\alpha}(t)] \cdot \mathbf{e}_i.$$
(8)

The positions of the particles obey their own Heisenberg equations of motion [20]

$$m_{\alpha}\ddot{\mathbf{r}}_{\alpha}(t) = q_{\alpha}\mathbf{E}_{\text{tot}}(\mathbf{r}_{\alpha}, t) + \frac{q_{\alpha}}{2}[\dot{\mathbf{r}}_{\alpha}(t)$$
$$\times \mathbf{B}_{\text{tot}}(\mathbf{r}_{\alpha}, t) - \mathbf{B}_{\text{tot}}(\mathbf{r}_{\alpha}, t) \times \dot{\mathbf{r}}_{\alpha}(t)]. \tag{9}$$

In Eq. (9)  $\mathbf{E}_{tot} = \mathbf{E} + \mathbf{E}_{ext}$  and  $\mathbf{B}_{tot} = \mathbf{B} + \mathbf{B}_{ext}$ , where  $\mathbf{E}$  and  $\mathbf{B}$  are the electric and magnetic fields due to the motion of the particles and  $\mathbf{E}_{ext}$  and  $\mathbf{B}_{ext}$  are the corresponding imposed external fields due to the laser pulse. The formal solution of Eq. (7) is

$$a_{i}(t) = e^{-i\omega_{i}t}a_{i} + i(2\hbar\omega_{i}\varepsilon_{0}L^{3})^{-1/2} \int_{0}^{t} dt' e^{-i\omega_{i}(t-t')}\Delta_{i}(t').$$
(10)

From Eqs. (5), (6), and (10) we deduce

$$\begin{split} \bar{n}_{i}(t) &= \langle a_{i}^{\dagger}a_{i} \rangle + i(2\hbar\omega_{i}\varepsilon_{0}L^{3})^{-1/2} \int_{0}^{t} dt' e^{i\omega_{i}t'} \langle a_{i}^{\dagger}\Delta_{i}(t') \rangle \\ &- i(2\hbar\omega_{i}\varepsilon_{0}L^{3})^{-1/2} \int_{0}^{t} dt'' e^{-i\omega_{i}t''} \langle \Delta_{i}^{\dagger}(t'')a_{i} \rangle \\ &+ (2\hbar\omega_{i}\varepsilon_{0}L^{3})^{-1} \int_{0}^{t} dt' \int_{0}^{t} dt'' e^{-i\omega_{i}(t'-t'')} \langle \Delta_{i}^{\dagger}(t')\Delta_{i}(t'') \rangle. \end{split}$$

$$(11)$$

If the field modes are assumed to be unpopulated initially, then  $\rho_{\text{field}}(0) = |\mathbf{0}\rangle \langle \mathbf{0}|$ , where  $|\mathbf{0}\rangle = \Pi_k |\mathbf{0}_k\rangle$  stands for the initial state vector of the field. Using the relation  $a_i |\mathbf{0}\rangle = 0$ , we simplify Eq. (11) to

$$\bar{n}_i(t) = (2\hbar\omega_i\varepsilon_0 L^3)^{-1} \int_0^t dt' \int_0^t dt'' e^{-i\omega_i(t'-t'')} \langle \Delta_i^{\dagger}(t')\Delta_i(t'') \rangle.$$
(12)

Although Eq. (12) gives the exact formula for the sought quantity, it is practically impossible to compute, since in order to do so one would need to solve the coupled Heisenberg equations of motion (7) and (9) in the infinite-dimensional

representation based on the direct product of complete sets of field-mode and particle states.

From Eq. (9) it is clear that the particles are driven by the external field and from Eqs. (7) and (8) that the moving particles drive the field modes. In turn, the field modes, which are implicit in  $\mathbf{E}[\mathbf{r}_{\alpha}(t), \{a_j(t)\}]$  and  $\mathbf{B}[\mathbf{r}_{\alpha}(t), \{a_j(t)\}]$  and therefore contribute to  $\mathbf{E}_{\text{tot}}$  and  $\mathbf{B}_{\text{tot}}$ , react to drive the particles. If this "back-reaction" of the field modes on the particles is neglected, the motion of the particles uncouples from that of the field modes. Equation (9) then reduces to

$$m_{\alpha}\ddot{\mathbf{r}}_{\alpha}(t) = -\nabla_{\mathbf{r}_{\alpha}}V_{c} + q_{\alpha}\mathbf{E}_{ext}(\mathbf{r}_{\alpha}, t) + \frac{q_{\alpha}}{2}[\dot{\mathbf{r}}_{\alpha}(t) \\ \times \mathbf{B}_{ext}(\mathbf{r}_{\alpha}, t) - \mathbf{B}_{ext}(\mathbf{r}_{\alpha}, t) \times \dot{\mathbf{r}}_{\alpha}(t)], \quad (13)$$

where now the motion of the particles is determined solely by the external field. [Note that the Coulombic force (the first term on the right side of Eq. (13)) is equivalent to the electric force on particle  $\alpha$  due to the longitudinal component of the electric field, which is implicit in  $\mathbf{E}_{tot}$  in Eq. (9).] Now one needs to solve "only" the Heisenberg equations [Eq. (13)] for the particles and then perform the trace implicit in Eq. (12) over "only" particle states. Although the computational problem has been simplified by "an order of infinity," it nevertheless remains formidable, particularly since the particles are subject to different external fields effectively, because of spatial variations in the field associated with the prescribed laser pulse.

To simplify the description even further, one commonly resorts to the "long-wavelength approximation" [21], assuming that the wavelengths associated with the external (laser) field, as well as with the relevant modes of the dynamically generated field, are long compared with the dimensions of the interaction region. In this limit A and  $A_{ext}$  become independent of position and Eq. (13) reduces further to

$$m_{\alpha}\ddot{\mathbf{r}}_{\alpha}(t) = -\nabla_{\mathbf{r}_{\alpha}}V_{c} + q_{\alpha}\mathbf{E}_{\text{ext}}(\mathbf{0}, t), \qquad (14)$$

where the system is taken to be located near the origin. [Note that  $\mathbf{B}_{\text{ext}} = \nabla \times \mathbf{A}_{\text{ext}}(\mathbf{0}, t)$  vanishes.] Likewise, the expression for  $\Delta_i(t)$  given by Eq. (8) becomes

$$\Delta_i(t) = \sum_{\alpha} q_{\alpha} \dot{\mathbf{r}}_{\alpha}(t) \cdot \mathbf{e}_i \equiv \dot{\mathbf{D}} \cdot \mathbf{e}_i, \qquad (15)$$

where **D** is the dipole moment of the system. Then from Eq. (12) we obtain a simplified expression for the population of photons in field mode *i*:

$$\bar{n}_{i}(t) = (2\hbar\omega_{i}\varepsilon_{0}L^{3})^{-1} \int_{0}^{t} dt' \int_{0}^{t} dt'' e^{-i\omega_{i}(t'-t'')}$$
$$\times \langle \dot{\mathbf{D}}(t') \cdot \mathbf{e}_{i} \dot{\mathbf{D}}(t'') \cdot \mathbf{e}_{i} \rangle.$$
(16)

Hence, the HPNS is seen to be proportional to a double FT of the time-autocorrelation function of the dipole velocity. We note that Eq. (16) corresponds to Eq. (7) of Sundaram and Milonni [6]. The essential difference, however, is that Eq. (16) here involves the dipole velocity whereas Eq. (7) of Ref. [6] involves the dipole moment itself.

The formula for  $\overline{n}_i(t)$  in Eq. (16) can be further simplified, if we assume the material system to consist of a pure dilute gas. The dipole moment of the whole system can then be recast as a sum of moments of atoms

$$\mathbf{D} = \sum_{a}^{N} \mathbf{d}_{a},\tag{17}$$

where the index a labels atoms, of which there are supposed to be N. We can then rewrite the time-correlation function in Eq. (16) as

$$C_{i}(t',t'') \equiv \langle \mathbf{D}(t') \cdot \mathbf{e}_{i} \mathbf{D}(t'') \cdot \mathbf{e}_{i} \rangle$$
  
=  $\sum_{a}^{N} \operatorname{Tr} \{ \rho_{\text{part}}(0) \dot{\mathbf{d}}_{a}(t') \cdot \mathbf{e}_{i} \dot{\mathbf{d}}_{a}(t'') \cdot \mathbf{e}_{i} \}$   
+  $\sum_{a}^{N} \sum_{b \neq a}^{N} \operatorname{Tr} \{ \rho_{\text{part}}(0) \dot{\mathbf{d}}_{a}(t') \cdot \mathbf{e}_{i} \dot{\mathbf{d}}_{b}(t'') \cdot \mathbf{e}_{i} \}.$  (18)

Since the gas atoms are coupled only through weak, shortrange (e.g., van der Waals) forces, we can neglect their interactions with one another. In this approximation we can express the initial density operator of the system as a direct product of the initial density operators of the presumed independent atoms as

$$\rho_{\text{part}}(0) = \prod_{c} \rho_c(0). \tag{19}$$

Substitution of Eq. (19) into Eq. (18) yields

$$C_{i}(t',t'') = \sum_{a}^{N} \operatorname{Tr}\{\rho_{a}(0)\dot{\mathbf{d}}_{a}(t') \cdot \mathbf{e}_{i}\dot{\mathbf{d}}_{a}(t'') \cdot \mathbf{e}_{i}\} + \sum_{a}^{N} \sum_{b \neq a}^{N} \operatorname{Tr}\{\rho_{a}(0)\dot{\mathbf{d}}_{a}(t') \cdot \mathbf{e}_{i}\}\operatorname{Tr}\{\rho_{b}(0)\dot{\mathbf{d}}_{b}(t'') \cdot \mathbf{e}_{i}\}.$$
(20)

Now within the long-wavelength approximation every atom is subject to the same external field at the same instant and every atom is characterized initially by the same density operator. Consequently, we can rewrite Eq. (20) as

$$C_{i}(t',t'') = N \operatorname{Tr}\{\rho(0)\dot{\mathbf{d}}(t') \cdot \mathbf{e}_{i}\dot{\mathbf{d}}(t'') \cdot \mathbf{e}_{i}\}$$
$$+ N(N-1)\operatorname{Tr}\{\rho(0)\dot{\mathbf{d}}(t') \cdot \mathbf{e}_{i}\}\operatorname{Tr}\{\rho(0)\dot{\mathbf{d}}(t'') \cdot \mathbf{e}_{i}\},$$
(21)

where  $\rho(0)$  and **d** now refer to a single representative atom in the interaction region. Since  $N \ge 1$ , we can neglect the first term on the right side of Eq. (21) and rewrite Eq. (16) approximately as

$$\overline{n}_{i}(t) = \frac{N^{2}}{2\hbar\omega_{i}\varepsilon_{0}L^{3}} \left| \int_{0}^{t} dt' e^{-i\omega_{i}t'} \operatorname{Tr}\{\rho(0)\dot{\mathbf{d}}(t') \cdot \mathbf{e}_{i}\} \right|^{2}.$$
(22)

We conclude that, under the customary assumptions, the "harmonic photon-number spectrum" is proportional to the squared magnitude of the Fourier transform of the expectation value of the dipole velocity of the representative atom.

Now  $\bar{n}_i(t)$  is the (expected) number of photons produced at time *t* in the single, discrete mode *i*, which is specified by wave vector  $\mathbf{k}_i [=(k_i, \theta_i, \phi_i)$ , in spherical polar coordinates] and polarization unit vector  $\mathbf{e}_i$ . As  $L \to \infty$ , the modes become continuously distributed. The number of modes with frequency in the range  $d\omega$  about  $\omega$ , direction in the element of solid angle  $d\Omega$ , and fixed polarization  $\mathbf{e}$  is then [22]

$$dN_m = (L/2\pi c)^3 \omega^2 d\omega d\Omega.$$
<sup>(23)</sup>

Therefore, the total number of photons produced at time t with these specifications (i.e., with frequency in the range  $d\omega$  about  $\omega$ , direction in the element of solid angle  $d\Omega$ , and fixed polarization **e**), which is the theoretical quantity that corresponds to the hypothetical single-atom HPNS that could in principle be measured, is given as the product of  $\bar{n}_i(t)$  and  $dN_m$ :

$$dn_{\rm tot}(t) = \frac{N^2 \omega}{2\hbar\varepsilon_0 (2\pi c)^3} \left| \int_0^t dt' e^{-i\omega t'} {\rm Tr}\{\rho(0)\dot{\mathbf{d}}(t') \cdot \mathbf{e}\} \right|^2 d\omega d\Omega.$$
(24)

We note in passing that the quadratic dependence of the HPNS on the number of gas atoms in the interaction region (or, equivalently, the density of the gas) was observed in early experiments on HG by Li *et al.* [23]. It is also in agreement with recent findings of Lorin *et al.* [24], who solved the coupled Maxwell-Schrödinger equations for a one-dimensional model of  $H_2^+$  gas.

The argument t of  $dn_{tot}(t)$  or  $\overline{n}_i(t)$  serves as a reminder that the hypothetical HPNS depends on the length of the

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period during which photons are collected. For real measurements [1] the period of collection is essentially infinite compared with the period (t) over which  $\text{Tr}\{\rho(0)\dot{\mathbf{d}}(t')\cdot\mathbf{e}\}$  can be reliably computed, even for the simplest atom. Optimum agreement between hypothetical and real measurements of the HPNS would therefore be expected to occur when  $\text{Tr}\{\rho(0)\dot{\mathbf{d}}(t')\cdot\mathbf{e}\}$  vanishes sufficiently rapidly after the passage of the laser pulse.

The above formal analysis demonstrates that the hypothetical single-atom HPNS is proportional to the squared magnitude of the FT of the dipole velocity. This expression is the one that should be implemented in numerical simulations of the HPNS. Of course, the FT of the dipole velocity can be converted, by integration by parts, to either the dipolemoment or dipole-acceleration form, or to any of an infinite number of other equivalent forms which may be more or less well suited to numerical analysis. All of these forms are related to the original dipole-velocity form through "limit contributions" arising from the parts' integrations and yield identical HPNS if the limit contributions are accounted for. However, one cannot simply choose a form out of numerical convenience and use it alone to compute the HPNS without regard for the limit contributions, which generally depend on the character of the laser pulse.

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