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1972 J. Phys. A: Gen. Phys. 5 L119

(http://iopscience.iop.org/0022-3689/5/11/014)

J. Phys. A: Gen. Phys., Vol. 5, November 1972. Printed in Great Britain. © 1972.

LETTER TO THE EDITOR

Quantum effects in second harmonic generation

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MS received 21 August 1972

Abstract. A simple quantum-mechanical model shows a new qualitative feature of the process of second harmonic generation. This feature is the impossibility of complete depletion of the fundamental wave and is not present in the classical description.

The process of second harmonic generation (SHG) is usually schematized by means of a classical model, in which two electromagnetic modes of frequency ω_1 and $\omega_2 = 2\omega_1$ are made to interact while propagating in a nonlinear crystal. A complete analytical treatment of this model has been given by Armstrong *et al* (1962). In particular, whenever the SH field is not present at the boundary $z = 0$ of the crystal, the real amplitude $\rho_1(z)$ of the fundamental wave $E_1(z, t) = \rho_1(z)\exp\{i(k_1 z-\omega_1 t)\}$ evolves, in the case of perfect phase-matching and provided that $|\partial^2\rho_1/\partial z^2| \ll k_1|\partial\rho_1/\partial z|$, as

$$\rho_1(z) = \rho_1(0)\,\mathrm{sech}(s\rho_1(0)z), \tag{1}$$

z being the length of the travelled path and s a suitable constant. According to equation (1), a complete power transfer to the SH field takes place as z goes to infinity.

The simplest quantum-mechanical description of this model is in terms of the Hamiltonian:

$$H = \hbar\omega_1 a_1^\dagger a_1 + \hbar\omega_2 a_2^\dagger a_2 + \hbar g a_1^2 a_2^\dagger + \hbar g^* a_1^{\dagger 2} a_2, \tag{2}$$

where a_i and $a_i^\dagger$ label the annihilation and creation operators relative to the ith mode, and g is the coupling constant between the two modes. The equations of motion for the number operators $n_i(t) = a_i^\dagger(t)a_i(t)$ are obtained by resorting to the Heisenberg equation of motion valid for any operator A:

$$i\hbar\frac{dA}{dt} = [A, H] \tag{3}$$

and taking into account the commutation relations

$$[a_i, a_j] = [a_i^\dagger, a_j^\dagger] = 0, \qquad [a_i, a_j^\dagger] = \delta_{ij}. \tag{4}$$

They can be cast, by means of simple operator algebra, in the convenient form of a second order system of equations

$$\frac{d}{dt}n_1 + 2\frac{d}{dt}n_2 = 0 \qquad \text{(energy conservation)} \tag{5}$$

$$\frac{d^2}{dt^2}n_1 = 4|g|^2(4n_1n_2 - n_1^2 + n_1 + 2n_2), \tag{6}$$

which have to be supplemented with the initial conditions

$$\langle\psi|n_1(0)|\psi\rangle = n_{10} \tag{7}$$

and

$$n_2(0)|\psi\rangle = 0, \qquad \left\langle\psi\left|\left(\frac{\mathrm{d}n_1}{\mathrm{d}t}\right)_{t=0}\right|\psi\right\rangle = 0, \tag{8}$$

which corresponds to the initial absence of quanta of frequency ω_2, $|\psi\rangle$ representing the state of the system. The equation of motion for the expectation value $\langle n_1(t)\rangle = \langle\psi|n_1(t)|\psi\rangle$ is immediately obtained by means of equations (5), (6), (7) and (8) in the form

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}\langle n_1(t)\rangle = 4|g|^2\{2\langle n_1(t)n_1(0)\rangle + n_{10} - 3\langle n_1{}^2(t)\rangle\}. \tag{9}$$

We observe that, since $\langle n_1{}^2\rangle \geqslant \langle n_1\rangle$, equation (9) yields

$$\left[\frac{\mathrm{d}^2}{\mathrm{d}t^2}\langle n_1(t)\rangle\right]_{t=0} \leqslant 0,$$

the equality holding if $|\psi\rangle$ is a linear combination of $|0\rangle$ and $|1\rangle$ number states. This has the physical meaning that the SHG process cannot start if the probability of finding at least two photons of frequency ω_1 is zero, since energy conservation requires the annihilation of two quanta with energy $\hbar\omega_1$ in order to obtain a quantum with energy $\hbar\omega_2$.

It is obvious that solving equation (9), without the help of a hierarchy of equations for higher order expressions of the kind $\langle n_1{}^k(t)n_1{}^h(0)\rangle$, requires a suitable factorization hypothesis on $\langle n_1(t)n_1(0)\rangle$ and $\langle n_1{}^2(t)\rangle$. Anyway, it is possible to derive a relevant consequence of equation (9) without any particular assumption. More precisely, the following property holds: for any given time $t_0 > 0$, the relation

$$\langle n_1{}^2(t)\rangle < \tfrac{1}{3}n_{10} - \epsilon, \tag{10}$$

ϵ being an arbitrary small positive quantity, cannot be identically satisfied for all $t > t_0$. This in turn physically means that it is impossible to have complete depletion of the fundamental mode. In order to prove this property, let us suppose equation (10) to be valid for any $t > t_0$. From equations (9) and (10) it immediately follows that:

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}\langle n_1(t)\rangle > 4|g|^2\{2\langle n_1(t)n_1(0)\rangle + 3\epsilon\} \geqslant 12|g|^2\epsilon; \tag{11}$$

on the other hand, a function $f(t)$ for which $\mathrm{d}^2f/\mathrm{d}t^2 > C > 0$ for $t > t_0$ must satisfy

$$\lim_{t\to\infty} f(t) = +\infty,$$

so that

$$\lim_{t\to\infty} \langle n_1(t)\rangle = +\infty.$$

This in turn implies, since $\langle n_1{}^2(t)\rangle \geqslant \langle n_1(t)\rangle$,

$$\lim_{t\to\infty} \langle n_1{}^2(t)\rangle = +\infty$$

which contradicts the assumption contained in equation (10).

An explicit solution of equation (9) can be obtained by assuming the factorization hypotheses

$$\langle n_1^2(t)\rangle = \langle n_1(t)\rangle^2 + \langle n_1(t)\rangle \tag{12}$$

$$\langle n_1(t)n_1(0)\rangle = \langle n_1(t)\rangle n_{10} + \langle n_1(t)\rangle, \tag{13}$$

which hold true if the fundamental field is assumed to remain (approximately) 'coherent' (Glauber 1963) during the interaction. This choice assures, whenever $\langle n_1(t)\rangle \gg 1$, that the fundamental field is as close as possible to a classical field with a well-stabilized amplitude (Glauber 1963). According to equations (12) and (13), equation (9) reduces to

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}\langle n_1(t)\rangle = -4|g|^2\{3\langle n_1(t)\rangle^2 - (2n_{10}-1)\langle n_1(t)\rangle - n_{10}\}, \tag{14}$$

whose solution, verifying the initial conditions given in equations (7) and (8), can be implicitly written in terms of an elliptic integral as (see, for example, Abramowitz and Segun 1965)

$$t = \left(\frac{1}{8|g|^2}\right)^{1/2} \int_{n_1(t)}^{n_{10}} \frac{\mathrm{d}x}{\{-(x-n_{10})(x-\beta_2)(x-\beta_3)\}^{1/2}}, \tag{15}$$

with

$$\beta_{2,3} = -\tfrac{1}{2} \pm \tfrac{1}{2}(1+8n_{10})^{1/2}.$$

Equation (15) can be put in explicit form for $n_{10} \gg 1$, thus obtaining (Abramowitz and Segun 1965)

$$\langle n_1(t)\rangle = n_{10}\,\mathrm{sech}^2(\gamma t) + \beta_2 \tanh^2(\gamma t) \tag{16}$$

with $\gamma = (2|g|^2 n_{10})^{1/2}$, which can be compared with $\rho_1{}^2(z)$ as given by equation (1). The quantum effect is present in the term $\beta_2 \tanh^2(\gamma t)$, a contribution which tends to a nonvanishing value β_2 as $t \to \infty$.

It is worthwhile to observe that our quantum model refers to a closed lossless cavity. Anyway, our results can be adapted to the travelling wave situation by means of the substitution $t = z/c$ and considering the cavity to possess a volume $V = ScT$, where S labels the transverse section of the beam, c is the velocity of light and T the time during which the counting apparatus remains active (Shen 1967).

We wish finally to recall that a general method for treating quantum-mechanically nonlinear optical phenomena has been introduced by Walls (1971). Its applicability, at least for what concerns the research of analytical solutions, is however limited for our problem to the case in which a small number of quanta n_{10} is present.

This work was supported in part by the Italian National Council of Research.

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