Why you should *not* use the electric field to quantize in nonlinear optics

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Abstract

We show that using the electric field as a canonical quantization variable in nonlinear optics leads to incorrect expressions for the squeezing parameters in SPDC and conversion rates in frequency conversion.

Quantization recipe

1. Write a Hamiltonian H in terms of the relevant fields $\mathbf{F}$ so that it is numerically equal to the energy.
2. Define a set of canonical commutation relations for the fields.
3. Check that the Heisenberg equations of motion (EOM)

$$\dot{\mathbf{F}} = \frac{[\mathbf{F}, H]}{i\hbar} \tag{1}$$

are the Equations of Motion for the fields [1].

For the Electromagnetic field (EMF) for example [1, 2]:

The Hamiltonian is

$$H = \int d^3\mathbf{r} \left(\frac{\mathbf{B}^2}{2\mu_0} + \epsilon_0 \mathbf{E} \cdot \left\{ \frac{1+\chi^{(1)}}{2} \mathbf{E} + \sum_{n\geq 2} \frac{n}{n+1} \chi^{(n)} : \mathbf{E}^n \right\} \right) \tag{2}$$

$$= \int d^3\mathbf{r} \left(\frac{\mathbf{B}^2}{2\mu_0} + \sum_{n\geq 1} \frac{1}{n+1} \mathbf{D} \cdot \eta^{(n)} : \mathbf{D}^n \right) \tag{3}$$

The (only nonzero) commutation relation is

$$[D^i(\mathbf{r}), B^j(\mathbf{r}')] = i\hbar \epsilon^{ijk} \frac{\partial}{\partial r^k} \delta(\mathbf{r} - \mathbf{r}') \tag{4}$$

and the Heisenberg EOMs are Maxwell's equations

$$\dot{\mathbf{D}} = \nabla \times \mathbf{B}/\mu_0, \quad \dot{\mathbf{B}} = -\nabla \times \mathbf{E} \tag{5}$$

if the following constitutive relation is assumed

$$\mathbf{E} = \frac{\mathbf{D} - \mathbf{P}}{\epsilon_0} = \sum_n \eta^{(n)} : \mathbf{D}^n \tag{6}$$

which defines macroscopic polarization in terms of $\mathbf{D}$ and $\eta^{(n)}$. Also can define in terms of $\mathbf{E}$ and nonlinear susceptibilities $\chi^{(n)}$

$$\eta^{(1)} = \epsilon_0^{-1}(1+\chi^{(1)})^{-1}, \tag{7}$$

$$\eta^{(2)} = -\epsilon_0 \eta^{(1)} \chi^{(2)} : \eta^{(1)} \eta^{(1)} \tag{8}$$

What goes wrong with the electric field?

Can you quantize with $\mathbf{E}$ and still satisfy Maxwell's Equations in a nonlinear medium?

Let's try to write

$$\mathbf{E}(\mathbf{r}) = \sum_J \int d\mathbf{k}\ \mathbf{e}_J(\mathbf{k},\mathbf{r}) a_J(\mathbf{k}) + \text{H.c.} = \text{poly}_1(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger)$$

$$\mathbf{B}(\mathbf{r}) = \sum_J \int d\mathbf{k}\ \mathbf{b}(\mathbf{k},\mathbf{r}) a_J(\mathbf{k}) + \text{H.c.} = \text{poly}_1(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger),$$

$$[a_J(\mathbf{q}), a^\dagger_{J'}(\mathbf{q}')] = \delta_{JJ'} \delta(\mathbf{q}' - \mathbf{q}) \tag{9}$$

$\text{poly}_n(x,y)$: polynomial of degree n in variables x, y. Then,

$$H = \text{poly}_{n+1}(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger)(\text{if } \chi^{(n)} \neq 0) \tag{10}$$

Now let us look at Faraday's law of induction

$$-\nabla \times \mathbf{E} = [\mathbf{B}, H]/(i\hbar) = \dot{\mathbf{B}}$$

$$\text{poly}_1(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger) \stackrel{?}{=} [\text{poly}_1(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger), \text{poly}_{n+1}(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger)]$$

$$\text{poly}_1(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger) \stackrel{?}{=} \text{poly}_n(a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger) \tag{11}$$

The electric field has to be a *nonlinear* function of $a_J(\mathbf{k}), a_J(\mathbf{k})^\dagger$.

No, you cannot.

Quantization with $\mathbf{D}$

If quantization is done with $\mathbf{D}$ and $\mathbf{B}$ then

$$\mathbf{D}(\mathbf{r}) = \sum_J \int d\mathbf{k}\ \mathbf{d}_J(\mathbf{k},\mathbf{r}) a_J(\mathbf{k}) + \text{H.c.} \tag{12}$$

Constitutive relation Eq. (6) gives $\mathbf{E}$ as a *nonlinear* function of the creation and annihilation operators

$$\mathbf{E} = \frac{\mathbf{D} - \mathbf{P}}{\epsilon_0} = \underbrace{\eta^{(1)}\mathbf{D}}_{\mathbf{E}_{\text{linear}}} + \sum_{n>1} \eta^{(n)} : \mathbf{D}^n \tag{13}$$

$$= \sum_n \eta^{(n)} : \left(\sum_J \int d\mathbf{k}\ \mathbf{d}_J(\mathbf{k},\mathbf{r}) a_J(\mathbf{k}) + \text{H.c.} \right)^n \tag{14}$$

This same conclusion is found by looking at Eq. (11).

Consequences for bright SPDC

- 3 guided modes on a waveguide labeled A, B, C
- Phase and energy matched

$$\bar{k}_C = \bar{k}_A + \bar{k}_B \text{ and } \bar{\omega}_C = \bar{\omega}_A + \bar{\omega}_B. \tag{15}$$

- Nonlinear part of the Hamiltonian in the interaction picture with respect to free propagation is

$$H_I(t) = \hbar\theta' \int dk_A dk_B dk_C e^{i(\omega_A(k_A)+\omega_B(k_B)-\omega_C(k_C))t} \tag{16}$$

$$\times \Phi(k_A, k_b, k_c) a^\dagger_A(k_A) a^\dagger_B(k_B) a_C(k_C) + \text{H.c.}$$

Using $\mathbf{E}_{\text{linear}}$ the interaction picture Hamiltonian is a factor of two larger than what is predicted with the correct quantization.

Compare factor of $\frac{n}{n+1}$ in Eq. (2) vs. $\frac{1}{n+1}$ in Eq. (3).

Effect on twin beams generation

- Mode C in coherent state with $N \gg 1$ photons:

$$a_C(k_C) \sim \langle a_C(k_C) \rangle = \sqrt{N} \alpha(k_C) \tag{17}$$

- The mean number of photons in the twin beams is

$$\langle N_{A/B} \rangle = \sinh^2(\theta\sqrt{N}). \tag{18}$$

- With $\mathbf{E}_{\text{linear}}$: $\langle N_{A/B\ \text{linear}} \rangle = 4 \langle N_{A/B} \rangle^2$.

References

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