

# Comments on a Proposed Quantization of Interacting Fields

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The meaning of a recently proposed formalism for quantization of interacting fields is discussed by studying the consequences of the time-dependent unitary transformation which is essential for this approach. It turns out that non-relativistic quantum electrodynamics in dipole approximation may serve as a useful, although rather singular, example for this method. In the relativistic case a different point of view is suggested in order to avoid inconsistent interpretation. It is further possible to give arguments for a reasonable choice of the unitary transformation concerned.

## 1. Introduction

In a recent paper concerning the quantization of interacting fields<sup>1</sup> it was pointed out that the canonical quantization rules, when applied to classical electrodynamics, do not lead to a unique formulation of quantum electrodynamics (QED). The reason is that one can perform time-dependent unitary transformations before quantizing the electromagnetic field. The proposed alternative for quantization of interacting fields<sup>1</sup> uses the possibility of decomposing the total Hamiltonian of the system (particle plus radiation field) into three terms: the Hamiltonian of the particle  $H_0$  (e. g. a bound or free electron), that of the free radiation field  $H_{\text{rad}}$ , and the interaction term  $H_{\text{int}}$ . While the radiation field at first is considered as classical, the Hamiltonian  $H_0 + H_{\text{int}}$  is subjected to a time-dependent unitary transformation  $U = \exp\{iS(t)\}$ . The motivation for the special choice of the transformation is that it should lead to a transformed system where the interaction term is not present any more, or at least is minimized. The reaction of the particle on the field is then taken into account by quantizing the free radiation field and adding its Hamiltonian to  $\tilde{H}_0 = U H_0 U^{-1}$ . On the resulting system, described by  $\tilde{H}_0 + H_{\text{rad}}$ , one now applies the inverse transformation which leads to a Hamiltonian that is different from the usual one but should yield the same physics. The aim (or hope) of course is to obtain a description of the system that avoids the difficulties inherent in the usual formulation. The method is illustrated in<sup>1</sup> by considering two cases. First the non-relativistic electron interacting with the radiation

field (approximated by the dipole term) is studied, and then the possibility of generalizing to relativistic QED is discussed. And in fact in the case of the first example a Hamiltonian is obtained for which no explicit renormalization of the electron mass is necessary any more. The term usually added to the Hamiltonian in an ad hoc way in order to get a finite non-relativistic Lamb shift appears here as the result of the transformation mentioned, after having quantized the radiation field.

Because of this encouraging result and its possible generalization it seems necessary to look more closely at the proposed quantization procedure. The purpose of the present paper is to discuss and to clarify the interpretation of this formalism. In order to find the correct transformation a more general approach will be used. First we shall find that the non-relativistic case in dipole approximation indeed may serve as a useful, although rather singular, example for this quantization prescription. On the other hand, in the case of generalization to relativistic QED it turns out that the transformation under discussion is not able to transform the system to the free case as claimed in<sup>1</sup>. In order to avoid arbitrariness and inconsistent interpretation a different point of view regarding this problem is proposed.

## 2. The Time-Dependent Unitary Transformation

To begin with we consider the Hamiltonian  $H_0$  of a massive particle  $m$  of charge  $e$  coupled by  $H_{\text{int}}$  to the classical radiation field  $H_{\text{rad}}$ . Following the method proposed in<sup>1</sup> we first apply a time-dependent

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<sup>1</sup> W. C. HENNEBERGER, Nucl. Phys. B 23, 365 [1970].



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unitary transformation (containing the classical electromagnetic field) to the system described by

$$(H_0 + H_{\text{int}}) \psi(\mathbf{r}, t) = i \frac{\partial}{\partial t} \psi(\mathbf{r}, t) \quad (1)$$

More precisely one is looking for a transformation  $\psi \rightarrow \tilde{\psi} = U \psi$  such that in the transformed system the interaction term  $H_{\text{int}} \rightarrow \tilde{H}_{\text{int}} = U H_{\text{int}} U^{-1}$  is not present any more. In order to see how this could be achieved in general we apply the transformation, unspecified at first,

$$U = \exp\{i S(t)\} \quad (2)$$

to the system (1) and obtain

$$\begin{aligned} \{U H_0 U^{-1} + U H_{\text{int}} U^{-1}\} \tilde{\psi} &= i \left\{ U \frac{\partial}{\partial t} U^{-1} \right\} \tilde{\psi}, \\ \left\{ \tilde{H}_0 + H_{\text{int}} + \frac{i}{1!} [S, H_{\text{int}}] + \frac{(i)^2}{2!} [S, [S, H_{\text{int}}]] \right. \\ &\quad \left. + \frac{(i)^3}{3!} [S, [S, [S, H_{\text{int}}]]] + \dots \right\} \tilde{\psi} \quad (3) \\ &= i \left\{ \frac{\partial}{\partial t} - \frac{i}{1!} \dot{S} - \frac{(i)^2}{2!} [S, \dot{S}] \right. \\ &\quad \left. - \frac{(i)^3}{3!} [S, [S, \dot{S}]] - \dots \right\} \tilde{\psi}, \end{aligned}$$

because  $[S, \partial/\partial t] \tilde{\psi} = -\dot{S} \tilde{\psi}$ . Putting now

$$\dot{S} = H_{\text{int}}, \quad (4)$$

the interaction Hamiltonian indeed cancels, but unfortunately the other terms will remain. Although the commutator series on each side of Eq. (3) look very similar

$$\begin{aligned} &\left\{ \tilde{H}_0 + \frac{i}{1!} [S, H_{\text{int}}] + \frac{(i)^2}{2!} [S, [S, H_{\text{int}}]] + \dots \right\} \tilde{\psi} \\ &= \left\{ i \frac{\partial}{\partial t} + \frac{i}{2!} [S, H_{\text{int}}] + \frac{(i)^2}{3!} [S, [S, H_{\text{int}}]] + \dots \right\} \tilde{\psi}, \end{aligned}$$

they are not identical because of the slightly different factorials in front of the commutators. Therefore, if one wants to get rid of the interaction term  $H_{\text{int}}$ , besides condition (4) one has to require

$$[S, H_{\text{int}}] = 0, \quad (5)$$

and thus obtains

$$\tilde{H}_0 \tilde{\psi} = i(\partial/\partial t) \tilde{\psi}. \quad (6)$$

### 3. Non-Relativistic Example

It is indeed possible to find a transformation of this kind, at least in a special case. Consider for

instance as in <sup>1,2</sup> the Schrödinger equation for a bound non-relativistic electron (without spin) in a classical radiation field given by the transverse vector potential  $\mathbf{A}(\mathbf{r}, t)$

$$H_0 = (\mathbf{p}^2/2m) + V(\mathbf{r}); \quad H_{\text{int}} = -(e/m) \mathbf{A}(t) \cdot \mathbf{p},$$

where for  $\mathbf{A}(\mathbf{r}, t)$  the dipole approximation  $\mathbf{A}(\mathbf{r}, t) \approx \mathbf{A}(0, t)$  is already introduced. According to (4) one has

$$S = i(e/m) \mathbf{Z}(t) \cdot \nabla, \quad (7)$$

where

$$\mathbf{Z}(t) = \int_{-\infty}^t d\tau \mathbf{A}(\tau)$$

is the Hertz vector of the electromagnetic field. Up to now  $\mathbf{A}$  and  $\mathbf{Z}$  are still classical quantities. Therefore condition (5) evidently is fulfilled<sup>3</sup>. At this stage the system is completed by including the quantized radiation field which has to satisfy the usual canonical commutation relations<sup>4</sup>.

The total Hamiltonian then reads

$$H_{\text{total}} = \tilde{H}_0 + H_{\text{rad}}, \quad H_{\text{rad}} = \frac{1}{2} \sum_{\mathbf{k}, \alpha} (p_{\mathbf{k}, \alpha}^2 + \omega_{\mathbf{k}}^2 q_{\mathbf{k}, \alpha}^2) \quad (8)$$

where  $p_{\mathbf{k}, \alpha}$  and  $q_{\mathbf{k}, \alpha}$  are the canonical conjugate operators as defined by the Fourier expansion of the radiation field  $\mathbf{A}(\mathbf{r}, t)$ . Performing now the inverse transformation one has to take into account the commutation relations of the radiation field and the fact that in the Schrödinger picture  $U$  and  $H_{\text{total}}$  are time-independent operators. The time derivative in the Schrödinger equation therefore does not apply to  $U$  and one obtains

$$(H_0 + U^{-1} H_{\text{rad}} U) \psi = i(\partial/\partial t) \psi. \quad (9)$$

The commutator series for  $U^{-1} H_{\text{rad}} U$  breaks off at the third term and leads to the interesting result

$$U^{-1} H_{\text{rad}} U = H_{\text{rad}} - (e/m) \mathbf{A} \cdot \mathbf{p} + \frac{\delta m}{m} \frac{\mathbf{p}^2}{2m},$$

where  $\delta m$  is the well known mass renormalization term

$$\delta m = \frac{4\alpha}{3\pi} \int_0^{k_{\text{max}}} dk.$$

Therefore, in this formulation there is no need for an explicit mass renormalization in calculating for instance the non-relativistic Lamb shift.

\*  $\hbar = c = 1$

<sup>2</sup> For the literature concerning this special case we point to the quotations in <sup>1</sup>.

<sup>3</sup> This is not true if  $\mathbf{A}$  does depend on space coordinates.

<sup>4</sup> See Ref. <sup>1</sup>, Eq. (11).

#### 4. Generalization to Relativistic QED

It is of course tempting to generalize this method to relativistic QED. In this case it is shown in <sup>1</sup> that the interaction term proportional to  $e^2$  obtained by the analogous procedure does not contain contributions to second-order  $S$ -matrix elements which correspond to (the diagonal terms such as) the electron self energy, photon self energy, and the disconnected graph. However, it should be remarked that in getting this interesting result a crucial assumption is made which, in our opinion, is not fulfilled. Indeed it is assumed that the transformation under discussion (2) has been carried out before quantizing the radiation field. But it is easy to see that in this case condition (5) cannot be satisfied. From the relativistic interaction

$$H_{\text{int}} = -\int d^3\mathbf{r} j_\mu(\mathbf{r}) A_\mu(\mathbf{r}, t);$$

$$j_\mu(\mathbf{r}) = ie : \psi(\mathbf{r}) \gamma_\mu \psi(\mathbf{r}) :$$

one obtains according to (4)

$$S = -\int d^3\mathbf{r} j_\mu(\mathbf{r}) Z_\mu(\mathbf{r}, t) \quad (10)$$

with the generalization of the Hertz vector  $Z_\mu(\mathbf{r}, t)$  satisfying

$$\frac{\partial}{\partial t} Z_\mu(\mathbf{r}, t) = A_\mu(\mathbf{r}, t).$$

A calculation of the commutator (5) leads to

$$[S, H_{\text{int}}] = \int d^3\mathbf{r} d^3\mathbf{r}' Z_\mu(\mathbf{r}, t) A_\nu(\mathbf{r}', t) \times$$

$$\times [j_\mu(\mathbf{r}), j_\nu(\mathbf{r}')] \neq 0, \quad (11)$$

because the commutator of currents does not vanish in general at equal times <sup>5</sup>

$$[j_\mu(\mathbf{r}), j_\nu(\mathbf{r}')] \sim$$

$$\begin{cases} \delta(\mathbf{r} - \mathbf{r}') \psi^\dagger(\mathbf{r}) \sigma_{mn} \psi(\mathbf{r}') & \text{for } m \neq n \neq 4 \\ 0 & \text{for } \mu=4; \nu=4; \mu=\nu, \end{cases}$$

where  $\sigma_{\mu\nu} = 1/2 i[\gamma_\mu, \gamma_\nu]$ .

Therefore the transformation with (2), (10) does not furnish the rotated Hamiltonian  $\tilde{H}_0$  which is a suitable starting point before introducing the quantized radiation field. On the other hand one finds in <sup>1</sup> that only  $\tilde{H}_0$  has been used and the terms arising from the commutator  $[S, H_{\text{int}}] \neq 0$  have not

been discussed. As a consequence it is no longer possible to maintain the statement that the Hamiltonian used in <sup>1</sup> yields the same physics as the usual one. Whether this holds true is still an open question.

One could of course avoid this inconsistency if one is willing to accept a rather different point of view. According to the usual treatment of canonical field quantization one adds to the free Hamiltonian of the system the interaction term (in correspondence to the classical expression) and then imposes on the different fields the quantization rules. Instead of following this line or the equivalent procedure discussed in Sect. 3 one may propose to start with the free particle Hamiltonian  $H_0$  rotated by a special time-dependent unitary transformation

$$H_0 \rightarrow \tilde{H}_0 = U H_0 U^{-1}.$$

For the rotated system the reaction of the particle on the radiation field is now taken into account by quantizing the radiation field according to the canonical commutation rules and adding the Hamiltonian  $H_{\text{rad}}$ . Performing then the inverse rotation one obtains the Hamiltonian (in the Schrödinger picture)

$$H = H_0 + U^{-1} H_{\text{rad}} U$$

which now contains the interaction in the second term.

The crucial question of course is to find (motivate) a reasonable and unique choice of the transformation  $U$ . Looking at the commutator series

$$U^{-1} H_{\text{rad}} U = H_{\text{rad}} + \frac{i}{1!} [H_{\text{rad}}, S]$$

$$+ \frac{(i)^2}{2!} [[H_{\text{rad}}, S], S] + \dots \quad (12)$$

it seems natural to demand for instance that the first commutator should reproduce the usual coupling of the charge current operator to the radiation field which corresponds to the interaction known from classical theory

$$i[H_{\text{rad}}, S] = -\int d^3\mathbf{r} j_\mu(\mathbf{r}) A_\mu(\mathbf{r}). \quad (13)$$

One finds that this condition can be realized by the expression <sup>7</sup>

$$S = -\int d^3(\mathbf{r}) j_\mu(\mathbf{r}) Z_\mu(\mathbf{r}).$$

<sup>5</sup> See also for instance KÄLLÉN <sup>6</sup>, p. 225. We follow the notation used in Ref. <sup>6</sup>.

<sup>6</sup> G. KÄLLÉN, Handbuch der Physik, Bd. V/1, Springer-Verlag, Berlin 1958.

<sup>7</sup> Notice that this expression for  $S$  just yields the transformation used in Ref. <sup>1</sup>.

It should be emphasized that the interaction introduced by the commutator series

$$U^{-1} H_{\text{rad}} U = H_{\text{rad}} - \int d^3\mathbf{r} j_{\mu}(\mathbf{r}) A_{\mu}(\mathbf{r}) \quad (14) \\ + \frac{i}{2!} \int d^3\mathbf{r} d^3\mathbf{r}' [j_{\mu}(\mathbf{r}) A_{\mu}(\mathbf{r}), j_{\nu}(\mathbf{r}') Z_{\nu}(\mathbf{r}')] + \dots$$

is different from the usual one. The term proportional to  $e^2$  in the series (14) leads to encouraging results which are discussed in <sup>1</sup>.

## 5. Conclusion

In conclusion it should be remarked that the proposed quantization scheme leads to the correct clas-

sical limit because for  $c$ -numbers the commutators in (12) vanish. But of course this treatment of interacting fields has to be confronted with all the well established experimental results of QED. It may turn out that the commutator series (12) breaks off at some term for renormalizable theories, as found in the non-relativistic example. For answering these questions a discussion of the higher commutators in (14) is necessary.

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# Optische Kern-Spin-Polarisation in Molekül-Kristallen \*

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This paper deals with the polarisation of nuclei in molecular crystals due to their hyperfine interaction with optically excited triplet states and excitons, a phenomenon which is termed optical nuclear polarisation (ONP).

A theory is presented which extends the mechanisms of dynamic nuclear polarisation (Overhauser effect) to the case of spin systems containing triplet states with  $S=1$  and nuclei with  $I=1/2$ . In this mechanism the optical electron polarisation (OEP) caused by symmetry selection rules for intersystem crossing to and from the magnetic triplet sublevels is assumed to be transferred to the nuclear magnetic substates by efficient hyperfine relaxation transitions.

The adiabatic fast passage is used to detect the nuclear polarisation. The advantages and conditions of this technique as compared to other nmr techniques are discussed.

ONP results at room temperature are given as a function of the external field  $H_0$ , its orientation with respect to the crystalline axes, the intensity and frequency of the exciting light and the concentration and types of guest molecules. In phenazine crystals relative polarisation factors up to 50 are found with marked orientation dependencies even in fields as high as 11 kG. These results can be interpreted in principle in terms of the presented theory. However, in low fields (0–200 G) where ONP caused by hyperfine relaxation vanishes, large polarisations are found in doped crystals of fluorene and anthracene. The maximum absolute polarisation in fluorene doped with acridine is  $3.6 \cdot 10^{-4}$  at  $H_0 = 80$  G corresponding to a relative polarisation factor of  $\sim 10^4$ . The effect of doping is discussed. Reference is made to a possible ONP mechanism <sup>3</sup> which is able to produce large nuclear polarisations at low fields.

## I. Einleitung

Die 1967 entdeckte <sup>1</sup> Polarisation von Protonen durch Licht beruht auf Wechselwirkungen zwischen Kernen mit dem Spin  $I=1/2$  und den beiden Elektronen mit dem Spin  $S=1$  von optisch angeregten paramagnetischen Triplettzuständen. In dieser Arbeit wird über eine systematische Untersuchung dieses

als optische Kernpolarisation (ONP) bezeichneten Effekts in aromatischen Molekülkristallen bei Zimmertemperatur berichtet. Dabei wurden folgende Parameter variiert:

1. Art und Konzentration der Gastmoleküle,
2. Größe des äußeren Magnetfeldes  $H_0$  und seine Orientierung relativ zu den Kristallachsen,

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<sup>1</sup> G. MAIER, U. HAEERLEN, H. C. WOLF u. K. H. HAUSSER, Phys. Lett. 25 A, 384 [1967].