On quantum logic operations based on photon-exchange interactions in an ensemble of non-interacting atoms

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The recently proposed idea to generate entanglement between photon states via exchange interactions in an ensemble of atoms (J. D. Franson and T. B. Pitman, Phys. Rev. A **60**, 917 (1999) and J. D. Franson *et al.*, (quant-ph/9912121)) is discussed using an S-matix approach. It is shown that if the nonlinear response of the atoms is negligible and no additional atom–atom interactions are present, exchange interactions cannot produce entanglement between photons states in a process that returns the atoms to their initial state. Entanglement generation requires the presence of a nonlinear atomic response or atom–atom interactions.

I. INTRODUCTION

In some recent papers Franson *et al.*, [1,2] suggested that exchange interactions of two photons in a macroscopic ensemble of identical, non-interacting atoms could lead to large conditional phase shifts. In contrast to "conventional" nonlinear optics which requires scattering of both photons from the same atom, exchange interactions are present even when the two photons interact with different atoms. This makes them much more likely to occur in a dense medium. The large magnitude of the predicted conditional phase shifts would make such systems very attractive for quantum logical operation. However, whether or not exchange interactions are capable of generating entanglement between photons has been subject of some debate [3,4]. In view of the claimed potential advantages, the requirements and limitations of the proposed schemes need to be examined.

In the present note I want to discuss a special type of exchange interactions. In particular I will analyze the possibility to entangle photon states through interactions in an ensemble of atoms under the conditions considered in [2]. Namely: (*i*) All processes are unitary, i.e. losses are negligible; (*ii*) The atomic system returns to the same state as before the interaction; (*iii*) The "conventional" nonlinear response of the atoms is assumed to be negligible; (*iv*) It is assumed that there are no atom–atom interactions, except those through the quantized radiation modes under consideration. Conditions (*i*) and (*ii*) enshure that the pair of qubits, represented by the photons undergoes an effective unitary evolution and is asymptotically disentangled from the atoms and the environment. It will be shown in the following that in a system that fulfills conditions (*i-iv*) entanglement between a pair of photons in distinguishable modes can *not* be generated. Any initially factorizable state will evolve into a factorizable state.

II. MODEL AND EFFECTIVE TIME-EVOLUTION OPERATOR

Let me consider the interaction of the quantized radiation field with a large number of identical atoms in dipole and rotating-wave approximation as proposed in [1,2]. In addition to the photon field, the atoms may be coupled to some external *classical* fields to allow for manipulations of the states after or during the interaction with the photons. The Hamiltonian of the system has the following general form

$$H = H_{\rm field} + H_{\rm atom}(t) + V, \tag{1}$$

where $H_{\rm field}$ is the free Hamiltonian of the quantized photon field and $H_{\rm atom}(t)$ is the free Hamiltonian of the atoms including the interaction with the (time-dependent) external, classical fields. For simplicity it is assumed that each mode of the photon field couples only to one atomic transition. It is however straight forward to lift this restriction. The interaction operator has thus the following general structure

$$V = -\hbar \sum_k g_k \sum_{j=1}^{N} \left[\hat{\sigma}^\dagger_{j,k} \hat{a}_k\, f_k(\vec{r}_j) + \hat{\sigma}_{j,k} \hat{a}^\dagger_k f^*_k(\vec{r}_j)\right]. \tag{2}$$

Here $\hat{a}_k$ and $\hat{a}^\dagger_k$ are annihilation and creation operators of the photon field. k is a mode index and $f_k(\vec{r})$ is the associated mode function. f_k is not restricted to plane waves but could also represent e.g. localized wave packets, distinguishable by their arrival time. The modes are assumed to be orthogonal, such that $[a_k, a^\dagger_{k'}] = \delta_{kk'}$. $\hat{\sigma}_{j,k}$ denotes a flip operator of atom j corresponding to the transition coupled to the mode k with coupling strength g_k. (Introducing flip operators for different k-values takes into account that the individual modes of the quantized field may be coupled to different dipole transitions.)

It is assumed that initially ($t = t_0$) all atoms are in their ground states, i.e. the total initial state vector has the form

$$|\psi(t_0)\rangle = |\phi(t_0)\rangle\, |g\rangle, \tag{3}$$

where $|\phi(t_0)\rangle$ is the initial field state and $|g\rangle$ the collective ground state of the atoms.

The Schrödinger-equation for the state vector in the interaction picture can formally be solved by

$$|\psi(t)\rangle = \mathsf{T} \exp\left\{-\frac{i}{\hbar}\int_{t_0}^{t} {\rm d}t'\, V(t')\right\} |\psi(t_0)\rangle, \tag{4}$$

where T is the time ordering operator.

It is clear that photon-atom interactions in general entangle both sub-systems. This is however not of interest here. The question I want to address is, whether the interaction can generate an entangled state of the *photons* given that the atomic system returns to its initial ground state at some time t_1. Thus we require

$$|\psi(t_1)\rangle \longrightarrow |\phi(t_1)\rangle |g\rangle. \tag{5}$$

In this case the atomic and photonic components of $|\psi(t_1)\rangle$ factorize and the photonic part is given by

$$|\phi(t_1)\rangle = \langle g|\,\mathsf{T}\exp\left\{-\frac{i}{\hbar}\int_{t_0}^{t_1}\mathrm{d}t'\,V(t')\right\}|g\rangle\,|\phi(t_0)\rangle = \mathsf{S}(t_1,t_0)\,|\phi(t_0)\rangle. \tag{6}$$

The operator S describes the conditional evolution of the photon field when the atomic system returns to its ground state.

In order to calculate the action of S, we make use of a generalization of the cumulant generation function for a classical statistical variable X

$$\Big\langle \exp\{sX\}\Big\rangle_X = \exp\left\{\sum_{m=0}^{\infty}\frac{s^m}{m!}\langle\langle X^m\rangle\rangle\right\}. \tag{7}$$

Here $\langle\langle X^m\rangle\rangle$ denotes the mth order cumulant, i.e. $\langle\langle X\rangle\rangle = \langle X\rangle$, $\langle\langle XY\rangle\rangle = \langle XY\rangle - \langle X\rangle\langle Y\rangle$ etc. Applying eq.(7) to S yields

$$\begin{aligned}\mathsf{S}(t_1,t_0) = \mathsf{T}\exp\Bigg\{&\int\!\mathrm{d}1\!\int\!\mathrm{d}2\;\hat a^\dagger_{k_1}(\tau_1)\,\mathcal{P}(1;2)\,\hat a_{k_2}(\tau_2)\\ &+\int\!\mathrm{d}1\!\int\!\mathrm{d}2\!\int\!\mathrm{d}3\!\int\!\mathrm{d}4\;\hat a^\dagger_{k_1}(\tau_1)\hat a^\dagger_{k_2}(\tau_2)\,\mathcal{P}^{(2)}(1,2;3,4)\,\hat a_{k_3}(\tau_3)\hat a_{k_4}(\tau_4)+\cdots\Bigg\}\end{aligned} \tag{8}$$

where $\int \mathrm{d}1$ stands for integration over time τ_1 and summation over the mode index k_1. It was assumed here for simplicity that the average dipole moment of the atoms vanishes.

$$\mathcal{P}(1,2) = \sum_j \mathcal{P}^j(1,2), \tag{9}$$

where

$$\mathcal{P}^j(1,2) = -g_k^2 f^*_{k_1}(\vec r_j) f_{k_2}(\vec r_j)\Big\langle\Big\langle \mathsf{T}\hat\sigma^\dagger_{jk_1}(\tau_1)\hat\sigma_{jk_2}(\tau_2)\Big\rangle\Big\rangle \tag{10}$$

describes the *linear* response of the jth atom to the quantized radiation field. The higher-order terms $\mathcal{P}^{(n)}$ characterize the "conventional" nonlinear response. The scattering of two photons off the same atom is for example determined by $\mathcal{P}^{(2)}$. It should be emphasized here, that cumulants containing operators of different atoms vanish, since it was assumed that atom–atom correlations can be built up only by the quantized radiation field. As a consequence each term $\mathcal{P}^{(n)}$ scales only linearly with the number of atoms N. Thus "conventional" nonlinear interactions of increasing order require increasing photon densities or large coupling constants g_k.

Franson *et al.* argued in [2] that a nonlinear phase shift between two photons could emerge even if the "conventional" nonlinear couplings, characterized by the higher-order cumulants in eq.(8), are negligible. Such phase shifts should arize from exchange interactions resulting from to the symmetrization requirements imposed by the bosonic nature of the photons. Let me therefore consider in the following the case were all higher-order cumulants are neglected. In this situation S reduces to:

$$\mathsf{S} \approx \mathsf{T}\,\exp\Bigg\{\int\!\mathrm{d}1\!\int\!\mathrm{d}2\,\hat a^\dagger_{k_1}(\tau_1)\,\mathcal{P}(1,2)\,\hat a_{k_2}(\tau_2)\Bigg\}. \tag{11}$$

It should be emphasized that although the evolution operator (11) is bilinear in the photon operators, it takes fully into account any exchange interaction. The implicit summation over mode indices accounts for processes where photon 1 is seen by atom A and photon 2 by atom B as well as the case where photon 1 is seen by atom B and photon 2 by atom A. It will now be shown that the conditional evolution $t_0 \to t_1$ described by S cannot generate entanglement. I.e. any initially factorizable state will evolve into a factorizable state after the interaction.

III. STATE EVOLUTION

In order to discuss the evolution of photons described by S in (11), I consider the case of the field initially being in a factorizable two-mode state with at most one photon in each mode. $|\phi(t_0)\rangle = |\phi_1\rangle\,|\phi_2\rangle\,|\{0_k\}\rangle$ with

$$|\phi_1\rangle = \left(\alpha_1 + \beta_1 \hat{a}^\dagger_{k_1}\right)\,|0_1\rangle, \qquad |\phi_2\rangle = \left(\alpha_2 + \beta_2 \hat{a}^\dagger_{k_2}\right)\,|0_2\rangle. \tag{12}$$

Here $|0_1\rangle, |0_2\rangle$ are the vacuum states of modes k_1 and k_2 and $|\{0_k\}\rangle$ is the vacuum state of all other modes.

I proceed with discussing the evolution of the individual components of $|\phi(t_0)\rangle$. The vacuum component remains of course unaffected and it is sufficient to consider

$$|\chi_1(t_1)\rangle = \mathsf{S}(t_1,t_0)\,|\chi_1(t_0)\rangle = \mathsf{S}(t_1,t_0)\,\hat{a}^\dagger_{k_1}(t_0)|0\rangle, \tag{13}$$

and

$$|\chi_{1,2}(t_1)\rangle = \mathsf{S}(t_1,t_0)\,|\chi_{1,2}(t_0)\rangle = \mathsf{S}(t_1,t_0)\,\hat{a}^\dagger_{k_1}(t_0)\,a^\dagger_{k_2}(t_0)|0\rangle. \tag{14}$$

To formally calculate these expressions we make use of Wick's theorem, which states that a time-ordered operator expression can be replaced by the sum of all normally ordered expressions with all possible "contractions". Contractions refer here to a replacement of any operator pairs $\hat{a}^\dagger_{k'}(\tau')$ and $\hat{a}_{k''}(\tau'')$ by the T-ordered propagator

$$\mathcal{D}(1,2) = \left\langle 0\right|\mathsf{T}\,\hat{a}^\dagger_{k'}(\tau_1)\hat{a}_{k''}(\tau_2)\left|0\right\rangle. \tag{15}$$

We first note that since t_0 is the smallest time, the creation operators $\hat{a}^\dagger_{k_1}(t_0)$ and $\hat{a}^\dagger_{k_2}(t_0)$ in eqs.(13) and (14) can be included in the T-ordering. Since $\mathsf{S}\,\hat{a}^\dagger_{k_1}(t_0)$ and $\mathsf{S}\,\hat{a}^\dagger_{k_1}(t_0)\hat{a}^\dagger_{k_2}(t_0)$ respectively act on the vacuum state, out of all normally ordered expressions only those survive which have no photon annihilation operator left.

Now $\mathsf{S}\,\hat{a}^\dagger_{k_1}(t_0)$ can be expanded into a power series and Wick's theorem applied to each term. This leads to the following perturbation series

$$\begin{aligned}|\chi_1(t_1)\rangle = \Bigg\{\hat{a}^\dagger_{k_1}(t_0) + \int\!\mathrm{d}1\!\int\!\mathrm{d}2\,\mathcal{D}(0,1)\Bigg[\mathcal{P}(1,2) + \int\!\mathrm{d}3\!\int\!\mathrm{d}4\,\mathcal{P}(1,3)\mathcal{D}(3,4)\mathcal{P}(4,2)\\ + \int\!\mathrm{d}3\!\int\!\mathrm{d}4\!\int\!\mathrm{d}5\!\int\!\mathrm{d}6\,\mathcal{P}(1,3)\mathcal{D}(3,4)\mathcal{P}(4,5)\mathcal{D}(5,6)\mathcal{P}(6,2) + \cdots\Bigg]\,\hat{a}^\dagger_{k''}(\tau_2)\Bigg\}|0\rangle,\end{aligned} \tag{16}$$

where $|0\rangle$ denotes the vacuum of all field modes. The first term results from contractions of photon operators within S. The other terms arise from all possible contractions of $\hat{a}^\dagger_{k_1}$ with operators from S.

Eq.(16) can be given the compact form

$$|\chi_1(t_1)\rangle = \left[\hat{a}^\dagger_{k_1}(t_0) + \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0,1)\, \Pi(1,2)\, \hat{a}^\dagger_{k''}(\tau_2)\right] |0\rangle \tag{17}$$

where $\Pi(1,2)$ is the solution to the linear integral equation (Dyson equation)

$$\Pi(1,2) = \mathcal{P}(1,2) + \int \mathrm{d}2 \int \mathrm{d}3\, \mathcal{P}(1,3)\, \mathcal{D}(3,4)\, \Pi(4,2). \tag{18}$$

In fact one easily verifies that an interactive solution of this equations generates the whole perturbation series of (16). That the quantum evolution can formally be solved in such a simple way is not surprising since the system is linear. Eq.(18) describes nothing else than multiple scattering of the incoming photon at the atoms with all nonlinearities being absent. In a diagrammatic language, the Dyson equation (18) corresponds to a sum of chain-like diagrams without branching or merging.

In a similar way as above one can proceed with $\mathsf{S}\, \hat{a}^\dagger_{k_1} \hat{a}^\dagger_{k_2}$. In this case contractions only within S generate a term proportional to the product $\hat{a}^\dagger_{k_1} \hat{a}^\dagger_{k_2}$ similar to the first term in eq.(16). Then two series of terms emerge where either $\hat{a}^\dagger_{k_1}$ or $\hat{a}^\dagger_{k_2}$ is contracted with operators from S. These leads to expressions identical to the higher-order terms in (16) multiplied with either $\hat{a}^\dagger_{k_1}$ or $\hat{a}^\dagger_{k_2}$. Finally there is a series of terms resulting of contractions of *both* $\hat{a}^\dagger_{k_1}$ and $\hat{a}^\dagger_{k_2}$ with operators from S. This yields

$$\begin{aligned}
|\chi_{12}(t_1)\rangle = \Big\{ & \hat{a}^\dagger_{k_1}(t_0) \hat{a}^\dagger_{k_2}(t_0) + \\
+ & \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0',1) \Big[\mathcal{P}(1,2) + \int \mathrm{d}3 \int \mathrm{d}4\, \mathcal{P}(1,3) \mathcal{D}(3,4) \mathcal{P}(4,2) + \cdots\Big]\, \hat{a}^\dagger_{k_2}(t_0)\, \hat{a}^\dagger_{k''}(\tau_2) + \\
+ & \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0'',1) \Big[\mathcal{P}(1,2) + \int \mathrm{d}3 \int \mathrm{d}4\, \mathcal{P}(1,3) \mathcal{D}(3,4) \mathcal{P}(4,2) + \cdots\Big]\, \hat{a}^\dagger_{k_1}(t_0)\, \hat{a}^\dagger_{k''}(\tau_2) + \\
+ & \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0',1) \Big[\mathcal{P}(1,2) + \int \mathrm{d}3 \int \mathrm{d}4\, \mathcal{P}(1,3) \mathcal{D}(3,4) \mathcal{P}(4,2) + \cdots\Big] \times \\
\times & \int \mathrm{d}\tilde{1} \int \mathrm{d}\tilde{2}\, \mathcal{D}(0'',\tilde{1}) \Big[\mathcal{P}(\tilde{1},\tilde{2}) + \int \mathrm{d}\tilde{3} \int \mathrm{d}\tilde{4}\, \mathcal{P}(\tilde{1},\tilde{3}) \mathcal{D}(\tilde{3},\tilde{4}) \mathcal{P}(\tilde{4},\tilde{2}) + \cdots\Big]\, \hat{a}^\dagger_{k''}(\tau_2)\, \hat{a}^\dagger_{\tilde{k}''}(\tilde{\tau}_2) \Big\} |0\rangle.
\end{aligned} \tag{19}$$

Here $0'$ and $0''$ stand for $\{t_0, k_1\}$ and $\{t_0, k_2\}$ respectively. This expression can again be brought into a compact form

$$\begin{aligned}
|\chi_{12}(t_1)\rangle &= \hat{a}^\dagger_{k_1}(t_0) \hat{a}^\dagger_{k_2}(t_0)\, |0\rangle \\
&+ \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0',1)\, \Pi(1,2) \hat{a}^\dagger_{k''}(\tau_2) \hat{a}^\dagger_{k_2}(t_0) |0\rangle + \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0'',1)\, \Pi(1,2) \hat{a}^\dagger_{k''}(\tau_2) \hat{a}^\dagger_{k_1}(t_0) |0\rangle \\
&+ \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0',1)\, \Pi(1,2) \int \mathrm{d}\tilde{1} \int \mathrm{d}\tilde{2}\, \mathcal{D}(0'',\tilde{1})\, \Pi(\tilde{1},\tilde{2})\; \hat{a}^\dagger_{k''}(\tau_2)\, \hat{a}^\dagger_{\tilde{k}''}(\tilde{\tau}_2)\, |0\rangle.
\end{aligned}$$

One immediately recognizes that $|\chi_{12}(t_1)\rangle$ can be written as

$$\begin{aligned}
|\chi_{12}(t_1)\rangle = \quad & \left[\hat{a}^\dagger_{k_1}(t_0) + \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0',1)\, \Pi(1,2)\, \hat{a}^\dagger_{k''}(\tau_2)\right] \\
& \otimes \left[\hat{a}^\dagger_{k_2}(t_0) + \int \mathrm{d}\tilde{1} \int \mathrm{d}\tilde{2}\, \mathcal{D}(0'',\tilde{1})\, \Pi(\tilde{1},\tilde{2})\, \hat{a}^\dagger_{\tilde{k}''}(\tilde{\tau}_2)\right] |0\rangle
\end{aligned} \tag{20}$$

The evolution of $|\phi\rangle$ from t_0 to t_1 is hence given by

$$
\begin{aligned}
|\phi(t_0)\rangle &= \left(\alpha_1 + \beta_1 \hat{a}^\dagger_{k_1}\right) \left(\alpha_2 + \beta_2 \hat{a}^\dagger_{k_2}\right) |0\rangle \\
&\downarrow \\
|\phi(t_1)\rangle &= \left[\alpha_1 + \beta_1\left(\hat{a}^\dagger_{k_1}(t_0) + \int \mathrm{d}1 \int \mathrm{d}2\, \mathcal{D}(0',1)\,\Pi(1,2)\,\hat{a}^\dagger_{k''}(\tau_2)\right)\right] \otimes \\
&\quad \left[\alpha_2 + \beta_2\left(\hat{a}^\dagger_{k_2}(t_0) + \int \mathrm{d}\tilde{1} \int \mathrm{d}\tilde{2}\, \mathcal{D}(0'',\tilde{1})\,\Pi(\tilde{1},\tilde{2})\,\hat{a}^\dagger_{\tilde{k}''}(\tilde{\tau}_2)\right)\right] \Big|0\Big\rangle .
\end{aligned} \tag{21}
$$

Thus if the process starts with a factorizable state with photons in distinguishable modes, i.e. if $(\alpha_1 + \beta_1 \hat{a}_{k_1})|0\rangle$ is orthogonal to $(\alpha_2 + \beta_2 \hat{a}_{k_2})|0\rangle$ and if the process generates photons in distinguishable modes, i.e. if

$$
\left[\alpha_1 + \beta_1\left(\hat{a}^\dagger_{k_1} + \iint \mathcal{D}\,\Pi\,\hat{a}^\dagger_{k''}\right)\right] \Big|0\Big\rangle \qquad \text{and} \qquad \left[\alpha_2 + \beta_2\left(\hat{a}^\dagger_{k_2} + \iint \mathcal{D}\,\Pi\,\hat{a}^\dagger_{\tilde{k}''}\right)\right] \Big|0\Big\rangle
$$

are orthogonal, then the generated state vector remains factorizable.

IV. CONCLUSION

In the present note I have shown it is not possible to generate entanglement between photons using *solely* exchange interactions in a large ensemble of atoms, if the atoms are left in the same quantum state after the interaction as they were initially. From a diagrammatic point of view entanglement between photons can not be generated if all possible diagrams are chain-like. To produce entanglement non-trivially connected diagrams are needed, as emerge for example from nonlinear atomic responses or from atom–atom interactions due to e.g. dipole-dipole or collisional interactions.

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