Long-range steady entanglement in a resonantly laser-excited atomic ensemble

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We show that slow two-level atoms in a weak resonant laser field, are entangled. The two considered groups of atoms can be separated by a macroscopic distance, and be parts of a larger atomic ensemble. In a dilute regime, for two very distant groups of atoms, we determine the maximum attainable entanglement negativity, and a laser intensity below which they are certainly entangled. They both decrease with increasing distance between the two groups, but increase with enlarging groups sizes.

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The impact of the environment on quantum entanglement is manifold. On the one hand, an initial entanglement between independent systems, can be destroyed by their coupling to their surroundings. This fragility of quantum entanglement is substantiated by the fact that it can vanish in finite time, as first shown for two-level atoms in distinct vacuum cavities \(^1\). On the other hand, the environment mediates interactions, Hamiltonian or not, between the considered systems, and can thus induce correlations between them, and hence potentially entanglement. It has been shown that finite entanglement can develop between two initially uncorrelated two-level systems, or qubits, sharing the same surroundings, but otherwise uncoupled \(^2\). However, in the realistic case of a finite separation between the two systems, this effect is only transient if the environment is in thermal equilibrium \(^1\). This is not the case when the surroundings does not reduce to a thermal bath. The systems steady state, reached asymptotically from any initial state, can present finite entanglement, as has been shown for two qubits in diverse environments, such as the electromagnetic vacuum and a resonant laser field \(^3\), two heat reservoirs at different temperatures \(^4\), and the electromagnetic field emitted by two bodies at different temperatures \(^5\). The entanglement obtained in these works, can be essentially traced back to one of the two familiar features of a pair of infinitely close qubits, which are, the divergence of the dipole-dipole level shifts, and the decoupling, from the environment, of the so-called subradiant state \(^6\). For distant atoms, the dipole-dipole interaction and the collective contributions to spontaneous emission are less pronounced, and a potential entanglement mechanism cannot simply rely on one or the other of their peculiarities. A possible strategy to obtain entanglement between remote effective two-level atoms, consists in a fine environment-engineering, based on dispersive laser-induced transitions to upper levels properly shifted by static electric and magnetic fields, to achieve, as well as possible, a desired purely dissipative dynamics \(^7\).

Following this proposal, long-lived entanglement of two atomic ensembles separated by 50 cm, has been observed experimentally \(^16\), \(^17\). An important difficulty with elaborate surroundings is that each added part not only plays the role it is intended for but also introduces undesired influences which can be detrimental to entanglement.

Here we show that a resonant laser field suffices to induce steady entanglement between groups of two-level atoms, potentially separated by a macroscopic distance, provided its intensity is low enough. The mutual influence between the atoms we consider, arises only from their coupling to the electromagnetic field. Thus, the atoms internal state does not evolve according to a desired effective master equation, but according to the natural Lehmberg’s master equation \(^8\), \(^9\), \(^10\), \(^11\). The underlying physical origin of the found entanglement, is that, for low laser intensities, the atoms internal dynamics is dominated by the dipole-dipole interaction, laser photon absorption and collective radiative decay, which all preserve the state purity. As a result, the atoms steady state is practically pure, and correlated, and hence entangled. Moreover, this remains true if the atoms of interest are surrounded by other identical atoms, and thus, the two considered groups of atoms can be parts of a larger atomic ensemble.

We consider an ensemble of two-level atoms evolving under the influence of a laser field. Within the dipolar approximation for the coupling to the electromagnetic field \(^12\), and a semiclassical approximation for the motion of the atoms \(^13\), the dynamics of the atoms internal state is governed by the Hamiltonian

\[
H = H_e + \omega_0 \sum_{\mu} \sigma_{\mu}^+ \sigma_{\mu} - \sum_{\mu} (\sigma_{\mu}^+ + \sigma_{\mu}) \left[ \mathbf{d} \cdot \mathbf{E}(\mathbf{r}_{\mu}) + \text{Re}(w(\mathbf{r}_{\mu}) \mathbf{e}^{-i\omega t}) \right],
\]

where \(\omega_0\) is the atomic resonance frequency, \(\mathbf{r}_{\mu}\) is the classical position of atom \(\mu\), and \(\Omega\) is proportional to the laser field amplitude. Throughout this paper, units are
used in which $\hbar = 1$. The Hamiltonian $H_c$ and field $E$ read, respectively, $H_c = c \int d^3k |k| (a^\dagger_{k1}a_{k1} + a^\dagger_{k2}a_{k2})$, and

$$E(r) = \int d^3k \left( -\frac{e|k|}{4\pi^2\epsilon_0} \right)^{1/2} \sum_{p=1,2} e_{kp} e^{ik \cdot r} a_{kp} + h.c.,$$

where $c$ is the speed of light, $\epsilon_0$ is the vacuum dielectric permittivity, $\epsilon_{k1}$ and $\epsilon_{k2}$ are unit vectors orthogonal to $k$ and to each other, and the electromagnetic field operators $a_{kp}$ bosonize commutation relations $[a_{kp}, a^\dagger_{k'p'}] = \delta_{pp'} \delta(k - k')$. The atomic operator $\sigma_\mu$ is defined by $\sigma_\mu = |g\rangle \langle \mu|$, where $|g\rangle$ and $|\mu\rangle$ are, respectively, the ground and excited states of atom $\mu$. The vector $d = |\mu\rangle \langle D_\mu|g\rangle_\mu$ where $D_\mu$ is the dipole moment of atom $\mu$, is assumed real and the same for all the atoms. The laser frequency $\omega \simeq \omega_0$ is close to the atomic resonant frequency, and the spatial function $w$ depends on the laser beam considered, $w(r) = \exp(iK \cdot r)$ for a plane wave of wave vector $K$, for example.

For fixed positions $r_\mu$, the timescales relevant to the dynamics of the atoms internal state $\rho$ are $\omega_0^{-1}$, $|r_\mu - r_v|/c$, and $\Gamma^{-1}$ where $\Gamma = |d|^2 \omega_0^3 / 3 \pi \epsilon_0 \alpha^3$ is the spontaneous decay rate of an isolated atom. The ratio $\Gamma / \omega_0$ is of the order of $\alpha^3$ where $\alpha \approx 7 \times 10^{-3}$ is the fine-structure constant. Thus, for distances $|r_\mu - r_v| < k_0^{-1} \alpha^{-3}$ where $k_0 = \omega_0/c$, the timescale $\Gamma^{-1}$ is very long compared to the other ones, and it can be shown that the time evolution of $\rho$ is well described by the master equation

$$\partial_t \rho = -\Gamma \mathcal{L}(\{r_\mu\}) \rho - i[H_a + H_I, \rho],$$

where $H_a$ and $H_I$ are, respectively, the second and last terms of Hamiltonian (1). The superoperator $\mathcal{L}$ is defined by

$$\mathcal{L} = \sum_{\mu,\nu} \left[ z_{\mu\nu} \sigma_\nu \sigma_\mu + z_{\mu\nu}^* \sigma_\mu \sigma_\nu - 2 \gamma_{\mu\nu} \sigma_\nu \sigma_\mu \right],$$

where $\sigma_\mu$ is any matrix, $\gamma_{\mu\nu} = \text{Re} z_{\mu\nu}$, $z_{\mu\nu} = 1/2$, and, for $\mu \neq \nu$,

$$z_{\mu\nu} = \frac{3 \epsilon_{\mu\nu}^r}{4 \pi^3} \left[ 1 - (3 \hat{\mathbf{d}} \cdot \hat{\mathbf{r}})^2 (i + r) - i [1 - (\hat{\mathbf{d}} \cdot \hat{\mathbf{r}})]^2 \right],$$

with $r = k_0 |r_\mu - r_v|$, $\hat{\mathbf{r}} = (r_\mu - r_v) / |r_\mu - r_v|$, and $\hat{\mathbf{d}} = d / |d|$. For atoms moving with velocities $\partial_t r_\mu \ll c / k_0 |r_\mu - r_v|$, equation (3) remains valid with the time-dependent positions $r_\mu(t)$ [21]. For an energy $\omega_0$ of some eV, and velocities of the order of the order of 100 m s$^{-1}$, the above two conditions are satisfied for distances of the order of decimeter.

We consider atoms velocities such that

$$\partial_t r_\mu \ll \Gamma k_0^{-1}.$$  

In terms of the temperature $T$ of the atomic ensemble, this condition can be rewritten as $T / \Lambda \ll 1$ K where $\Lambda$ is the mass number of the atoms. Since the characteristic length scale of both $H_I$ and $\mathcal{L}$, is $k_0^{-1}$, the displacements of the atoms during a time interval of length $\Gamma^{-1}$, can be neglected in equation (3). Consequently, at each instant $t$, $\rho$ is essentially equal to the asymptotic solution of this equation with atom positions $r_\mu(t)$ assumed fixed. It is hence of the form $\rho = \sum_\mu \rho_\mu \exp(-\imath \omega t)$. Due to the small value of $\Gamma / \omega_0$, the matrices $\rho_\mu$ are practically given by their zeroth order expansions in this ratio. Thus, they obey $[H_a, \rho_\mu] = p \omega_0 \rho_\mu$, and are determined by

$$i \mathcal{L} \rho_\mu + p \delta \rho_\mu + \eta [W, \rho_{\mu+1}] + \eta [W^\dagger, \rho_{\mu-1}] = 0,$$

where $\delta = (\omega - \omega_0)/\Gamma$ is a dimensionless laser detuning, $\eta = \Omega / 2 \Gamma$ and $W = \sum_\mu \gamma_{\mu\nu} w(r_\mu \nu)^*$. Note that only the rotating wave part of $H_I$, i.e., $\Omega W \exp(i \omega t)/2 + h.c.$, appears in these equations.

As we are concerned with low laser intensities, we solve equations (7) perturbatively in the ratio $\eta = \Omega / 2 \Gamma$. We find, up to second order,

$$\rho = \left( \langle \psi | \psi \rangle^{-1} | \psi \rangle \langle \psi | \right)^{[2]},$$

where the superscript $[2]$ means that only terms up to second order are kept, and

$$| \psi \rangle = |G \rangle + \sum_\mu u_\mu | \mu \rangle + \eta^2 \sum_{\mu < \nu} (u_\mu u_\nu + v_\mu \nu) | \mu \nu \rangle,$$

see Appendix. In this expression, $|G \rangle = \otimes_\mu |g \rangle_\mu$ is the ground state of the atomic Hamiltonian $H_a$, $| \mu \rangle = \sigma_\mu^z | \mu \rangle$ and $| \mu \nu \rangle = \sigma_\mu^y | \nu \rangle$. The components $u_\mu$ and $v_\mu \nu$ obey

$$\sum_\xi (z_{\mu\xi} v_{\xi \mu} - i \delta v_{\mu \xi}) = i w_{\mu \nu},$$

$$\sum_\xi (z_{\mu\xi} v_{\xi \mu} + z_{\nu\xi} v_{\xi \mu}) - 2 i \delta v_{\mu \nu} = z_{\mu \nu} (w_{\mu \nu}^2 + w_{\nu \mu}^2),$$

where $\mu < \nu$, $w_{\mu \nu} = w(r_\mu \nu) \exp(-i \omega t)$, $v_{\mu \nu}$ equals 0 for $\mu = \nu$, $v_{\mu \nu}$ for $\mu < \nu$, and $v_{\mu \nu}$ for $\mu > \nu$. The state of atom $\mu$, which reads $\rho_\mu = |\phi \rangle_\mu \langle \phi | - \eta^2 |u_\mu^2| |g \rangle_\mu |g \rangle_\mu^* | \mu \rangle | \mu \rangle$, where $|\phi \rangle_\mu = |g \rangle_\mu + \eta u_\mu | \mu \rangle$, is determined by $u_\mu$, and the correlations between atoms $\mu$ and $\nu$ are determined by $v_{\mu \nu}$, since $\rho_{\mu \nu} - \rho_{\mu} \otimes \rho_{\nu} = \eta^2 v_{\mu \nu} \sigma_\mu \sigma_\nu + h.c.$ where $\rho_{\mu \nu}$ is the state of the pair of atoms $\mu$ and $\nu$.

The fact that $\rho$ coincides, up to second order, with a pure state, plays an essential role in the following. The origin of this effective purity can be understood as follows. The first two terms of the superoperator (4), which describe the dipole-dipole interaction between the atoms and the decay of the excited atomic levels due to spontaneous emission, can be interpreted in terms of an effective complex Hamiltonian. This is not the case of the last one, which accounts for the populating, by spontaneous emission, of $|k \rangle |l \rangle$ where $|k \rangle$ and $|l \rangle$ are any eigenstates of $H_a$, from matrix elements $\langle k' | \hat{p} | l' \rangle$ such that $\epsilon_{k'} > \epsilon_k$ where $\epsilon_k = |k| H_a |k\rangle$. However, for small $\eta$, and in the
long time regime, this process essentially does not contribute to \( \rho \), since the order, in \( \eta \), of \( \langle k | \rho | l \rangle \) increases with \( \epsilon_k \). Up to second order, it only results in a correction to the ground state population \( \langle G | \rho | G \rangle \), which simply ensures the normalisation of \( \rho \). Due to the decline of \( \langle k | \rho | l \rangle \) with increasing \( \epsilon_k \), stimulated emission is also negligible. Consequently, equations (7) can be replaced by the Schrödinger-like equation

\[
\partial_t |\psi\rangle = -\left[iH_a + \sum_{\mu,\nu} z_{\mu\nu}\sigma^\dagger_\mu\sigma_\nu - i\frac{\Omega}{2}e^{-i\omega t}W^\dagger\right]|\psi\rangle,
\]
(11)

where the last term describes laser photon absorption. This equation is satisfied, up to second order, by a state of the form (9), provided \( u_\mu \) and \( v_{\mu\nu} \) fulfill eq. (10).

An important property of the state (8) is that the ensuing state of any subensemble of atoms, is given by an expression of the same form. Consider the system \( S \) consisting of the atoms \( \mu = 1, \ldots, n \), and the complementary system \( \overline{S} \) consisting of all the other atoms. The pure state (9) can be expanded on the basis \( \{|G\rangle, |\sigma\rangle, |\psi\rangle, \ldots \} \) of system \( \overline{S} \), where \( |G\rangle = \otimes_{\nu>n}|\nu\rangle \), as \( |\psi\rangle = |G\rangle|\psi\rangle_{S} + \ldots \). The state \( |\psi\rangle_{S} \) is given by expression (9), but with sums running only over the first \( n \) atoms, and \( |G\rangle, |\mu\rangle \) and \( |\mu\nu\rangle \), replaced by the corresponding states for system \( S \). The point is that the following terms in the above expansion of \( |\psi\rangle \), either do not contribute to the second-order state \( \rho_S \) of system \( S \), or contribute simply to a correction to the population \( \langle G | \rho_S | G \rangle \), or system \( S \), in the ground state population \( \langle G | \rho_S | G \rangle \). Consequently, \( \rho_S \) is given by eq. (8) with \( |\psi\rangle \) replaced by \( |\psi\rangle_{S} \).

We now show that, as a consequence of the above obtained results, any two subgroups of atoms, say \( A \) and \( B \), are entangled for low enough laser intensities. A sufficient, but in general not necessary, condition for \( A \) and \( B \) to be entangled, is that the partial transpose \( \rho^T_{AB} \) of their collective state \( \rho_{AB} \), has negative eigenvalues \( (22) \). Using expression (8), the eigenvalues of \( \rho^T_{AB} \) can be determined up to second order in \( \eta \). Since all the atoms are in their ground state for \( \eta \to 0 \), one of them goes to 1 in this limit. The others, denoted \( \lambda_q \) in the following, are at least of second order. It can be shown that the lowest order contribution to \( \lambda_q \) is an eigenvalue of \( \eta^2(V + V^\dagger) \)

\[
V = \sum_{\mu \leq n_A < \nu \leq n_A + n_B} v_{\mu\nu} |\mu\rangle_{AB} \langle \nu|, \tag{12}
\]

where \( n_A \) and \( n_B \) are the numbers of atoms, suitably numbered, of systems \( A \) and \( B \), respectively, and \( |\mu\rangle = \sigma^\dagger_\mu \otimes_{\nu \leq n_A + n_B} |\nu\rangle \), see Appendix. The eigenvalues of the Hermitian operator \( V + V^\dagger \) are real. Since it is traceless, some of them are negative as soon as \( V \neq 0 \) \( (23) \). Thus, for small \( \eta \), \( A \) and \( B \) are either uncorrelated or entangled. This is similar to the pure state case, and results from the fact that, up to second order, \( \rho_{AB} \) coincides with a pure state, as discussed above.

As seen above, any two subgroups of atoms, are generically entangled for small enough \( \eta \). The opposite limit, \( \Omega \gg \Gamma \), corresponds to the saturation regime, where \( p_{AB} \) is proportional to the identity matrix, and hence \( A \) and \( B \) are uncorrelated. Thus, there is a particular value of \( \eta \), that depends on \( A \) and \( B \), where \( p_{AB} \) goes from entangled to separable. In the general case, determining this value requires solving equations (7) for finite \( \eta \), which is not straightforward, even for only two atoms \( (8) \). Moreover, for more than two atoms, there is no simple necessary and sufficient condition for entanglement \( (22) \). However, for atoms separated by distances much larger than \( k_0^{-1} \), which is of the order of 0.1\( \mu m \) for \( \omega_0 \) of some eV, a laser intensity threshold below which \( A \) and \( B \) are certainly entangled, can be evaluated. In this dilute regime, equations (10) can be solved perturbatively in the coefficients \( z_{\mu\nu} \approx k_0^{-1}|\mathbf{r}_\mu - \mathbf{r}_\nu|^{-1} \) with \( \mu \neq \nu \). This leads to the dominant contribution

\[
v_{\mu\nu} = -4z_{\mu\nu}(1 - 2\delta)^{-3}(w^2_\mu + w^2_\nu), \tag{13}
\]

to the matrix elements of operator (12). Note that the correlations between atoms \( \mu \) and \( \nu \) are then the same in the presence and absence of the other atoms. The eigenvalue \( \lambda_q \) expands, in powers of \( \eta \), as \( \lambda_q = \eta^2 \lambda_q^{(2)} + \eta^4 \lambda_q^{(4)} + \ldots \), where \( \lambda_q^{(2)} \) is an eigenvalue of \( V + V^\dagger \). For negative \( \lambda_q^{(2)} \), \( \lambda_q \) changes sign for a certain value of \( \Omega \), which we denote by \( \Omega_q \). Since the matrix elements of \( V \) are given by eq. (13), \( \lambda_q^{(2)} \) is small in the dilute regime considered here. On the contrary, \( \lambda_q^{(4)} \) attains a finite value in the limit of vanishing \( z_{\mu\nu} \). We find the positive asymptotic value

\[
\lambda_q^{(4)} = (1/4 + \delta^2)^{-2} \sum_{\mu \leq n_{AB}} |w_\mu|^4 |\langle \mu|\varphi_q\rangle|^2, \tag{14}
\]

where \( n_{AB} = n_A + n_B \), and \( |\varphi_q\rangle \) is the eigenstate of \( V + V^\dagger \) corresponding to \( \lambda_q^{(2)} \), see Appendix. This leads, for negative \( \lambda_q^{(2)} \), to \( \Omega_q \simeq \Gamma |\lambda_q^{(2)}|/|\lambda_q^{(4)}|^{1/2} \). As long as \( \Omega < \max_{q}\Omega_q \), at least one eigenvalue \( \lambda_q \) is negative, and hence \( A \) and \( B \) are necessarily entangled.

To study more quantitatively long-range entanglement, we consider two regions of characteristic size \( L \), separated by a large distance \( D \gg k_0L^2 \), and assume the laser beam is a plane wave of wave vector \( \mathbf{K} \). Systems \( A \) and \( B \) consist of the atoms lying in these regions. In this case, equations (5), (12), and (13), give

\[
V = \frac{3i\sin^2\theta e^{ik_0D}}{k_0D(1 - 2\delta)^{-1}} \sum_{\mu \leq n_A < \nu \leq n_A + n_B} |\tilde{\mu}\rangle \langle \tilde{\nu}|(w^2_\mu + w^2_\nu), \tag{15}
\]

where \( w_\mu = \exp(i\mathbf{K} \cdot \mathbf{r}_\mu - i\omega t) \), \( \theta \) is the angle between \( \mathbf{d} \) and the approximate line joining \( A \) and \( B \), and \( |\tilde{\mu}\rangle = \exp(-ik_0e \cdot \mathbf{r}_\mu)|\mu\rangle_{AB} \) with \( e \) the unit vector pointing from \( A \) to \( B \). Noting that this operator can be written in terms of four kets, one finds two negative eigenvalues \( \lambda_q^{(2)} \), see
Appendix. For randomly distributed atoms and large enough numbers \(n_A\) and \(n_B\), these two negative \(\lambda_q^{(2)}\) are essentially equal. Since \(|w_\mu| = 1\) for all the atoms of A and B, the sum in expression \(14\) reduces to 1. Finally, A and B are necessarily entangled for

\[
\Omega < \frac{\sqrt{3}}{4} \Gamma(1 + 4\delta^2)^{1/4} \left( \frac{D_0}{D} \right)^{1/2}, \tag{16}
\]

where \(D_0 = k_0^{-1}(n_A n_B)^{1/2} \sin^2 \theta\). A measure of the entanglement between A and B is the negativity, which is the absolute sum of the negative \(\lambda_q\) \[22\]. It reaches its maximum

\[
N_{\text{max}} = \frac{9}{32} (1 + 4\delta^2)^{-1} \left( \frac{D_0}{D} \right)^2, \tag{17}
\]

for \(\Omega\) equal to the right side of inequality \(16\) divided by two. As the distance between systems A and B increases, the interval of laser amplitudes that lead to non-zero negativity, shrinks, and the maximum attainable negativity \(N_{\text{max}}\) diminishes. However, since \(D\) appears in equations \(16\) and \(17\), divided by \(D_0\), the unfavorable impact of increasing the distance can be counterbalanced by enlarging the numbers \(n_A\) and \(n_B\). For example, the same negativity can be reached, for any distance \(D\), by adjusting appropriately these numbers and the laser intensity. Another interesting consequence of the dependence on the numbers \(n_A\) and \(n_B\) is the following. All the other parameters, including \(D\) and the laser amplitude \(\Omega\), being fixed, eq. \(16\) shows that big enough groups of atoms, separated by the large distance \(D\), are necessarily entangled. Let us finally discuss the influence of the laser detuning. As \(\delta\) is increased, the laser intensity threshold given by eq. \(16\), grows. However, our resonant approach, based on eq. \(7\), is valid only for not too large \(\delta\). Moreover, the reachable values of negativity vanish with increasing \(\delta\), see eq. \(17\).

In summary, we have shown that two groups of two-level atoms, A and B, can be entangled by a weak resonant laser field, even if the distance between them is macroscopic, and even in the presence of surrounding atoms. A laser amplitude below which A and B are certainly entangled, and the maximum attainable value of negativity, have been derived, in a dilute regime, for far separated A and B, in a plane wave laser beam. It has been found that both these threshold amplitude and maximal negativity diminish with increasing distance between A and B. But these tendencies can be counterbalanced by enlarging the sizes of A and B. In this work, we assumed that the motion of the atoms is slow enough that its impact on the dynamics of their internal state can be disregarded, which, depending on the atomic mass, can be valid for temperatures of the order of 10 K. A natural extension of our study would be to examine how the found laser-induced entanglement depends on the atoms velocities for higher temperatures, and whether it disappears at some temperature. The quantitative results presented for very distant systems A and B have been derived in the dilute regime. It would be of interest to determine how general they are, especially the positive impact of enlarging the number of considered atoms. We finally remark that, though we focus on atoms in this paper, the studied entanglement mechanism may be relevant to other physical realizations of qubits, such as nuclear spins, coupled to a common environment, and to oscillating fields.

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[23] The non-zero eigenvalues of \(V + V^\dagger\) are \(\pm \Lambda_q^{1/2}\) where \(\Lambda_q\) are the non-zero eigenvalues of both positive operators \(VV^\dagger\) and \(V^\dagger V\).
APPENDIX

In the absence of the laser field, the atoms steady state is the ground state, i.e., \( \rho_p^{(0)} = \delta_{\rho_0} |G\rangle\langle G| \). To solve equations (7) for small \( \eta \), we expand the Fourier components \( \rho_p \) as \( \rho_p = \rho_p^{(0)} + \eta \rho_p^{(1)} + \ldots \), and rewrite these equations as \( (i \mathcal{L} + \gamma \rho_p^{(n+1)}) = -[W, \rho_p^{(n)}] - [W^\dagger, \rho_p^{(n)}] \). This recursive relation gives

\[
\rho_p^{(1)} = \delta_{\rho_1} \sum_\mu \hat{u}_\mu |\mu\rangle \langle \mu| + \delta_{\rho_{-1}} \sum_\mu \hat{u}_{\mu}^* |\mu\rangle \langle \mu|, \tag{18}
\]

where the components \( \hat{u}_\mu \) obey eq. (10) with \( t = 0 \). In deriving this result, we used the fact that the matrix elements \( \langle \mu |\rho_{\pm}\rangle \langle \nu | \) are at most of second order. They are actually of third order. Similarly, we find

\[
\rho_p^{(2)} = \delta_{\rho_2} \left( -\sum_\mu |u_{\mu}|^2 |G\rangle\langle G| + \sum_{\mu,\nu} \hat{u}_{\mu} \hat{u}_{\nu}^* |\mu\rangle \langle \nu| \right) + \delta_{\rho_{-2}} \sum_{\mu,\nu} s_{\mu\nu} |\mu\rangle \langle \nu|, \tag{19}
\]

where the components \( s_{\mu\nu} \) obey \( \sum_\xi (s_{\mu}(\xi \mu + z_{\xi \mu} \xi \mu) - 2i\delta_{\xi\mu} = \hat{u}_{\mu}^* |\nu\rangle \langle \mu| = 0 \). Using the first equality of eq. (11), it can be shown that \( v_{\mu\nu} = (s_{\mu\nu} - \hat{u}_{\mu}^* |\nu\rangle \langle \mu| \) satisfies the second equality of eq. (11). Finally, the atoms state \( \rho = \sum_\rho \rho_p \exp(-i\eta \omega t) \) can be written, up to second order, under the form (8).

From the above expressions, it follows that the Fourier components of the state \( \rho_{AB} \) of A and B, are given, up to second order, by eq. (13) and eq. (19) with sums running only over the atoms \( \mu \leq n_{AB} = n_A + n_B \). Consequently, the only non-vanishing matrix elements of its second-order partial transpose are

\[
|G\rangle \langle \rho_{AB}^{(2)}|G\rangle = 1 - \eta^2 \sum_{\mu \leq n_{AB}} |u_\mu|^2, \tag{20}
\]

\[|G\rangle \langle \rho_{AB}^{(2)}|\mu\rangle = \eta u_\mu \quad \text{for} \quad \mu \leq n_A, \tag{21}\]

\[|\mu\rangle \langle \rho_{AB}^{(2)}|\nu\rangle = \eta^2 u_\mu u_{\nu}^* \quad \text{for} \quad \nu \leq n_A, \quad \mu < \nu, \tag{22}\]

\[|G\rangle \langle \rho_{AB}^{(2)}|\mu\nu\rangle = \eta^2 (u_\mu u_{\nu} + v_{\mu\nu}^*) \quad \text{for} \quad \mu < \eta \leq n_A, \tag{23}\]

\[|\mu\rangle \langle \rho_{AB}^{(2)}|G\rangle = |G\rangle \langle \rho_{AB}^{(2)}|\mu\rangle^*, \quad \text{and} \quad |\mu\nu\rangle \langle \rho_{AB}^{(2)}|G\rangle = |G\rangle \langle \rho_{AB}^{(2)}|\mu\nu\rangle^*, \tag{24}\]

understood here as \( |G\rangle = \otimes_{\xi \leq n_{AB}} |\xi\rangle \), \( |\mu\rangle = \sigma_\mu^G |G\rangle \) and \( |\mu\nu\rangle = \sigma_\mu^G \sigma_\nu^G |G\rangle \), with \( \mu < \nu \leq n_{AB} \). Writing \( \rho_{AB}^{F}(\varphi_q) \) and expanding both \( |\varphi_q\rangle \) and \( \lambda_q \) in powers of \( \eta \), with \( \lambda_q^{(0)} = 0 \), lead to \( \lambda_q^{(1)} = 0 \), and \( (V + V^\dagger)|\varphi_q\rangle^{(0)} = \lambda_q^{(2)}|\varphi_q\rangle^{(0)} \), where the operator \( V \) is given by eq. (12).

Pursuing this perturbation calculation to higher orders, we find, after a lengthy but straightforward derivation, \( \lambda_q^{(3)} = 0 \), and

\[\lambda_q^{(4)} = \langle \varphi_q | (\rho_{AB}^F)^{(4)} | \varphi_q \rangle - |\tau_q^G|^2 (\lambda_q^{(2)} + \sum_{\mu} |u_\mu|^2) \tag{24}\]

\[+ \lambda_q^{(2)} \sum_{\mu < \nu} |\tau_{\mu\nu}|^2 + 2 \text{Re} (\tau_q^G \langle \varphi_q | (\rho_{AB}^F)^{(3)} | G \rangle), \tag{25}\]

where \( |\varphi_q\rangle = |\varphi_q\rangle^{(0)} \), and \( \tau_q^k = \langle k | \varphi_q \rangle^{(1)} \). The only component of \( |\varphi_q\rangle^{(1)} \) required for our purpose, is \( \tau_q^G = -\sum \hat{u}_\mu^* |\mu\rangle \langle \varphi_q| \), where \( \hat{u}_\mu \) is \( u_\mu \) for \( \mu \leq n_A \), and \( u_\mu \) for \( \mu > n_A \). We are concerned with the value of \( \lambda_q^{(4)} \) in the limit of infinitely distant atoms, in which \( \rho_{AB} \) converges to the uncorrelated state \( \rho_{AB}^{(0)} = \rho_{\leq n_A} \rho_{\leq n_B} \) where

\[\rho_{\leq n_A} = (1 - p_\mu) \sigma_\mu^G \sigma_\mu^G + p_\mu \sigma_\mu^G \sigma_\mu^G + c_\mu \sigma_\mu^G + c_\mu^* \sigma_\mu^G, \tag{25}\]

The components of the above kets are \( |\mu| \langle \phi_{+} | + | \phi_{+} | \langle \phi_{-} | \tag{26}\]

The last result can be derived more directly as follows. The conditions \( s_A, s_B \leq 1 \) mean \( \varphi_{\pm} \varphi'_{\pm} \approx 0 \). In this case, it is immediate to see that the non-zero eigenvalues of both \( V + V^\dagger \) and \( V \), which are the squares of the non-zero eigenvalues of \( V + V^\dagger \) 25, are \( \langle \phi_{\pm} | \phi_{\pm} | \langle \phi'_{\pm} | \phi'_{\pm} | = y \).