Dynamics of entanglement between two free atoms with quantized motion

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Abstract

The electronic entanglement between two free atoms initially at rest is obtained including the effects of photon recoil, for the case when quantum dispersion can be neglected during the atomic excited-state lifetime. Unlike previous treatments using common or statistically independent reservoirs, a continuous transition between these limits is observed, which depends on the inter-atomic distance and degree of localization. The occurrence of entanglement sudden death and birth as predicted here deviates from the case where the inter-atomic distance is treated classically by a static value. Moreover, the creation of a dark state is predicted, which manifests itself by a stationary entanglement that even may be created from an initially separable state.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

If a quantum system is brought into contact with another system with much larger mode space, the system suffers decoherence [1], which in the Born–Markov approximation results in an exponential decay of its coherences. Within this paradigm, one may conclude that entanglement [2] should also naturally suffer a degradation with a similar behaviour: an asymptotic decay to zero. However, recently it has been shown that many exceptions from this expected behaviour exist, that show a vanishing of entanglement at finite time, including its possible reappearance at later times. These features have been denoted as entanglement sudden death (ESD) and birth (ESB), respectively [3–9].

The occurrence of ESD and/or ESB has created increasing interest in studying the dynamics of entanglement of various types of bipartite systems, where the concurrence [10] can be used as a measure of entanglement. These systems include, among others, systems of continuous variables [11, 12], atoms in cavities [7, 13–15], cavity fields [16, 17], spin chains [18, 19] and atoms in free space [20–23]. Also non-Markovian effects have been studied for the dynamics of bipartite entanglement [24–26]. Furthermore, experiments have been performed to model [27] or directly measure [28] the dynamics of entanglement.

To the best of our knowledge, at present, theoretical analysis has been performed only within the framework of a master equation in the Born–Markov approximation, where the atoms are supposed to have classical and fixed positions [29, 30]. In such a treatment [21] the dynamics of the concurrence has been shown to reveal in general two different time scales, one given by the spontaneous decay rate and the other by a distance-dependent collective decay rate. Furthermore, the existence of a sequence of more than one ESD and one ESB has been stated. In the limit of infinite inter-atomic distance, this model reproduces the well-known results for two spin systems in statistically independent reservoirs [7]. However, in such a treatment the influence of the relative quantized motion of the atoms is discarded.

Physically, this may only be realized by situating the two atoms in independent traps and in the limit where the trap frequency is $\omega \gg E_r/\hbar$, where $E_r$ is the recoil energy of the atoms. In this Lamb–Dicke limit the photon recoil is received by the trapping mechanism (Mössbauer effect) and the atomic motion is unaffected. In all other cases, the photon recoil affects the motion of the two atoms and in this way the dynamics of the electronic entanglement between the atoms may be modified. This is most relevant in the limit of free atoms. There the relative quantized motion has to be included, in order to allow the distinguishability between the atoms for properly addressing the issue of their mutual entanglement [23]. We will briefly review the arguments given in [23].

Given two identical two-level atoms, initially at rest and distinguishable by their differing positions, their
distinguishability should be maintained at least during the atoms’ excited-state lifetime $\tau_0 = 2\pi/\gamma_0$, $\gamma_0$ being the natural linewidth of the electronic transition. This condition requires that the quantum dispersion of the relative-position wave-packet be sufficiently weak, so that during $\tau_0$ the rms spread is

$$\Delta r \ll \bar{r}, \quad (1)$$

where $\bar{r}$ is the mean inter-atomic distance. Due to quantum dispersion, the initial rms spread $\Delta r_i$ is enlarged during the interval $\tau_0$ to

$$\Delta r_i = \Delta r_i \sqrt{1 + \left(\frac{l_d}{\Delta r_i}\right)^4}, \quad (2)$$

where the dispersion length is $l_d = \sqrt{\hbar/\gamma_0 m}$ with $\hbar$ being Planck’s constant and $m$ the atomic mass.

Equation (2) together with condition (1) leads to the condition

$$\bar{r} \gg \Delta r_i \gg \lambda_0, \sqrt{E_i/\hbar\gamma_0}, \quad (3)$$

where $\lambda_0$ is the wavelength of the electronic transition and the recoil energy is $E_i = (\hbar k_0^2/2m)$ with $k_0 = 2\pi/\lambda_0$. As typically $E_i \ll \hbar\gamma_0$, the minimum distance and rms spread may still be much smaller than $\lambda_0$. However, a zero rms spread is not permitted, as then quantum dispersion would rapidly render the atoms indistinguishable. Thus, to be consistent with the requirement of distinguishability during $\tau_0$, a finite initial spread $\Delta r_i$ within the limits (3) is required.

In a realistic experiment, the atoms cannot be treated as point particles but as quantum wavepackets with minimum allowed widths, as shown above; a quantized description of the atomic motion is required. Furthermore, as the atoms may freely move, the photon recoil associated with spontaneous emissions will generate non-vanishing atomic velocities for atoms that initially were at rest. As the quantized motion is correlated to photon emissions, it is indirectly correlated to electronic-state changes. Thus, the quantized motion represents a subsystem that may be correlated with the electronic state of the atoms. Therefore, to obtain the reduced electronic dynamics and its entanglement it is necessary to include the inter-atomic motion in its quantized form. In consequence, in this paper we present a fully quantized treatment of both the electronic and relative motion. By use of a Wigner–Weisskopf approach it is shown how the dynamics of the entanglement between the atoms is affected by the photon recoil during spontaneous emissions. The present work extends our previous results in [23], where two atoms with initially only one excitation were considered, to the more complex case of initially two excitations.

The outline of the paper is as follows. In section 2, the interaction between the two atoms and the electromagnetic (em) field is treated in the Wigner–Weisskopf approach and the dynamics of the relevant probability amplitudes is solved. Using these solutions, in section 3 the electronic reduced density matrix is obtained and then used in section 4 to derive the concurrence as a measure of entanglement. Finally, in section 5, a summary and conclusions are given.

2. Dynamics of two two-level atoms interacting with the electromagnetic field

2.1. Hamilton operator

The system consists of two atoms of mass $m$ and with inter-atomic distance vector $\bar{r}$ and corresponding relative momentum $\hat{p}$. Each atom is composed of two electronic levels with the transition frequency $\omega_{0a}$ described by the atomic pseudo-spin operator $\hat{S}_a$ with $a = \pm 1$ being the index for the two atoms. Both atoms interact with the em field via their electronic transition dipole moment $\bar{d}$.

The complete Hamilton operator for this system is given by

$$\hat{H} = \int d^3k \sum_{\sigma} \hbar \epsilon_{\bar{k},\sigma} \hat{a}_{\bar{k},\sigma}^\dagger \hat{a}_{\bar{k},\sigma} + \frac{\hbar}{2} \bar{d} \cdot \bar{e}_{\bar{k},\sigma} E_{\bar{k}} + \hbar \text{A.}, \quad (4)$$

Here $\hat{a}_{\bar{k},\sigma}$ is the annihilation operator for a photon in the plane-wave mode defined by the wave vector $\bar{k}$ and the polarization $\sigma$. The interaction between atoms and em field reads

$$\bar{V} = \int d^3k \sum_{\sigma} \hbar \lambda_{\bar{k},\sigma} \hat{S}_{a,\sigma} \hat{a}_{\bar{k},\sigma} e^{-i\bar{k} \cdot \bar{r}}/2 + \text{H.a.}, \quad (5)$$

where the atom–photon coupling strength is given by the rate

$$\lambda_{\bar{k},\sigma} = \bar{d} \cdot \bar{e}_{\bar{k},\sigma} E_{\bar{k}}/\hbar, \quad (6)$$

with $\bar{e}_{\bar{k},\sigma}$ being the polarization unit vector and with the vacuum electric-field strength

$$E_{\bar{k}} = \sqrt{\hbar\epsilon_{\bar{k}}/16\pi^3\epsilon_0}. \quad (7)$$

2.2. Equations of motion for the probability amplitudes

Assuming an initial vacuum state for the em field and an initial electronic state of the atoms consisting of a superposition of only $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$, the general form of the wavefunction for the system is given by

$$|\Psi(t)\rangle = \int d^3r \langle\bar{r}\rangle_{\text{rel}} \otimes \left\{ \psi_{\bar{k},\sigma}(\bar{r}, t)|\bar{g}\rangle + \psi_{\bar{k},\sigma}(\bar{r}, t)|\bar{e}\rangle \right\}$$

$$+ \int d^3k \sum_{\sigma} \sum_{a=\pm} \psi_{a,\bar{k},\sigma}(\bar{r}, t)|a; \bar{k}, \sigma\rangle$$

$$+ \int d^3k \int d^3k' \sum_{\sigma,\sigma'} \psi_{\bar{k},\sigma,\bar{k}',\sigma'}(\bar{r}, t)|\bar{k}, \sigma; \bar{k}', \sigma'\rangle \}. \quad (8)$$

Here the electronic–photonic states are defined as

$$|\bar{g}\rangle = |\downarrow\downarrow\rangle_{\text{el}} \otimes |\text{vac}_{\text{em}}\rangle, \quad (9)$$

$$|\bar{e}\rangle = |\uparrow\uparrow\rangle_{\text{el}} \otimes |\text{vac}_{\text{em}}\rangle, \quad (10)$$

$$|a; \bar{k}, \sigma\rangle = |\bar{k}, \sigma\rangle_{\text{el}} \otimes \hat{a}_{\bar{k},\sigma}^\dagger |\text{vac}_{\text{em}}\rangle, \quad (11)$$

$$|\bar{k}, \sigma; \bar{k}', \sigma'\rangle = |\bar{k}, \sigma\rangle_{\text{el}} \otimes \hat{a}_{\bar{k}',\sigma'}^\dagger |\text{vac}_{\text{em}}\rangle, \quad (12)$$

$$|\bar{k}, \sigma; \bar{k}', \sigma'\rangle = |\downarrow\downarrow\rangle_{\text{el}} \otimes \hat{a}_{\bar{k},\sigma}^\dagger \hat{a}_{\bar{k}',\sigma'}^\dagger |\text{vac}_{\text{em}}\rangle, \quad (13)$$

where the state (13) is not normalized to unity in order to contain the bosonic enhancement when two identical photons are emitted.
With the general form (8) the Laplace-transformed Schrödinger equation,
\[ i\hbar x |\Psi(r)(s)\rangle = i\hbar |\Phi\rangle + \hat{H}|\Psi(r)(s)\rangle, \tag{14} \]
has to be solved with the initial condition \(|\Psi(r(0))\rangle = |\Phi\rangle\).

The initial state \(|\Phi\rangle\) is analogously expanded as equation (8) with the initial probability amplitudes being \(\phi_s(r)\), \(\phi_e(r)\), whereas \(\phi_{a,k,s}(r) = \phi_{a,k,e,s}(r) = 0\) because of the initial absence of photons. The corresponding Laplace-transformed coupled equations for the probability amplitudes then read
\[
(i\hbar + \omega_0)\tilde{\psi}_a(r, s) = i\phi_a(r), \tag{15}
\]
\[
(i\hbar - \omega_0)\tilde{\psi}_e(r, s) = i\phi_e(r)
+ \int d^3k \sum_{\alpha} \kappa_{a,s} \tilde{\psi}_{a,k,s}(r, s) e^{i\vec{k}\cdot\vec{r}/2}, \tag{16}
\]
\[
(i\hbar - c\kappa)\tilde{\psi}_{a,k,s}(r, s)
= \kappa_{a,s}^* \tilde{\psi}_a(r, s) e^{-i\vec{k}\cdot\vec{r}/2}
+ \int d^3k' \sum_{\alpha'} \kappa_{a,s} \tilde{\psi}_{a,k,s}(r, s) e^{-i\vec{k}'\cdot\vec{r}/2}
\times [\tilde{\psi}_{a,k,s}(r, s) + \tilde{\psi}_{a,k,s}(r, s)] e^{-i\vec{k}'\cdot\vec{r}/2}. \tag{17}
\]

Here it was assumed that the resulting spectral width of the electronic resonances is much larger than the Doppler shifts induced by the slow relative motion. Under this assumption the kinetic energy of this relative motion can be safely neglected.

Note that with the appearance of the state \(|\tilde{r}\rangle\rangle\) in equation (8), a full quantized treatment of the relative distance between the atoms is warranted and included by the use of the correspondingly defined amplitudes. As a consequence, the neglectation of the kinetic energy in the equations of motion for the amplitudes, cf equations (15)–(18), is equivalent to the Raman–Nath approximation in atomic-beam diffraction. There the kinetic energy is neglected, but the recoil still generates orders of diffraction of the passing atoms which correspond to dynamical changes of their atomic momenta.

2.3. Solution of the probability amplitudes

From equation (15) it can be readily seen that the ground-state amplitudes have the simple solution
\[ \psi_a(r, t) = \phi_a(r) e^{-i\omega_0 t}. \tag{19} \]

To obtain the solution for the excited-state amplitude \(\tilde{\psi}_e\), we first insert equation (18) into equation (17) to obtain
\[
(i\hbar - c\kappa)\tilde{\psi}_{e,k,s}(r, s)
= \kappa_{e,s}^* \tilde{\psi}_e(r, s) e^{-i\vec{k}\cdot\vec{r}/2}
+ \int d^3k' \sum_{\alpha'} \frac{2|\kappa_{e,s}|^2}{i\hbar + \omega_0 - c(\vec{k} + \vec{k}')}
\times [\tilde{\psi}_{a,k,s}(r, s) + \tilde{\psi}_{a,k,s}(r, s)] e^{-i\vec{k}'\cdot\vec{r}}. \tag{20}
\]
Assuming now the form
\[ \tilde{\psi}_{a,k,s}(r, s) = \tilde{\psi}_e(r, s) \chi_{a,k,s}(r, s), \tag{21} \]
and assuming that we may expand the required solution \(\chi_{a,k,s}\) in powers of the atom–photon interaction rate \(\kappa_{e,s}\),
\[ \chi_{a,k,s}(r, s) = \kappa_{e,s}^* e^{-i\vec{k}\cdot\vec{r}/2}
\times \left[ \chi^{(1)}_{a,k,s}(r, s) + |\kappa_{e,s}|^2 \chi^{(3)}_{a,k,s}(r, s) + \cdots \right]. \tag{22} \]
we obtain from the first-order terms of equation (20) the condition
\[ \chi^{(1)}_{a,k,s}(r, s) = \frac{1}{i\hbar + \omega_0 - c\kappa}, \tag{23} \]
and likewise from the third-order terms the condition
\[ |\kappa_{e,s}|^2 \chi^{(3)}_{a,k,s}(r, s) = 2 \chi^{(1)}_{a,k,s}(r, s) \tag{24} \]
and assuming that we may expand the required solution \(\chi_{a,k,s}\) in powers of the atom–photon interaction rate \(\kappa_{e,s}\),
\[ \chi_{a,k,s}(r, s) = \kappa_{e,s}^* e^{-i\vec{k}\cdot\vec{r}/2}
\times \left[ \chi^{(1)}_{a,k,s}(r, s) + |\kappa_{e,s}|^2 \chi^{(3)}_{a,k,s}(r, s) + \cdots \right]. \tag{22} \]

Inserting then equation (21) into equation (16) and using expansion (22), the excited state amplitude is obtained as
\[ \tilde{\psi}_e(r, s) = f(r, s) \phi_e(r), \tag{25} \]
where the function is defined as
\[ f(r, s) = \frac{1}{s + i\hbar + \omega_0 + \Omega(r, s)}, \tag{26} \]
with the complex-valued frequency shift being
\[ \Omega(r, s) = \Omega^{(1)}(r, s) + \Omega^{(3)}(r, s) + \cdots. \tag{27} \]

The first-order term of this frequency shift results from \(\chi^{(1)}\) and reads
\[ \Omega^{(1)}(r, s) = 2 \int d^3k \sum_{\sigma} \frac{|\kappa_{e,s}|^2}{i\hbar - c\kappa}, \tag{28} \]
whereas the third-order term results from \(\chi^{(3)}\) as
\[ \Omega^{(3)}(r, s) = 4 \int d^3k \sum_{\sigma} \frac{|\kappa_{e,s}|^2}{i\hbar - c\kappa}
\times \int d^3k' \sum_{\sigma'} \frac{|\kappa_{e,s}|^2}{i\hbar - c\kappa - c\kappa'}
\times \left[ \frac{1}{i\hbar - c\kappa} + \frac{1}{i\hbar - c\kappa'} \right] [1 + \cos(\vec{k} - \vec{k}') \cdot \vec{r}]. \tag{29} \]

We note that each occurrence of the factor \(|\kappa_{e,s}|^2\) in equations (28) and (29) will result as the natural line width \(\gamma_0\) of the atomic electronic transition, which is defined as
\[ \gamma_0 = \frac{\hbar^2}{2\Lambda_0^3}, \tag{30} \]
Therefore, \( \Omega^{(1)}(\vec{r}, s) \propto \gamma_0 \) and \( \Omega^{(3)}(\vec{r}, s) \propto \gamma_0^2 \), where \( \gamma_0 \ll \omega_0 \), so that the pole in equation (26) is located very near to \( s = -i\omega_0 \) and \( \Omega \) represents a relative displacement of this pole of the order of \( \varepsilon = \gamma_0/\omega_0 \ll 1 \). Thus, we may approximate this frequency shift by taking the limiting value

\[
\Omega(\vec{r}) \approx \lim_{\omega \to \omega_0} \Omega(\vec{r}, s) = \lim_{\omega \to \omega_0} \Omega^{(1)}(\vec{r}, s) + \mathcal{O}(\gamma_0^2).
\]  

(31)

The limiting value is obtained by evaluating the residues of expression (28) as \( \lim_{\omega \to \omega_0} \Omega^{(1)}(\vec{r}, s) = -i\gamma_0 \), so that we may use the approximation

\[
\Omega \approx -i\gamma_0.
\]  

(32)

In the time domain, the function \( f(\vec{r}, t) \) can thus be approximated as

\[
f(\vec{r}, t) \approx f(t) = \exp[-(i\omega_0 + \gamma_0)t],
\]

(33)

being independent of the inter-atomic distance vector \( \vec{r} \). Correspondingly the amplitude (21) can be consistently approximated as

\[
\varphi_{a,k,\sigma}(\vec{r}, s) = \frac{\kappa_{a,k,\sigma}^2}{i\omega - ck} \int \psi(\vec{r}).
\]

(34)

Equations (19), (25) and (34) together with the approximation (33) constitute the required information to evaluate the reduced electronic density operator of the system.

3. Electronic density matrix of the two atoms

3.1. Reduced electronic density matrix

The reduced density operator of the electronic systems of the two atoms reads

\[
\hat{\rho}(t) = \int d^3r \, \text{Tr}_{\text{ee}}(\hat{\rho} \, |\psi(t)\rangle \langle \psi(t)|\hat{\rho}).
\]

(35)

In the standard basis \(|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle\rangle\), it can be written as the reduced density matrix matrix

\[
\rho(t) = \begin{pmatrix}
\rho_{e,e} & 0 & 0 & 0 \\
0 & \sigma_{e,+} & \sigma_{e,-} & 0 \\
0 & \sigma_{e,-} & \sigma_{e,+} & 0 \\
0 & 0 & 0 & \rho_{g,g}
\end{pmatrix},
\]

(36)

where the matrix elements are given by

\[
\rho_{e,e}(t) = \int d^3r \left| \psi_e(\vec{r}, t) \right|^2,
\]

(37)

\[
\rho_{e,g}(t) = \int d^3r \int d^3k \sum_{\sigma} \psi_e^{*}(\vec{r}, s) \psi_g(\vec{r}, t),
\]

(38)

\[
\sigma_{a,b}(t) = \int d^3r \int d^3k \sum_{\sigma} \psi_{a,k,\sigma}(\vec{r}, t) \psi_{b,k,\sigma}^{*}(\vec{r}, t),
\]

(39)

and due to the unit trace,

\[
\rho_{e,g}(t) = 1 - \rho_{e,e}(t) - \sum_{a=\pm} \sigma_{a,a}(t).
\]

(40)

This latter definition of \( \rho_{e,g}(t) \) ensures consistency with the approximations performed for the other density matrix elements. Furthermore, equation (36) represents a physical density matrix because its Kraus decomposition exists, as shown in the appendix.

For the evaluation of these matrix elements we will assume initial conditions where the relative motion of the atoms is factorized from the electronic state, i.e.

\[
|\phi_e(\vec{r})|^2 = \rho_{e,e}(0) w(\vec{r}),
\]

(41)

\[
|\phi_g(\vec{r})|^2 = \rho_{e,g}(0) w(\vec{r}),
\]

(42)

\[
\phi_e(\vec{r}) \phi_g^{*}(\vec{r}) = \rho_{e,g}(0) w(\vec{r}),
\]

(43)

where \( w(\vec{r}) \) is the probability density for the inter-atomic distance vector \( \vec{r} \).

Given the approximation (33), the solutions for the excited-state population and of the outer off-diagonal matrix element then become

\[
\rho_{e,e}(t) = \rho_{e,e}(0) e^{-2\gamma_0 t},
\]

(44)

\[
\rho_{e,g}(t) = \rho_{e,g}(0) e^{-2\omega_0 t - \gamma_0 t}.
\]

(45)

Due to the two possible decay channels \(|\uparrow\uparrow\rangle \Rightarrow |\uparrow\downarrow\rangle\) and \(|\uparrow\uparrow\rangle \Rightarrow |\downarrow\downarrow\rangle\), the decay of the excited state population occurs at twice the decay rate of a single atom.

3.2. Density matrix elements \( \sigma_{a,b} \)

From equation (39) the Laplace transformed diagonal elements \( \sigma_{\pm,a}(s) \) can be shown to be identical and of the form

\[
\sigma_{\pm,a}(s) = \rho_{e,e}(0) \int_0^s \int d\omega \omega \frac{|\kappa_{a,k,\sigma}^2|^2}{(ck - is)(ck + ip)}.
\]

(47)

After performing the sum over polarizations and integrating over spherical angles of the wave vector, this integral becomes

\[
I(s, p) = \int d^3k \sum_{\sigma} \frac{|\kappa_{a,k,\sigma}^2|^2}{(ck - is)(ck + ip)}.
\]

(48)

Using equations (33) and (48), the matrix elements (46) can be rewritten as

\[
\sigma_{\pm,a}(s) = \frac{\gamma_0}{2\pi \omega_0^3} \rho_{e,e}(0) \int_0^s \int d\omega \omega^3 g(\omega, s - s') f^{*}(\vec{r}, s') I(s - s', s'),
\]

(49)

where the new function \( g \) is given by

\[
g(\omega, s) = \frac{f(s)}{\omega + is} = \frac{i}{(s + i\omega)(s + i\omega(1 + \gamma_0))},
\]

(50)

where definition (26) together with equation (32) have been used. Thus, in the time domain, the diagonal matrix elements are

\[
\sigma_{\pm,a}(t) = \frac{\gamma_0}{2\pi \omega_0^3} \rho_{e,e}(0) \int_0^\infty d\omega \omega^3 |g(\omega, t)|^2.
\]

(51)

1 Note that the divergence of this integral is removed by the fact that it is integrated over \( s \) and \( p \), as given in equation (46).
The Laplace-transformed centred off-diagonal matrix elements can be obtained analogously by inserting equation (34) into equation (39). Both elements result as identical and read

$$\sigma_{\pm,\pm}(s) = \rho_{e,0}(0) \times \int_0^\infty \frac{ds'}{s'} f(\mathbf{r}, s - s') f^*(\mathbf{r}, s') I_\xi(s - s', s', \tau_r),$$

(52)

where the retardation time of light propagation between the atoms is \(\tau_r = r/c\). The integral is defined as

$$I_\xi(s, p, r) = \int d^3k \sum_\sigma |\mu_\xi^{(p)}(ck - is)(ck + ip)|^2, \quad (s \geq p),$$

(53)

with \(\theta = \angle(\mathbf{r}, \mathbf{k})\) and \(\xi = \sin^2 \angle(\mathbf{r}, \mathbf{d})\), assuming the induced electrical dipole moments of the two atoms to be parallel. After summing over polarizations and integrating over the spherical angles of the wavelight vector, this integral becomes

$$I_\xi(s, p, r) = \frac{\gamma_0}{2\pi \omega_0} \int_0^\infty d\omega \omega |\mu_\xi^{(p)}(\omega + ip)|^2, \quad (s \geq p),$$

(54)

where the dissipative part of the dipole–dipole interaction pattern is given by [29, 30]

$$\mu_\xi(\omega) = \frac{3}{2} \left[ (3\xi - 2) \left( \frac{\cos x}{x^3} - \frac{\sin x}{x^2} \right) + \frac{\sin x}{x} \right].$$

(55)

Inserting equation (54) into (52) and using the approximation (33) the off-diagonal matrix elements result as

$$\sigma_{\pm,\pm}(s) = \frac{\gamma_0}{2\pi \omega_0} \rho_{e,0}(0) \int_0^\infty d\omega \omega |\mu_\xi^{(p)}(\omega + ip)|^2 \times \int_0^\infty ds' g(\omega, s - s') g^*(\omega, s').$$

(56)

Thus, in the time domain, the matrix elements (56) become

$$\sigma_{\pm,\pm}(t) = \frac{\gamma_0}{2\pi \omega_0} \rho_{e,0}(0) \int_0^\infty d\omega \omega |P(\omega)(1 + e^{-2\gamma_0 t} - 2 \cos[(\omega - \omega_0)t]e^{-\gamma_0 t})|,$$

(57)

In both expressions, equations (51) and (57), the spectral weighting factor \(|g|^2\) results as a Lorentzian profile modulated by a damped harmonic oscillation

$$|g(\omega, t)|^2 = \frac{\pi}{\gamma_0} P(\omega)\{1 + e^{-2\gamma_0 t} - 2 \cos[(\omega - \omega_0)t]e^{-\gamma_0 t}\},$$

(58)

where the Lorentzian spectrum is given by

$$P(\omega) = \frac{1}{\pi} \frac{\gamma_0}{\omega^2 + \gamma_0^2/4}. $$

(59)

Due to the Lorentzian, the cosine in equation (58) contains the argument \((\omega - \omega_0)t \lesssim \gamma_0 t\). For \(\gamma_0 t \ll 1\) we may therefore approximate equation (58) by

$$|g(\omega, t)|^2 \approx \frac{\pi}{\gamma_0} P(\omega)(1 - e^{-\gamma_0 t})^2.$$ 

(60)

This approximation is consistent both with the fact that \(|g(\omega, t = 0)|^2 = 0\) and with the limiting value for \(\gamma_0 t \gg 1\). Therefore, it may be used also for large times without introducing large error.

Using the approximation (60), the off-diagonal elements finally result as

$$\sigma_{\pm,\pm}(t) = \frac{\rho_{e,0}(0)}{4} \frac{\bar{A}_\xi(r)(1 - e^{-\gamma_0 t})^2}{\bar{A}_\xi(r)},$$

(61)

where the spectrally averaged dissipative dipole–dipole pattern reads

$$\bar{A}_\xi(r) = \frac{2}{\omega_0^3} \int_0^\infty d\omega \omega^3 P(\omega)\mu_\xi^{(p)}(\omega r/c).$$

(62)

Performing analogous approximations for the diagonal matrix elements (51) results in

$$\sigma_{\pm,\pm}(t) \approx \frac{\rho_{e,0}(0)}{4} (1 - e^{-\gamma_0 t})^2.$$ 

(63)

As \(\bar{A}_\xi(r) = [1 + (\gamma_0/\omega_0)^2]^{3/2} \approx 1\), the latter diagonal elements become

$$\sigma_{\pm,\pm}(t) \approx \frac{\rho_{e,0}(0)}{4} (1 - e^{-\gamma_0 t})^2.$$ 

(64)

Note that the inner matrix elements, equations (61) and (64), have only one time scale, \(\gamma_0^{-1}\), at which they increase in time. This is different from the results obtained for two atoms with classical fixed positions [21]. There, two time scales have been derived, where the first coincides with \(\gamma_0^{-1}\) and the second is \([\pi \mu_\xi^{(p)}(k_0c)]^{-1}\), where \(r_c\) is the classical fixed inter-atomic distance.

3.3. Averaged dissipative dipole–dipole pattern

For the extreme far field, where the mean interatomic distance \(\bar{r} \gg r_c\) with the critical distance being defined by

$$r_c = (\frac{\omega_0}{\gamma_0}) \lambda_0,$$

(65)

in the average (62) the spectral width of the Lorentzian generates in the argument of the dipole–dipole pattern \(\mu_\xi\) a corresponding width \(\Delta x \gg 2\pi r_c\). Thus, the \(2\pi r_c\)-periodic oscillation of \(\mu_\xi\) will be effectively averaged over to result in \(\bar{A}_\xi(r) \approx 0\). Thus, in this case the off-diagonal matrix elements are

$$\sigma_{\pm,\pm}(t) = 0, \quad (\bar{r} \gg r_c).$$

(66)

In the opposite case, when the mean inter-atomic distance satisfies the condition \(\bar{r} \lesssim r_c\), which still permits very large distances between the atoms, the Lorentzian generates a sufficiently small width to provide a non-vanishing average \(\bar{A}_\xi\). Under these circumstances we may take the dipole–dipole pattern at the maximum \(\omega_0\) of the Lorentzian, to obtain

$$\bar{A}_\xi(r) \approx \mu_\xi^{(p)}(k_0c).$$

(67)

As equation (67) decays naturally when extending to the far and extreme far field, we may use it as a general approximation valid in the near, inductive and far field—including the extreme far field. Thus, the off-diagonal matrix elements can be further approximated as

$$\sigma_{\pm,\pm}(t) \approx \frac{\rho_{e,0}(0)}{4} \frac{\bar{A}_\xi(r)}{\mu_\xi^{(p)}(k_0c)}.$$ 

(68)

The average over the dissipative dipole–dipole pattern is illustrated in figure 1, where the function \(\mu_\xi^{(p)}(k_0c)\) is shown for the case when both atomic dipoles are perpendicular (solid
expressions can be written as \( \mu / \Delta_1 r \). For the density matrix (36), the concurrence takes the form

\[
\epsilon = \frac{1}{2} \left( \sqrt{p_{e,e}(0)^2 + p_{e,g}(0)^2} - p_{e,e}(0) - p_{e,g}(0) \right),
\]

where \( p_{e,e}(0) \) and \( p_{e,g}(0) \) are the amplitudes of the electronic states in the initial state. The zeros of these functions, \( c_1 \) and \( c_2 \), are functions of \( \mu_\xi \). The zeros of \( c_2 \) are plotted for values of \( \mu_\xi \) being: 1.0 (blue), 0.5 (green) and 0.0 (red).

\[
\xi = \frac{1}{2} \left( \sqrt{p_{e,e}(0)^2 + p_{e,g}(0)^2} - p_{e,e}(0) - p_{e,g}(0) \right),
\]

where \( p_{e,e}(0) \) and \( p_{e,g}(0) \) are the amplitudes of the electronic states in the initial state. The zeros of these functions, \( c_1 \) and \( c_2 \), are functions of \( \mu_\xi \). The zeros of \( c_2 \) are plotted for values of \( \mu_\xi \) being: 1.0 (blue), 0.5 (green) and 0.0 (red).
For a pure state \( (q = 1) \), the dependence of the zeros on the excited-state probability \( p \) and on the mean dipole–dipole pattern \( \mu_\xi \) is shown in figure 2, where \( \epsilon_1 \) is plotted in black and \( \epsilon_2 \) in blue (\( \mu_\xi = 1 \)), green (\( \mu_\xi = 1/2 \)) and red (\( \mu_\xi = 0 \)). It can be observed that for a given value of \( p \), the time of disentanglement, i.e. the time window between ESD and ESB, decreases with increasing mean \( \mu_\xi \).

A trivial case is obtained for \( p = 0 \), where both atoms start in their electronic ground states. In this case the concurrence vanishes at all times. For \( 0 < p < 1 \), which includes the case of initial maximal entanglement \( (p = 1/2) \), the concurrence first decays, ending with an ESD, stays absent for a period of time, and then revives in the form of an ESB, see figure 3. Apart from the case \( \mu_\xi = 0 \) (red curve) where the concurrence does not revive—which applies to far field, weak localization and strong localization near a node of \( \mu_\xi \)—the concurrence always revives and reaches a stationary value. Thus, there is no asymptotic decay to zero!

The special case \( p = 1 \), where initially both atoms are in their electronic excited state and thus initially are not entangled, is of particular interest. In this case, a ESB is present without any previous ESD or entanglement, see figure 4. This behaviour can be explained when considering that entanglement between the atoms and the em field is created when one of the atoms emits a photon, with the identity of the emitting atom being unknown. We will come back to this issue in section 4.2.

In the case of a mixed state, \( c_1 \) and thus also the zero \( \epsilon_1 \) are modified by the value of \( q \). This corresponds to an obvious modification of the initial value of the concurrence together with a modification of the decay and thus ESD. However, the rebirth of the concurrence is determined by the function \( c_2 \) and thus is not affected, as can be seen when comparing figure 3 with figure 5.

### 4.2. Stationary concurrence due to a dark state

For \( t \to \infty \) the two functions, equations (72) and (73), reach the stationary values

\[
\lim_{t \to \infty} c_1(t) = -\frac{\rho_{ee}(0)}{4}, \quad (78)
\]

\[
\lim_{t \to \infty} c_2(t) = \frac{\rho_{ee}(0)}{4} \mu_\xi, \quad (79)
\]

so that there exists a stationary value for the concurrence

\[
\lim_{t \to \infty} C(t) = \frac{\rho_{ee}(0)}{2} \mu_\xi. \quad (80)
\]
This result might at first sight appear counter-intuitive, as it states that there will be never any asymptotic decay to zero of the concurrence. However, when considering the electronic–photonic and motional dynamics it becomes clear that during the interaction of the atoms with the em field a so-called dark state is created. Starting from the two atoms being excited, the system may evolve into a superposition state where one photon has been emitted by either atom, see figure 6.

This superposition state is an eigenstate of the Hamiltonian (4) when neglecting the quantum dispersion, i.e. discarding the kinetic energy term. As a consequence, this state is stationary and thus a further photon emission does not occur—i.e. it is a dark state. This stationary state manifests entanglement between all partitions. If the emitted photon does not carry information on its emission centre, i.e. on the identity of the emitting atom, tracing over the em field does not destroy the entanglement between the two atoms. Moreover, if the associated photon recoil does not provide information on which atom emitted the photon, tracing over the inter-atomic distance does not diminish the electronic entanglement between atoms. In such a case, the dark state is stationary, the concurrence reaches a stationary value given by equation (80).

Contrary to this situation, when the emitted photon and/or the associated recoil carry information on the identity of the emitting atom, after tracing over the em field and inter-atomic distance, the reduced electronic state does not reveal entanglement. This situation is realized in various ways, all characterized by the condition $\mu_\xi = 0$. For instance $\mu_\xi = 0$ if (a) the atoms are weakly localized, (b) strongly localized but near a node of $\mu_\xi$ or (c) in the far field $\vec{r} \gg \lambda_0$. In the latter case, one may speak of two statistically independent reservoirs for the two atoms, where our result is in agreement with [7].

Of course, as the kinetic-energy term in equation (4) has been approximated, the discussed dark state is not an exact eigenstate of the full Hamiltonian (4). As a consequence, in a non-approximate treatment the dispersion of the inter-atomic distance would lead to a finite lifetime of this dark state. However, this lifetime would be much larger than the lifetime of the electronic excited state, $2\pi/\gamma_0$ (cf the discussion in section 1). In this sense, our approximation is consistent for typical timescales of electronic decay.

5. Summary and conclusions

In summary it has been shown that different from the case of two atoms with classical fixed positions, where more than one ESD and one ESB may occur [21], for the case of two free atoms with quantized relative motion and initially at rest, only one ESD and one ESB can occur. Furthermore, the inclusion of photon recoil permits the creation of a dark state by spontaneous emission. This dark state manifests itself in a stationary electronic entanglement between the atoms, that even may be created from an initially separable state. Whether this stationary entanglement appears or not, depends crucially via the photon recoil on the statistics of the inter-atomic distance.

We note that a similar behaviour has been observed in the non-Markovian calculation of [26]. We believe that these phenomena can be well explained within the framework of a dark state, as discussed here.

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Appendix. Kraus decomposition of the reduced density matrix

It can be shown that the reduced density matrix (36) can be decomposed as a Kraus representation

$$\rho(t) = \sum_{n=0}^{2} K_n(t) \cdot \rho(0) \cdot K_n^\dagger(t),$$

where $\rho(0)$ is the initial density matrix, which is a physical density matrix. The Kraus operators read in their unitary matrix form

$$K_0(t) = \begin{pmatrix} \alpha(t) & 0 & 0 & 0 \\ 0 & \beta(t) & 0 & 0 \\ 0 & 0 & \gamma(t) & 0 \\ 0 & 0 & 0 & \delta(t) \end{pmatrix},$$

$$K_1(t) = \begin{pmatrix} 0 & \varepsilon^\dagger(t) & \varepsilon(t) & 0 \\ \varepsilon^\dagger(t) & 0 & 0 & 0 \\ \varepsilon(t) & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},$$

$$K_2(t) = \begin{pmatrix} 0 & 0 & \zeta(t) & 0 \\ 0 & 0 & 0 & \zeta^\dagger(t) \\ 0 & 0 & 0 & 0 \\ \zeta(t) & 0 & 0 & 0 \end{pmatrix},$$

where always a solution can be found for the four real functions $\alpha$, $\beta$, $\gamma$ and $\delta$ and the two complex functions $\varepsilon$ and $\zeta$. Each Kraus operator $K_n$ corresponds to the emission of $n$ photons.

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