Atomic entanglement near a realistic microsphere

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Abstract. We study a scheme for entangling two-level atoms located close to the surface of a dielectric microsphere. The effect is based on medium-assisted spontaneous decay, rigorously taking into account dispersive and absorptive properties of the microsphere. We show that even in the weak-coupling regime, where the Markov approximation applies, entanglement up to 0.35 ebits between two atoms can be created. However, larger entanglement and violation of Bell’s inequality can only be achieved in the strong-coupling regime.

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1. Introduction

A bipartite quantum system is said to be entangled if its state cannot be represented as a convex sum of direct products of its subsystem states. Quantum entanglement entails correlations between outcomes of particular experiments at (spatially) separated objects which break certain Bell’s inequalities, predicted by local realistic theories [1]. Many experiments have been performed to test Bell’s inequalities [2], with the locality [3] and detection efficiency [4] loopholes recently reported to be closed. Despite this breakthrough, a decisive experiment to rule out any local realistic theory is yet to be performed [5]. Beyond the fundamental aspects, entanglement is a key resource for many applications in quantum information processing, including secret key distribution [6], dense coding [7], and teleportation [8].

Atoms can be entangled through interaction with a (common) electromagnetic field. The effect, which is very weak in free space, can be enhanced significantly in resonator-like equipments. Proposals have been made for entangling spatially separated atoms in Jaynes-Cummings systems through strong resonant atom-field coupling [9]. The coupling can be sequential or simultaneous. One of these schemes has been realized using Rydberg atoms coupled one by one to a high $Q$ microwave superconducting microcavity [10], with achieved probability of preparing a maximally entangled state in the range of 0.63 and two atoms separated by centimetric distances. To limit photon losses, off-resonant coupling, where the cavity mode is only virtually excited, can also be employed as recently proposed [11] and implemented [12]. Another proposal involves continuous monitoring of photons leaking out of the cavity. Provided no photon is detected outside the cavity, a pure entangled state between two atoms results [13, 14].

In this paper we first consider the more general problem of generation of two-atom entangled states in the presence of dispersing and absorbing dielectric bodies owing to the medium-assisted change of the spontaneous emission and the mutual dipole-dipole coupling of the atoms. Our investigation is based on a macroscopic approach to the electromagnetic field quantization which represents the potential and field operators in terms of a Green tensor expansion over appropriately chosen fundamental bosonic field variables (for a review, see [15]).

We apply the theory to the case of the two atoms being near a microsphere and show that the scheme, in contrast to much of previous work, does not require a strong atom-field coupling regime to realize entanglement, but the resulting state is not maximally entangled. Further, we study the strong-coupling regime, taking rigorously into account atomic spontaneous decay, photon leakage from the microsphere, and material absorption and dispersion. As an example, numerical calculations are performed for a microsphere whose permittivity is of Drude–Lorentz type.
2. Basic equations

Let us consider $N$ two-level atoms [positions $r_A$, transition frequencies $\omega_A$ ($A = 1, 2, \ldots, N$)] that resonantly interact with radiation via electric-dipole transitions (dipole moments $d_A$). Let us further assume that the atoms are sufficiently far from each other, so that the interatomic Coulomb interaction can be ignored. In this case, the electric-dipole and rotating wave approximations apply, and the minimal-coupling Hamiltonian takes the form of [13]

$$\hat{H} = \int d^3r \int_0^\infty d\omega \ h \omega \hat{f}^\dagger(r, \omega) \hat{f}(r, \omega) + \sum_A \frac{1}{2} h \omega_A \hat{\sigma}_{Az} - \sum_A [\hat{\sigma}_A \hat{E}^{(+)}(r_A) d_A + \text{H.c.}],$$

where the two-level atoms are described by the Pauli operators $\hat{\sigma}_A$, $\hat{\sigma}_A^\dagger$, and $\hat{\sigma}_{Az}$, and $\hat{f}(r, \omega)$ and $\hat{f}^\dagger(r, \omega)$ are bosonic field operators which play the role of the fundamental variables of the electromagnetic field and the medium, including a reservoir necessarily associated with the losses in the medium. The electric-field operator is expressed in terms of $\hat{f}(r, \omega)$ as

$$\hat{E}^{(+)}(r) = i \sqrt{\frac{h}{\pi \varepsilon_0}} \int_0^\infty d\omega \frac{\omega^2}{\varepsilon^2} \int d^3r' \sqrt{\varepsilon_1(r', \omega)} G(r, r', \omega) \hat{f}(r', \omega),$$

where $G(r, r', \omega)$ is the classical Green tensor and $\varepsilon(r, \omega) = \varepsilon_R(r, \omega) + i \varepsilon_I(r, \omega)$ the complex permittivity.

For a single-quantum excitation, the system wave function at time $t$ can be written as

$$|\psi(t)\rangle = \sum_A C_{U_A}(t) e^{-i(\omega_A - \bar{\omega})t} |U_A\rangle \{0\}$$

$$+ \int d^3r \int_0^\infty d\omega \left[ C_{Li}(r, \omega, t) e^{-i(\omega - \bar{\omega})t} |L\rangle \{1_i(r, \omega)\} \right]$$

($\bar{\omega} = \frac{1}{2} \sum_A \omega_A$). Here, $|U_A\rangle$ is the excited atomic state, where the $A$th atom is in the upper state and all the other atoms are in the lower state, and $|L\rangle$ is the atomic state, where all atoms are in the lower state. Accordingly, $\{0\}$ is the vacuum state of the rest of the system, and $\{1_i(r, \omega)\} = \hat{f}^\dagger_i(r, \omega) \{0\}$ is the state, where it is excited in a single-quantum Fock state. Note that this state is not a photonic state in general, but a state of the macroscopic medium dressed by the electromagnetic field. From the Schrödinger equation, we obtain the following (closed) system of integro-differential equations:

$$\dot{C}_{U_A}(t) = \sum_{A'} \int_0^t dt' K_{AA'}(t, t') C_{U_{A'}}(t') + f_A(t),$$

$$f_A(t) = -\frac{1}{\sqrt{\pi \varepsilon_0 h}} \int_0^\infty d\omega \int d^3r \left\{ \frac{\omega^2}{c^2} e^{-i(\omega - \bar{\omega})t} \left[ \sqrt{\varepsilon_1(r, \omega)} \right] d_A G(r_A, r, \omega) C_L(r, \omega, 0) \right\},$$

$$K_{AA'}(t, t') = -\frac{1}{h \pi \varepsilon_0 \varepsilon_I} \int_0^\infty d\omega \left[ \frac{\omega^2}{c^2} e^{-i(\omega - \bar{\omega})t'} e^{i(\omega - \bar{\omega})t} d_A \text{Im} G(r_A, r_A', \omega) d_{A'} \right].$$

Note that $K_{AA'}(t, t') = K_{A'A}(t', t)$ because of the reciprocity theorem.

The excitation can initially reside either in one of the atoms or the medium-assisted electromagnetic field. The latter case, i.e., $C_L(r, \omega, 0) \neq 0$ in equation (5), could be
realized, for example, by coupling the field first to an excited atom $D$ during a time interval $\Delta t$ such that

$$ C_L(\mathbf{r}, \omega, 0) = \int_{-\Delta t}^{0} dt' \frac{\omega^2}{\sqrt{\pi \varepsilon_0 \hbar}} e^{i(\omega - \omega_D)t'} \sqrt{\varepsilon_1(\mathbf{r}, \omega)} \mathbf{d}_D \mathbf{G}^*(\mathbf{r}_D, \mathbf{r}, \omega) C_U(t'), $$

where $C_U(t)$ describes the single-atom decay \[16\]. Substitution of the expression \[7\] into equation \[5\] then yields

$$ f_A(t) = \int_{-\Delta t}^{0} dt' K_{AD}(t, t') C_U(t'). \tag{8} $$

We now turn to the problem of two atoms, denoted by $A$ and $B$. For simplicity, let us consider atoms with equal transition frequencies $\omega_A = \omega_B = \omega_D$, so that

$$ K_{AA'}(t, t') \equiv K_{AA'}(t - t') \tag{9} $$

($A' = B, D$) and

$$ K_{AB}(t - t') = K_{BA}(t - t'), \tag{10} $$

and assume that the isolated atoms follow the same decay law,

$$ K_{AA}(t, t') = K_{BB}(t, t') \equiv K(t - t'). \tag{11} $$

Introducing the new variables

$$ C_\pm(t) = 2^{-1/2}[C_{UA}(t) \pm C_{UB}(t)], \tag{12} $$

it is not difficult to prove that the system of equations \[4\] [together with equation \[8\]] decouples as follows:

$$ \dot{C}_\pm(t) = \int_{-\Delta t}^{0} dt' K_\pm(t - t') C_ \pm(t') + \frac{1}{\sqrt{2}} \int_{-\Delta t}^{0} dt' \left[ K_{AD}(t - t') \pm K_{BD}(t - t') \right] C_U(t'), \tag{13} $$

where

$$ K_\pm(t - t') = K(t - t') \pm K_{AB}(t - t'). \tag{14} $$

Obviously, the $C_\pm(t)$ are the expansion coefficients of the wave function with respect to the (atomic) basis

$$ |\pm\rangle = 2^{-1/2}(|U_A\rangle \pm |U_B\rangle) \tag{15} $$

and $|L\rangle$ (instead of the basis $|U_A\rangle$, $|U_B\rangle$, and $|L\rangle$). Thus, $C_+(t)$ and $C_-(t)$ are the probability amplitudes of finding the total system in the states $|+\rangle\{0\}$ and $|-\rangle\{0\}$, respectively. In the further treatment of equation \[13\] one can distinguish between the weak- and the strong-coupling regime.

2.1. Weak Coupling

In the weak-coupling regime, the Markov approximation applies, and in equation \[13\] $C_\pm(t')$ can be replaced with $C_\pm(t)$, with the time integrals being $\zeta$-functions. In particular, when the medium-assisted field is initially not excited, then the second term
on the right-hand side of equation (13) vanishes and we are left with a homogeneous first-order differential equation, whose solution is

\[ C_\pm(t) = e^{(-\Gamma_\pm/2+i\delta_\pm)t}C_\pm(0), \] (16)

where \((\Gamma \equiv \Gamma_{AA}, \delta \equiv \delta_{AA})\)

\[ \Gamma_\pm = \Gamma \pm \Gamma_{AB}, \quad \delta_\pm = \delta \pm \delta_{AB}, \] (17)

\[ \Gamma_{AB} = \frac{2\omega^2_A}{\hbar \varepsilon_0 c^2} d_A \text{Im} \, G(r_A, r_B, \omega_A) d_B, \] (18)

\[ \delta_{AB} = \frac{P}{\pi \hbar \varepsilon_0} \int^\infty_0 d\omega \frac{\omega^2 d_A \text{Im} \, G(r_A, r_B, \omega) d_B}{\omega - \omega_A}. \] (19)

Obviously, \(\Gamma_\pm\) are the decay rates of the states \(|\pm\rangle\), and the assumption (11) means that the two atoms are positioned in such a way that they have equal single-atom decay rates and line shifts. Note that the values of \(\Gamma_+\) and \(\Gamma_-\) can substantially differ from each other, because of the interference term \(\Gamma_{AB}\) (of positive or negative sign).

At this point it should be mentioned that, on starting from the Hamiltonian (1), the reduced density operator for the atomic subsystem in the weak-coupling regime can be shown to obey the equation

\[ \dot{\rho} = -\frac{i}{2} \sum_{A,A'} \left( \hat{\sigma}^+_A \hat{\sigma}^+_A \rho - 2\hat{\sigma}^+_A \hat{\sigma}^+_A \rho + \hat{\sigma}^+_A \hat{\sigma}^+_A \right) + i \sum_{A,A'} \delta_{AA'} [\hat{\sigma}^+_A \hat{\sigma}^+_A, \rho] \] (20)

\((A, A' = 1, 2, ..., N)\). It is worth noting that this result is in agreement with the result given in [17], where Kubo’s formula is applied to the field correlation functions.

2.2. Strong Coupling

In the strong-coupling regime (i.e., when the atoms are in a resonator-like equipment of sufficiently high quality), the atoms are predominantly coupled to a sharp field resonance, whose mid-frequency approximately equals the atomic transition frequency. As a result, the probability amplitudes in equation (13) must not necessarily be slowly varying compared with the kernel functions and the Markov approximation thus fails in general. Regarding the line shape of the field resonance as being a Lorentzian, one can of course approximate the kernels \(K(t-t')\), \(K_{AB}(t-t')\) [and \(K_{AD}(t-t')\) and \(K_{BD}(t-t')\)] in a similar way as done in [16] for a single atom.

Equation (13) reveals that the motion of the states \(|\pm\rangle\) defined by equation (15) is governed by the kernel functions \(K_{\pm}(t-t')\), and it may happen that one of them becomes very small, because of destructive interference [cf. equation (14)]. In that case, either \(|+\rangle\) or \(|-\rangle\) is weakly coupled to the field, and thus the strong-coupling regime cannot be realized for both of these states simultaneously.
3. Entangled-state preparation

Let us consider a particular configuration of material bodies, namely, a microsphere (radius \( R \)), which can act as a microcavity [18]. It is well known that rays of suitable wavelengths may bounce around the rim by total internal reflection, forming the so-called whispering gallery (WG) waves. These field resonances can combine extreme photonic confinement with very high quality factors – properties that are crucial for cavity QED experiments [19] and many optoelectronical applications [18]. For a band-gap material and frequencies inside the band-gap, a microsphere can also give rise to high quality surface-guided (SG) waves [20].

Here we assume that the microsphere material can be characterized by a (single-resonance) Drude–Lorentz-type permittivity

\[
\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_T^2 - \omega^2 - i\omega\gamma},
\]

where \( \omega_P \) corresponds to the coupling constant, and \( \omega_T \) and \( \gamma \) are the medium oscillation frequency and the linewidth, respectively. Recall that the Drude–Lorentz model covers both metallic (\( \omega_T = 0 \)) and dielectric (\( \omega_T \neq 0 \)) matter and features a band gap between \( \omega_T \) and \( \omega_L = \sqrt{\omega_T^2 + \omega_P^2} \).

3.1. Weak Coupling

Let us restrict our attention to two identical (two-level) atoms located at diametrically opposite positions (\( r_A = -r_B \)) outside the microsphere and having radially oriented transition dipole moments. Obviously, the conditions (10) and (11) are fulfilled for such a system, so that from equations (17) and (18) together with the Green tensor for a microsphere [21] one can find that

\[
\Gamma_{\pm} = \frac{3}{2}\Gamma_0 \sum_{l=1}^{\infty} \text{Re} \left\{ \frac{l(l+1)(2l+1)}{(k_A r_A)^2} h_l^{(1)}(k_A r_A) \right\} \left[ j_l(k_A r_A) + B_l^N(\omega_A) h_l^{(1)}(k_A r_A) \right] \left[ 1 \mp (-1)^l \right] \cdot
\]

\[k_A = \omega_A/c; j_l(z) \text{ and } h_l^{(1)}(z), \text{ spherical Bessel and Hankel functions; } B_l^N(\omega_A), \text{ generalized reflection coefficient [21]; } \Gamma_0, \text{ decay rate of a single atom in free space]. When atom } A \text{ is initially in the upper state and atom } B \text{ is accordingly in the lower state, then the two superposition states } |+\rangle \text{ and } |-\rangle \text{ [equation (14)] are initially equally excited } [C_+(0) = C_-(0) = 2^{-1/2}]. \text{ If the atomic transition frequency coincides with a microsphere resonance, the single-atom decay rate } \Gamma \text{ may be approximated (for sufficiently small atom-surface distance) by [20]}

\[
\Gamma \simeq \frac{3}{2}\Gamma_0 l(l+1)(2l+1) \text{Re} \left\{ \frac{h_l^{(1)}(k_A r_A)}{k_A r_A} \right\}^2 B_l^N(\omega_A),
\]

and equation (22) can be approximated as follows:

\[
\Gamma_{\pm} \simeq \Gamma \left[ 1 \mp (-1)^l \right].
\]
Hence $\Gamma_- \gg \Gamma_+$ ($\Gamma_+ \gg \Gamma_-$) if $l$ is even (odd), i.e., the state $|\rangle$ ($|\rangle$) decays much faster than the state $|\rangle$ ($|\rangle$).

Figure 1. The dependence of the decay rates $\Gamma_+$ (solid line) and $\Gamma_-$ (dashed line) on (a) the transition frequency and (b) the distance of the atoms from a microsphere is shown for two atoms at (with respect to a sphere) diametrically opposite positions, radially oriented transition dipole moments, and a single-resonance Drude–Lorentz-type dielectric $[R = 10 \lambda_T \ (\lambda_T = 2\pi c/\omega_T); \ \omega_T = 0.5 \omega_T; \ \gamma = 10^{-6} \omega_T; \ \Delta r_B = \Delta r_A \geq 10^{-2} \lambda_T$; (a) $\Delta r_A = 0.02 \lambda_T$; (b) $\omega_A \simeq 1.0501 \omega_T$].

The (exact) frequency dependence of $\Gamma_\pm$ as given by equation (22) is illustrated in figure (a) for a frequency interval inside a band gap, and the dependence on the
atom-surface distance is illustrated in figure 1(b). We see that the values of $\Gamma_+$ and $\Gamma_-$ can be substantially different from each other before they tend to the free-space rate $\Gamma_0$ as the distance from the sphere becomes sufficiently large. In particular, the decay of one of the states $|+\rangle$ or $|-\rangle$ can strongly be suppressed at the expense of the other one, which decays rapidly. Note that $\Gamma_+$ also differs from $\Gamma_-$ for two atoms in free space [22]. However, the difference that occurs by mediation of the microsphere is much larger. For example, at the distance for which in figure 1(b) $\Gamma_+$ attains the minimum, the ratio $\Gamma_-/\Gamma_+ \simeq 6.7 \times 10^4$ is observed, which is to be compared with the free-space ratio $\Gamma_-/\Gamma_+ \simeq 1.0005$. The effect may become even more pronounced for larger microsphere sizes and lower material absorption, i.e., sharper microsphere resonances. Needless to say that it is not only observed for SG waves considered in figure 1, but also for WG waves.

From the above, there exists a time window, during which the overall system is prepared in an entangled state that is a superposition of the state with the atoms being in the state $|+\rangle$ (|−⟩) and the medium-assisted field being in the ground state, and all the states with the atoms being in the lower state $|L\rangle$ and the medium-assisted field being in a single-quantum Fock state. The window is opened when the state $|-\rangle$ (|+⟩) has already decayed while the state $|L\rangle$ emerges, and it is closed roughly after the lifetime of the state $|+\rangle$ (|−⟩). As a result, the two atoms are also entangled to each other. The state is a statistical mixture, the density operator of which is obtained from the density operator of the overall system by taking the trace with respect to the medium-assisted field. Within the approximations (16) and (24) it takes the form of

$$\hat{\rho} \simeq |C_{\pm}(t)|^2 |\pm\rangle\langle \pm| + 1 - |C_{\pm}(t)|^2 |L\rangle\langle L|,$$  

where

$$|C_{\pm}(t)|^2 \simeq 2^{-1}e^{-\Gamma_{\pm}t}. \quad (26)$$

Applying the separability criterion [23], it is not difficult to prove that the state (25) is indeed inseparable, in fact, for all times $t$. It is worth noting that the atoms become entangled within the weak-coupling regime, starting from the state $|U_A\rangle$ (or $|U_B\rangle$) and the vacuum field. In the language of (Markovian) damping theory one would probably say that the two atoms are coupled to the same dissipative system, which gives rise to the quantum coherence.

The time evolution of the entanglement of formation $E_F(\hat{\rho})$ (for the concept of entanglement of formation, see [24]) is shown in figure 4, where the entanglement is measured in ebits. It is clear from the structure of the coefficient $C_{\pm}(t)$ [equation (23)] that one can never achieve a maximally entangled state in the weak-coupling regime, since the state (23) is never pure. Applying the convexity property of entanglement measures, one realizes that the amount of entanglement contained in the state (25) is bounded according to [25]

$$E_F(\hat{\rho}) \leq |C_{\pm}(t)|^2. \quad (27)$$

Figure 2 reveals that at most 0.35 ebits can be achieved for $t \to 0$ (the limit $t \to 0$ is to be understood as the smallest time that is compatible with the requirement for the
fast decay channel to be already closed). It is further seen that the entanglement falls off faster than exponentially with time – a result that is already expected from the convexity property and equation (26).

Entangled-state preparation in the weak-coupling regime has the advantage that it could routinely be achieved experimentally. However, the value of $|C_{\pm}(t)|^2$ in equation (25) is always less than $1/2$. In order to achieve a higher degree of entanglement, a strong-coupling regime is required.

### 3.2. Strong Coupling

Let us assume that the two atoms are initially in the ground state and the medium-assisted field is excited. The field excitation can be achieved, for example, by coupling an excited atom $D$ to the microsphere and then making sure that the atomic excitation is transferred to the medium-assisted field. If the atom $D$ strongly interacts with the field, the excitation transfer can be controlled by adjusting the interaction time. Another possibility would be measuring the state populations and discarding the events where the atom is found in the upper state. Here we restrict our attention to the first method and assume that all three atoms $D$, $A$, and $B$ strongly interact with the same microsphere resonance (of mid-frequency $\omega_C$ and line width $\Delta \omega_C$). The upper-state probability amplitude $C_{UD}(t)$ of atom $D$ can then given by

$$C_{UD}(t) = e^{-\Delta \omega_C(t+\Delta t)/2} \cos[\Omega_D(t + \Delta t)/2],$$

(28)
with $\Omega_D$ being the corresponding Rabi frequency $\Omega = \sqrt{2 \Gamma C \Delta \omega_C}$, where $\Gamma_C$ is the value of $\Gamma$ at the frequency $\omega_C$; for the calculation of $\Delta \omega_C$, see [20]. For

$$
\Delta t = \pi/\Omega_D, \tag{29}
$$

the initially (i.e., at time $t = -\Delta t$) excited atom $D$ is at time $t = 0$ in the lower state $[C_U D(0) = 0]$. From the preceding subsection we know that when the resonance angular-momentum number $l$ is odd (even), then the state $|+\rangle$ ($|\rangle$) “feels” a sharply peaked high density of medium-assisted field states, so that a strong-coupling approximation applies. The state $|\rangle$ ($|+\rangle$), in contrast, “feels” a flat one and the (weak-coupling) Markov approximation applies. Assuming atom $A$ is at the same position as was atom $D$, from equations (13), (28), and (29) we then find that

$$
C_\pm(t) \simeq -e^{-\Delta \omega_C (t+\pi/\Omega_D)/2} \sin(\Omega_\pm t/2) \tag{30}
$$

($\Omega_\pm = \sqrt{2} \Omega$, with $\Omega$ being the Rabi frequency of atom $A$ or $B$), and

$$
C_\mp(t) \simeq 0 \tag{31}
$$

(the sign of $C_-(t)$ in equation (30) is reversed if atom $B$ is at the same position as was atom $D$). The two-atom entangled state is again of the form (25), but now the weight of the state $|+\rangle$ ($|\rangle$) can reach values larger $1/2$, provided that the resonance linewidth $\Delta \omega_C$ is small enough. An example of the time evolution of the entanglement of formation is shown in figure 3. The shape of the curve strongly depends on the ratios $\Delta \omega_C/\Omega_\pm$ and $\Delta \omega_C/\Omega_D$. Small values of $\Delta \omega_C/\Omega_\pm$ yield many oscillations with, on assuming that $\pi \Delta \omega_C/\Omega_D$ is also small, large achievable entanglement. This is the case in figure 3. Roughly speaking, $\Omega_D$ controls the maximal obtainable entanglement, $\Delta \omega_C$ the decrease of the envelope, and $\Omega_\pm$ the oscillation frequency. Highest entanglement is achieved if

$$
\Delta \omega_C \ll \Omega_\pm, \Omega_D. \tag{32}
$$
For example, choose a microsphere with a $Q$ factor of $2.4 \times 10^8$. At optical frequencies of $\omega_C \sim 3 \times 10^{15}$ Hz (i.e., $\lambda \approx 600$ nm) this amounts to $\Delta \omega_C \sim 10^7$ Hz. For $\lambda \approx 573$ nm, the spontaneous decay rate of a quantum dot, regarded as an artificial atom, has been measured to be $\Gamma \approx 5 \times 10^9$ Hz [24], so that it follows that $\Omega \sim 10^3 \ldots 10^2 \Delta \omega_C$. This justifies the parameters chosen in figure 3. The results also show that maximal entanglement of 1 ebit cannot be achieved in practice even in the strong-coupling regime.

3.3. Multiparticle entanglement

Besides entangling two atoms, the scheme can also be used to create multiparticle entanglement. Let us briefly discuss the problem of creating tripartite entanglement by a single excitation. Then, instead of the states $|U_A\rangle$, $|U_B\rangle$, and $|U_C\rangle$, and $|L\rangle$, it is helpful to use the states

$$|1\rangle = 3^{-1/2} (|U_A\rangle + |U_B\rangle + |U_C\rangle),$$  
$$|2\rangle = 6^{-1/2} (2|U_A\rangle - (|U_B\rangle + |U_C\rangle)),$$
$$|3\rangle = 2^{-1/2} (|U_B\rangle - |U_C\rangle),$$

and $|L\rangle$ as basis states. Note that the states $|1\rangle$ and $|2\rangle$ belong to the $W$ class of the tripartite entangled states [27]. If one assumes that the three atoms are identical and equivalently positioned near the microsphere so that

$$K_{II}(t-t') = K(t-t'), \quad I = A, B, C,$$
$$K_{AB}(t-t') = K_{BC}(t-t') = K_{CA}(t-t'),$$

the integro-differential equations for the amplitudes of the states $|i\rangle\{0\}$, $i = 1, 2, 3$ decouple [cf. equation (4)]:

$$\dot{C}_1 = \int_0^t dt' [K(t-t') + 2K_{AB}(t-t')]C_1(t') + f_1(t),$$
$$\dot{C}_{2(3)} = \int_0^t dt' [K_-(t-t')C_{2(3)}(t') + f_{2(3)}(t)],$$

where

$$f_1(t) = 3^{-1/2} [f_A(t) + f_B(t) + f_C(t)],$$
$$f_2(t) = 6^{-1/2} \{2f_A(t) - [f_B(t) + f_C(t)]\},$$
$$f_3(t) = 2^{-1/2} [f_B(t) - f_C(t)].$$

In the weak-coupling regime, on applying the Markov approximation, equations (38) and (39) can easily be solved to obtain

$$C_i(t) = e^{(-\Gamma_i/2 + i\delta_i)t}C_i(0), \quad i = 1, 2, 3,$$
$$\Gamma_1 = \Gamma + 2\Gamma_{AB}, \quad \delta_1 = \delta + 2\delta_{AB},$$
$$\Gamma_2 = \Gamma_3 = \Gamma_-, \quad \delta_2 = \delta_3 = \delta_-.$$

Here we have assumed that the excitation initially resides in the atomic subsystem. Suppose that atom $A$ is excited at $t = 0$, then $C_1(0) = 1/\sqrt{3}$, $C_2(0) = \sqrt{2}/3$, and...
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$C_3(0) = 0$, and it follows from equation (43) that $C_3(t) = 0$. If one can set up the system in such a way that $\Gamma_{AB} \sim -\Gamma/2$, then the state $|2\rangle$ decays fast, leaving the atomic subsystem in a mixed entangled state with the weight of $|1\rangle\langle 1|$ being less than $1/3$. Alternatively, if $\Gamma_{AB} \sim \Gamma$, then the state $|1\rangle$ decays fast, and the atomic subsystem is prepared in a mixed entangled state with the weight of $|2\rangle\langle 2|$ being less than $2/3$.

If the strong-coupling regime is realized for the state $|1\rangle$, and if the excitation is pumped into the system through the medium-assisted electromagnetic field in such a way that

$$f_A(t) = f_B(t) = f_C(t),$$

then $f_2(t) = f_3(t) = 0$ [see equations (41) and (42)], and an entangled state of the form (23), with $|1\rangle$ and $C_1(t)$ replacing $|\pm\rangle$ and $C_{\pm}(t)$, will be generated. The condition (46) can be fulfilled by, e.g., coupling the field first to an excited atom D placed at equidistance from the atoms $A$, $B$, and $C$. In the same way, more than three atoms can be entangled with each other.

4. Violation of Bell’s inequality

It is well known that entangled pairs of spatially separated particles contradict classical local realism – the idea that the properties of one particle cannot be affected instantaneously by a measurement made upon the other one that is sufficiently far away. This has been quantified in the form of Bell’s inequalities, which must be obeyed by any local realistic theory, but can be violated by quantum mechanics. Whereas violation of Bell’s inequalities always indicates entanglement, the opposite conclusion is wrong; entanglement must not necessarily lead to violation of Bell’s inequalities. It is therefore interesting to know the conditions under which violation of a Bell inequality could principally be observed. For spin system, the Bell inequality

$$B_S = |E(\theta_1, \theta_2) - E(\theta_1', \theta_2') + E(\theta_1', \theta_2) + E(\theta_1, \theta_2')| \leq 2 \quad (47)$$

is commonly considered, where

$$E(\theta_1, \theta_2) = \langle \hat{\sigma}_A^{\theta_1} \hat{\sigma}_B^{\theta_2} \rangle,$$

$$\hat{\sigma}_A^\theta = \cos \theta \hat{\sigma}_A^x + \sin \theta \hat{\sigma}_A^y. \quad (49)$$

When the state with both atoms simultaneously excited is not populated, as it is the case for a state of the type (23), it is not difficult to prove that

$$E(\theta_1, \theta_2) = E(\theta_1 - \theta_2, 0). \quad (50)$$

Let us choose

$$\theta = \theta_1 - \theta_2 = \theta_2 - \theta_1' = \theta_1' - \theta_2', \quad (51)$$

The inequality (47) then simplifies to

$$B_S = |3E(\theta, 0) - E(3\theta, 0)| \leq 2. \quad (52)$$
An entangled state of the type (25) can only give rise to a violation of the Bell’s inequality if $|C_\pm(t)|^2 \geq 2^{-1/2} \simeq 0.71$ [14], which cannot be achieved in the weak-coupling regime, equation (26). However, it can be achieved in the strong-coupling regime.
Atomic entanglement near a realistic microsphere

\[ E(\theta, 0) = \pm \cos \theta |C_\pm(t)|^2 = \pm \cos \theta e^{-\Delta \omega_C(t + \pi/\Omega_D)} \sin^2 \left( \Omega t/\sqrt{2} \right). \] (53)

Substitution of this expression into equation (52) yields, on choosing \( \theta = \pi/4 \),

\[ B_S = 2\sqrt{2} e^{-\Delta \omega_C(t + \pi/\Omega_D)} \sin^2 \left( \Omega t/\sqrt{2} \right), \] (54)

which clearly shows that \( B_S > 2 \) becomes possible as long as \( \Delta \omega_C(t + \pi/\Omega_D) \ll 1 \).

Examples of the temporal evolution of \( B_S \) are shown in figure 4(a). In figure 4(b) the dependence of the ratio \( \Omega/\Delta \omega_C \) on the distance of the atoms from the sphere is plotted. The strong-coupling regime can be observed for distances for which \( \Omega/\Delta \omega_C \gg 1 \) is valid. The inset reveals that the maximum value of \( B_S \) decreases with increasing atom-surface distance and reduces below the threshold value of 2 still in the strong-coupling regime.

It is well known that Bell’s inequality tests may suffer from low detection efficiencies or from distances between the two entangled parts which are smaller than the speed of light times the measurement time, thus allowing for the two particles to be connected by a signal during the measurement. In both cases, from the set of measured data it may be difficult to decide whether a Bell inequality is really violated or not. Following [14], the first loophole can be closed for massive particle entanglement considered here. So, the correlation function \( E(\theta, 0) \) can be determined experimentally by first applying laser pulses with appropriately chosen phases (single qubit rotations) on each of the two atoms and then measuring their state populations, which can be performed with extremely high efficiency. The second loophole, which typically occurs in Bell’s inequality tests for massive particle entanglement, has not fully been closed so far [4, 14]. The scheme proposed here for particle entanglement can be realized using, e.g., atomic beams passing nearby a microsphere or atoms (including quantum dots or other artificial atoms) at fixed positions. In particular, atomic-beam experiments might offer a possibility to close the light-cone loophole.

5. Summary and Conclusions

The spontaneous emission and the mutual dipole-dipole coupling of (two-level) atoms can drastically change due to the presence of macroscopic bodies. The effect can be used to entangle the atoms with each other. Apart from the shape of the bodies, their dispersive and absorptive properties are crucial. All these aspects can be taken into account in a consistent way by quantization of the phenomenological Maxwell field by means of a source-quantity representation of the field in terms of the (classical) Green tensor and appropriately chosen fundamental variables that describe collective excitations of the field and the matter including the reservoir. Basically, all that is needed is knowledge about the spatially varying, complex permittivity of the equipment. As functions of frequency, the real and imaginary parts of the permittivity must satisfy the Kramers-Kronig relations, which establish the fundamental relation between dispersion and absorption.
The case of two atoms located near a (dispersive and absorptive) microsphere and single-quantum excitation has been considered in some detail. It has been shown that in the weak coupling regime, where the Markov approximation applies, there is a time window for entangling the atoms. Entanglement up to 0.35 ebits can be achieved. The effect is somewhat unexpected, because it is commonly believed that only strong atom-cavity coupling can lead to interatomic entanglement. As shown, in the strong-coupling regime the created entanglement can be indeed much higher. However, perfect entanglement (in the sense of a pure Bell state) cannot be achieved in practice even in the strong-coupling regime. It is worth noting that Bell’s inequality can only be violated in the strong-coupling regime.

Needless to say that the formalism also applies to the study of the influence of other types of microcavities on the resonant atom-light interaction. Throughout the paper we have assumed that the mutual distance between the atom is large enough to disregard the interatomic Coulomb interaction. For sufficiently small distances this approximation fails. Moreover, to include in the theory the direct short-distance interaction between the atoms, the rotating-wave approximation may also fail. Both problems deserve further investigation.

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