The exponential laws for emission and decaying of entangled atoms

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Abstract
The first photon emission and the disentanglement of a pair of identical bosonic atoms in excited entangled states follow an exponential law. We extend the theory to distinguishable and identical fermionic two-atom systems. As a byproduct of the analysis we determine the symmetries of the fermionic wave function. We also derive the emission distributions of excited atoms in product states, which must take into account the presence of simultaneous detections. Comparing both distributions reveals a direct manifestation of the modifications induced by entanglement on the atomic emission properties.

1 Introduction

Entanglement plays an important role in the description of light-matter interactions in multi-particle systems. Several authors have analyzed different aspects of the problem ([1, 2, 3] and references therein), in particular, atomic absorption and emission [4]. Recently, two experiments have studied possible effects of entanglement in the emission distribution of excited hydrogen atoms generated in the photodissociation of $H_2$ molecules [5, 6]. An interpretation of the experiments has been presented in [7]. A fundamental element of this interpretation is the validity of exponential laws for the first photon emission and for the decay of the atomic pair to a non-entangled state.

In this paper we explore exponential laws in pairs of identical fermionic atoms and distinguishable ones. Following the strategy used for bosons, the analytical form of the decaying parameter can be evaluated by invoking the compatibility between the emission distributions with and without temporal ordering. In addition, in [7] it was shown that the explicit form of the decaying rate can be explained by a combination of three effects: entanglement, exchange effects (the photodissociated atoms are identical bosons) and disentanglement after the first spontaneous emission (the spontaneous emission by an atom leaves the multi-particle system in a non-entangled state [8, 9]). A similar argumentation can be
done here but, because of its similarity with the original one, we only sketch it in the Appendix. The only novel element of this argumentation is that, in the case of identical fermions, it determines the symmetries of the wave function previous to antisymmetrization.

Later, we consider the emissions by pairs of distinguishable atoms in excited product states. The evaluation of the distribution demands a careful consideration of the aspects related to the existence of simultaneous detections. The distributions for entangled and product states are different, providing a potential method to verify the dependence of the emission properties on entanglement. The main advantage of this approach is that, at variance with the arrangements [5, 6], this dependence appears independently of exchange effects. We can test without any interfering effect the influence of entanglement.

In the final part of the paper we discuss some conceptual aspects of the problem, in particular, the discontinuity between distributions associated with entangled and product states, the qualitative rather than quantitative influence of entanglement on emission, the characterization of multi-atom systems as single emitters, and the differences between exponential decaying laws and disentanglement in finite times [8, 9].

2 Emission in entangled states

In the case of pairs of excited identical bosonic atoms, the first photon emission obeys an exponential law [7]. As the first emission leads to the disentanglement of the atoms, the temporal variation of the number of entangled atoms must be expressed via the standard decaying law:

$$\frac{dn_e}{dt} = -\Gamma_f n_e$$

with $n_e$ as the number of entangled pairs and $\Gamma_f$ as the rate of first emissions. The trivial solution of the equation is $n_e(t) = n_0 \exp(-\Gamma_f t)$, where $n_0$ is the initial number of entangled pairs.

From the experimental point of view the interesting distribution is that of first emitted photons, much more simple to measure than that of entangled pairs. It is given by $N_f(t) = n_0(1 - \exp(-\Gamma_f t))$, which can be derived from $N_f(t) + n_e(t) = n_0$.

We shall explore the consequences of assuming that the decaying law also holds for pairs of distinguishable atoms. Thus, we assume that the two above equations for $n_e$ and $N_f$ remain valid when we consider a pair of distinguishable atoms denoted by $A$ and $B$.

We know the form of the emission distribution for each type of atom. If we do not compare the emission times of the two atoms, we have a single-atom process. Only when we compare these times we are dealing with multi-atom phenomena and a dependence on entanglement can be present [7]. Then the
distribution of each type of photon is that associated with an ensemble of single
atoms (or pairs of atoms in product states). The photon distribution is
\[ N_i(t) = n_0(1 - e^{-\Gamma_i t}) \] (2)
with \( i = A, B \) the two types of photons (denoted by the same symbols of the
emitting atoms), and \( \Gamma_i \) the emission rate of atoms of type \( i \).

From these distributions we can obtain the second photon emission \( N_s(t) =
N_A(t) + N_B(t) - N_f(t) \). However, in this paper we are not interested in this
distribution.

Next, we must evaluate the rate of first emissions. This can be done by
expressing \( N_i \) in terms of \( \Gamma_f \). We denote by \( n_i \) the number of excited atoms
of type \( i \) in product states, that is, the number of atoms whose companion in the
pair (of type \( j, j \neq i \)) has already emitted but it is yet in the excited state (the
second emission of the pair will be of type \( i \)). We have
\[ \frac{dN_i}{dt} = \gamma_i n_e + \Gamma_i n_i \] (3)
This expression shows that there are two contributions to the emission of pho-
tons of type \( i \). The first one, \( \gamma_i n_e \), is associated with the first emissions. It
represents the channel where the first emissions are of the type \( i \), denoting \( \gamma_i \)
the rate of first emissions in this channel. The second contribution refers to
non-entangled excited atoms of type \( i \) (atoms initially belonging to entangled
pairs where the first emission has been of the type \( j \neq i \)) that emit (is the
second emission of the pair). As the atoms are disentangled they emit with the
single-atom rate \( \Gamma_i \).

The variable \( n_i \) is an additional one in the problem, obeying the equation
\[ \frac{dn_i}{dt} = \gamma_j n_e - \Gamma_i n_i \quad i \neq j \] (4)
The number of non-entangled excited \( i \) atoms increases by first emissions in the
channel \( j \) (first emitted photons of type \( j \)) and decreases by second emissions
of type \( i \) leaving the atom in the ground state.

This equation can be easily solved. With the initial condition \( n_i(0) = 0 \)
(initially all the atoms are entangled) we obtain
\[ n_i(t) = \frac{n_0 \gamma_j}{\Gamma_i - \Gamma_f} (e^{-\Gamma_f t} - e^{-\Gamma_i t}) \] (5)

Using this expression we can rewrite Eq. (3) as
\[ \frac{dN_i}{dt} = n_0 \gamma_i e^{-\Gamma_i t} + \Gamma_i \frac{n_0 \gamma_j}{\Gamma_i - \Gamma_f} (e^{-\Gamma_f t} - e^{-\Gamma_i t}) \] (6)
On the other hand, from Eq. (2), the temporal variation of \( N_i \) can be also
expressed as
\[ \frac{dN_i}{dt} = n_0 \Gamma_i e^{-\Gamma_i t} \] (7)
Thus, we have obtained two expressions for the temporal variation of emitted photons of each type. The first one uses the temporal ordering of the emissions, whereas the second one is independent of it. The compatibility of both expressions demands them to be equal. The equality conditions leads to the four relations \((i, j = A, B)\)

\[
\gamma_j = \Gamma_f - \Gamma_i
\]  

(8)

and

\[
\gamma_i = \frac{\Gamma_i \gamma_j}{\Gamma_f - \Gamma_i}
\]  

(9)

It is immediate to obtain the solution:

\[
\gamma_i = \Gamma_i
\]  

(10)

and

\[
\Gamma_f = \Gamma_A + \Gamma_B
\]  

(11)

The emission rate of the first photon by the entangled system is the sum of the emission rates of the two atoms. This result agrees with the study of systems interacting simultaneously with two different weak noises, where the resulting decay rate is the sum of the separate rates \([9]\).

The above analysis is also valid for identical atoms. When the two atoms are identical the emission rate doubles that of the single atom, recovering the result presented in \([7]\) for bosons. For fermions we obtain the same decaying rate, \(\Gamma_f^F = 2\Gamma_i\).

In \([7]\) the analytical form of the emission rate was explained by an explicit calculation based on the matrix elements associated with the process. Here, we could follow a similar procedure. However, it would be almost identical and we shall not repeat it. We only present in the Appendix the distinctive features of the derivation for fermions and distinguishable atoms.

3 Emission in product states

In order to compare with the entangled case, we derive in this section the photon emission distributions of atoms in product states. The physical information we have about the physical system is the exponential form of the single atom emissions and the statistical independence of the two processes. From these facts we can deduce the first emission distribution.

We consider the probability of only one emission in a temporal interval \(\tau\) centered around \(t\), \(P(t, \tau)\). The temporal interval is dictated by the experimental window of detection. It can vary from experiment to experiment. As signaled before, a distinctive characteristic of non-entangled emitters is that the emissions are statistically independent. An immediate consequence of this independence is that we can have simultaneous emissions. For these events we
cannot define a first emitted photon and we must discard these pairs. We restrict our considerations to the post-selected set with only one emission in each interval. The unnormalized one emission probability is

\[ P^{un}(t, \tau) = P_A(t, \tau)\tilde{P}_B(t, \tau) + P_A(t, \tau)P_B(t, \tau) = P_A(t, \tau)(1 - P_B(t, \tau)) + (1 - P_A(t, \tau))P_B(t, \tau) = P_A(t, \tau) + P_B(t, \tau) - 2P_A(t, \tau)P_B(t, \tau) \] (12)

where \( P_i(t, \tau) \) is the probability of emission by the atom \( i = A, B \) in that interval and \( \tilde{P}_i(t, \tau) \) is the probability of not emission in the same interval. The superscript \( un \) denotes the unnormalized character of the distribution. The total probability of one emission is the probability of emission by atom \( A \) and not emission by \( B \) plus the probability of the same process with the roles of the two atoms interchanged. As the emissions by the two atoms are statistically independent, each one of the two terms is the product of the events probabilities.

The probabilities can be evaluated from the relation

\[ P_i(t, \tau) = \frac{N_i(t + \tau/2) - N_i(t - \tau/2)}{n_0} \approx \frac{\tau}{n_0} \frac{dN_i}{dt} = \tau \Gamma_i e^{-\Gamma_i t} \] (13)

that represents the ratio between the number of photons emitted in the interval and the total number of emitted photons (of type \( i \)), which can be approximated using Taylor’s rule.

We must normalize the probability distribution to unity, \( \int_0^\infty P(t, \tau)/\tau dt = 1 \), because we have assumed that the simultaneous detections have been removed. A trivial integration gives

\[ \int_0^\infty \frac{P^{un}(t, \tau)}{\tau} dt = 2 - \frac{2\tau \Gamma_A \Gamma_B}{\Gamma_A + \Gamma_B} = \alpha^{-1} \] (14)

Finally, the one-emission probability becomes

\[ P(t, \tau) = \alpha \tau (\Gamma_A e^{-\Gamma_A t} + \Gamma_B e^{-\Gamma_B t} - 2\tau \Gamma_A \Gamma_B e^{-(\Gamma_A + \Gamma_B)t}) \] (15)

The one-emission probability equals the first emission probability. From it we can easily obtain, with an argument similar to that used in Eq. (13), the distribution of first photon emissions. From \( P/\tau \approx (dN_f^p/dt)/n_0 \) and imposing the initial condition \( N_f^p(0) = 0 \) we have

\[ N_f^p(t) = n_0 \left( 1 - \alpha e^{-\Gamma_A t} - \alpha e^{-\Gamma_B t} + \alpha \frac{2\tau \Gamma_A \Gamma_B}{\Gamma_A + \Gamma_B} e^{-(\Gamma_A + \Gamma_B)t} \right) \] (16)

with the superscript \( p \) denoting that now we are dealing with atoms in product states.

In the limit of long times we have \( N_f^p(\infty) = n_0 \). Note that we obtain \( n_0 \) photon detections because we restrict our considerations to the post-selected ensemble. This is equivalent to have the total probability of only one detection
equal to one. If we had worked in the total ensemble including the simultaneous detections we would have obtained a number of first detections smaller than the initial number of excited atoms.

The first photon distribution for product states is not an exponential function but the sum of three different ones. In addition to the temporal dependence typical of single atoms, $\Gamma_A t$ and $\Gamma_B t$, we also have a dependence on the sum of both rates, $(\Gamma_A + \Gamma_B)t$. On the other hand, the distribution is also a function of the experimental temporal window $\tau$. Using different windows leads to different numbers of removed coincidence detections.

![Figure 1: Temporal dependence of the normalized photon emission distributions. The blue, red, black and green curves correspond respectively to $N_f/n_0$, $N_f^p/n_0$, $N_A/n_0$ and $N_B/n_0$. We represent the case $\Gamma_A = 1, \Gamma_B = 1.5$ and $\tau = 5/6$. The distributions are dimensionless and the time is in units of $\Gamma_A^{-1}$.](image)

Now, we can compare the first distributions for entangled (with the assumption of an exponential form) and product states. We present this graphical comparison in Figure 1. Typical values of atomic emission rates, $\Gamma$, are of the order of $10^9 \, s^{-1}$ (approximate value for the hydrogen atom). On the other hand, the temporal windows used in photon detection experiments range usually between $10^{-8} \, s$ and $10^{-10} \, s$. Note that our method is only valid for temporal windows obeying $\tau \Gamma_A \Gamma_B < \Gamma_A + \Gamma_B$ (Eq. 14).

We see that the first photon distributions for entangled and product states of excited distinguishable atoms differ. This result could be used (provided the experiments verify the exponential form for the entangled case) to directly
test the role of entanglement in spontaneous emission without the interfering contribution of the identity effects present in the case of identical particles.

4 Discussion

The ideas discussed in this paper are, in principle, testable with minor modifications of the arrangements in [5, 6]. In order to obtain the first-photon distributions for entangled states we only need to photodissociate molecules composed of identical fermionic atoms or distinguishable ones. As in [5, 6], the decaying products must be in excited states. This way we can test if they obey, as in the bosonic case, an exponential law. On the other hand, the distributions for product states could be obtained in a number of ways. For instance, we could even use two samples composed each one of a type of atom and compare the emission times.

The validity of the exponential law does not depend on the initial degree of entanglement of the two atoms. This result has been experimentally tested for identical bosonic atoms in [6] and has been theoretically derived here for fermionic and distinguishable ones. We do not have a smooth transition from the exponential law to a non-exponential one (but a sum of different exponentials) when the initial degree of entanglement decreases. There is a discontinuity in the emission distributions for the transition between entangled and non-entangled states. The type of decaying law ruling the process is only a function of the presence or absence of entanglement. We are dealing with a qualitative property of the system, not with a quantitative one.

The existence of an exponential law for the first emission of a composed system is also relevant from the fundamental point of view. In addition to the presence of non-classical correlations, the most distinctive conceptual characteristic of entangled systems is the loss of individuality of their components. If a system is entangled one cannot define the states of the components. Equivalently, one can ascribe a complete set of properties to the full system, but not to each one of the components (see [10, 1] for an excellent discussion of this point in the context of identical particles, but that can be easily translated to distinguishable ones). Closely related arguments can be developed in our case. We describe excited individual atoms as single emitters obeying an exponential law. The exponential law for the first emission of the two-atom system shows that the complete system, although composed, behaves like a single emitter. In contrast, when the two atoms are in product states, the complete system cannot be viewed as a single emitter. When the atomic pair is entangled a composed system has a property usually associated with single entities, reflecting a new aspect of the subtle relation between the parts and the whole in entangled systems.

As signaled in the introduction, the experiments of the type [5, 6] provide the first demonstration of disentanglement by spontaneous emission. Similar
experiments with identical fermionic atoms and distinguishable ones would pro-
vide a verification of the phenomenon for these types of atoms. In all the cases 
we expect an exponential decaying law for the entanglement. This behavior 
contrasts with that described in [8] where the decay occurs, for some values of 
the problem parameters, in a finite time. This difference can be explained by 
the different contexts associated with both cases. In [8] the excited atoms are 
placed in two cavities, whereas in our approach the atoms are in free space (only 
interacting with the vacuum electromagnetic field).

Appendix

Identical fermions. For fermions we can follow the presentation done in [7] 
for bosons. The two-atom initial state (just after the photodissociation in the 
case of a decaying molecule) is

\[ |\Psi_{F}^{0}\rangle = N_{F}^{0} (\Psi(x,y) - \Psi(y,x))|e>_{1}|e>_{2} \]  

The indices 1 and 2 denote the two fermionic atoms and the spatial variables 
x and y their center of mass (CM) position. \( \Psi \) is the CM wave function and 
\( |e>_{i}, i = 1, 2 \) refers to the excited internal state of the atoms. The minus sign 
between the CM wave functions reflects the antisymmetrization of the full state. 
The normalization coefficient is

\[ N_{F}^{0} = \frac{1}{(2 - 2Re(<\Psi(x,y)|\Psi(y,x)>))^{1/2}} \]  

After the first emission the two-atom state becomes

\[ |\Psi_{F}^{f}\rangle = \frac{1}{\sqrt{2}} (\psi(x)\phi(y)|g>_{1}|e>_{2} - \psi(y)\phi(x)|e>_{1}|g>_{2}) \]  

Because of the disentanglement inherent to the emission, the CM wave functions 
are product ones [7]. \( |g>_{1} \) denotes the ground state of the atoms.

Now, we can obtain the emission rate by evaluating the transition matrix 
element \( <\Psi_{F}^{f}|U|\Psi_{F}^{0}> \), with \( U \) as the evolution operator. This can be done 
by following step by step the derivation for bosons in [7]. It is immediate to 
see that the only difference is the sign between the wave functions (and in the 
normalization coefficient): it is positive for bosons and negative for fermions. It 
is also straightforward to realize that we reach the relation \( \Gamma_{F}^{f} = 2\Gamma_{i} \) if the CM 
wave function (previous to antisymmetrization) is antisymmetric

\[ \Psi(x,y) = -\Psi(y,x) \]  

This contrasts with the case of bosons, where the CM wave function (previous 
to symmetrization) must be symmetric. Note that for fermions the wave func-
tion must be antisymmetric all the time between the preparation and the first
emission. At the initial time it is an initial condition. For instance, if the atoms are generated by dissociation of a molecule it is natural to assume that being the atoms identical fermions the exchange effects lead to an initial antisymmetric state. The subsequent free evolution of the atoms preserves the antisymmetric character of the initial state. In effect, using the propagator $G$ we can write the wave function at times $t > 0$ as

$$\Psi(x, y, t) = \int d^3x_s d^3y_s G(x, y, t; x_s, y_s, 0)\Psi(x_s, y_s, 0) \quad (21)$$

For free evolution, the Schrödinger equation and, consequently, the propagator are symmetric. The simultaneous interchange $x \leftrightarrow y$ and $x_s \leftrightarrow y_s$ shows that the wave function at $t$ remains antisymmetric.

We conclude that the relation $\Gamma^f = 2\Gamma_i$ imposes a precise symmetry on the wave function (previous to antisymmetrization). Without that relation the symmetry properties of the wave function would remain undetermined.

**Distinguishable atoms.** When the atoms are distinguishable, the initially entangled state $\Phi(x, y)|e >_A |e >_B$ can evolve into the disentangled states $\psi(x)\phi(y)|g >_A |e >_B$ or $\psi_s(x)\phi_s(y)|e >_A |g >_B$. The two channels for the first emission are distinguishable because the emitted photons have different frequencies. Then we must add probabilities instead of probability amplitudes. Following the steps in [7] we have that the probability in each channel equals its emission rate and, finally, we obtain $\Gamma_f = \Gamma_A + \Gamma_B$.  

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