Entanglement, identity and disentanglement in two-atom spontaneous emission

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Abstract
Two recent experiments studying the potential effect of entanglement on the emission properties of excited atoms produced in molecular photodissociation have been interpreted in conflicting ways. We present a theoretical analysis of the problem, showing that the experimental results can be explained by a combination of three processes: entanglement, exchange effects associated with the identical nature of the atoms and disentanglement by spontaneous emission. According to our approach these experiments provide the first verification of the phenomenon of disentanglement by spontaneous emission.

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1 Introduction
Two recent experiments have explored the potential role of entanglement in the emission properties of pairs of excited hydrogen atoms produced in the photodissociation of \( H_2 \) molecules. In the original experiment [1], the coincidence time spectra of the emitted photons were measured. The authors concluded that the decay time constant of the spectra was dependent on the degree of entanglement in the system. Later [2], it was signaled that the above argument cannot be considered conclusive because the method of evaluation of the decay constant is based on the independence of the two emission processes, a controversial assumption for entangled systems. In that paper it was also suggested that the correct description should be based on a method of photon detection able to distinguish the temporal ordering of the two emission processes. A setup fulfilling that condition has been presented in [3]; where the emission and detection times can be measured, determining the temporal ordering of the two events. Those authors concluded, in marked contrast with [1], that the emission effects are not dependent on entanglement.
We present in this paper a theoretical analysis of the problem aimed at settling the question. During the analysis we realize that entanglement effects are actually present in the problem, but they alone cannot explain the complete phenomenon. Two other ingredients, exchange effects and disentanglement by spontaneous emission, must enter into the physical picture. The argument supporting this conclusion is simple: the compatibility between the emission distributions with and without temporal ordering imposes a precise relation between the emission rates of the pair of atoms and that of a single isolated atom. That relation leads, via some simple calculations, to the conclusion that the first photon emission depends on entanglement and identity and the second on the process of disentanglement by spontaneous emission [4, 5].

In order to clarify the problem further from the conceptual point of view we discuss, following well-known arguments in entanglement theory, how some measurable properties of the two-atom system depend on the non-separable character of the initial state and others do not. To be concrete, the temporally ordered distributions are dependent, whereas those without temporal ordering and the coincidence time spectra are not.

The phenomenon of disentanglement by spontaneous emission [4, 5] enters into the physical picture of the problem providing an elegant explanation of the behavior of the second photon emission. Conversely, setups of the type of [1] and [3] experimentally verify the phenomenon. This is a relevant result because it is its first direct demonstration. Disentanglement by spontaneous emission is a particular instance of the more general process of disentanglement induced by the environment (sometimes taking place in a finite time, then being denoted entanglement sudden death). Environment-induced disentanglement was first experimentally tested in [6] using an optical arrangement. Reference to later tests and developments can be found in [5]. However, to our knowledge, the loss of entanglement associated with spontaneous emission has not been experimentally verified.

2 Photon emission distributions

In the experiments in [1] and [3] $H_2$ molecules are photodissociated producing pairs of hydrogen atoms in excited states. Two detectors are placed at fixed directions outside the cell containing the molecules, and the arrival times at both positions are registered. They are denoted $t_1$ and $t_2$ (events registered at detectors 1 and 2). In [1] only the distribution $t_1 - t_2$ is measured. In [3] the synchrotron clock signal is also recorded. With this additional record one can also determine the times of the first ($t_f$) and second ($t_s$) photon detections.

We denote by $N_f(t)$ and $N_s(t)$ the number of first and second emitted photons recorded between the photodissocation ($t = 0$) and $t$. Consequently, the total number of emitted photons registered in that time interval is

$$N(t) = N_f(t) + N_s(t)$$ (1)
If we denote by \( n_0 \) the number of photodissociated molecules at the initial time the numbers of first and second photons are [2]

\[
N_f(t) = n_0(1 - e^{-\Gamma_f t})
\]

and

\[
N_s(t) = N_f(t) - n_0 \frac{\Gamma_f}{\Gamma_s - \Gamma_f} (e^{-\Gamma_f t} - e^{-\Gamma_s t})
\]

with \( \Gamma_f \) and \( \Gamma_s \) the first and second emission rates.

The total number can also be expressed in terms of the number of photons registered at each detector, \( N(t) = N_1(t) + N_2(t) \). Note that we have \( N_1(t) = N_2(t) \) because each emission process takes place in a random direction and for a large number of repetitions of the experiment both distributions become statistically similar. This property has been experimentally verified in [3]. This result can also be written as \( N_i(t) = N(t)/2 \) with \( i = 1, 2 \).

The experiment in [3] shows that the distributions \( N_i \) have the form \( N_i = n_0(1 - e^{-\Gamma t}) \) and, consequently

\[
N(t) = 2n_0(1 - e^{-\Gamma t})
\]

with \( \Gamma \) the emission rate for a single atom or, equivalently, of atoms in product states.

It is immediately seen that the three distributions are compatible when the conditions

\[
\Gamma_f = 2\Gamma
\]

and

\[
\Gamma_s = \Gamma
\]

hold. Naturally, this is the value obtained by the fits of the first and second emission curves [7].

We represent the three detection patterns graphically. We have done the previous presentation in terms of the total number of photons of a type detected between the initial time and \( t \). It can also be done, as in [3], using the distributions of photons of each type detected at a given time \( t \). For instance, for the first photons the number of detections in a short interval \( \Delta t \) around time \( t \) is \( N_f(t + \Delta t) - N_f(t) \approx n_0 \Gamma_f e^{-\Gamma_f t} \Delta t \). Then the normalized number of detections per unit time at time \( t \) is \( n_f(t) = (N_f(t + \Delta t) - N_f(t))/n_0 \Delta t \approx \Gamma_f e^{-\Gamma_f t} \). We represent the three distributions in Fig. 1. The detection patterns coincide with the count patterns in [3].

We devote the rest of the paper to understanding the physical meaning of the two conditions (5) and (6).
Figure 1: Temporal dependence of the normalized number of detections per unit time of photons at each detector \( (n_i, i = 1, 2, \text{black line}) \), first photons \( (n_f, \text{blue line}) \), and second photons \( (n_s, \text{red line}) \). The time and detection numbers are represented in units of \( \Gamma^{-1} \) and \( \Gamma \), respectively, with \( \Gamma^{-1} = 1.6 \times 10^{-9} \text{s} \).

3 Evaluation of the first photon emission rate

The emission rates can be evaluated from the matrix elements associated with these transitions. Just after the photodissociation, which we take as the initial time \( (t = 0) \), the state of the two atoms is

\[
|\Psi_0> = N_0 (\Psi(x,y) + \Psi(y,x)) |e>_{A} |e>_{B} \tag{7}
\]

with \( N_0 \) the normalization coefficient

\[
N_0 = \frac{1}{(2 + 2 \text{Re}(<\Psi(x,y) | \Psi(y,x)>))^{1/2}} \tag{8}
\]

The indices \( A \) and \( B \) refer to the two atoms. The coordinates in the wave function of the \( A \) and \( B \) atoms are denoted, respectively, \( x \) and \( y \). We represent the internal dynamics of the atoms by the kets \( |e> \) and \( |g> \), which refer to the excited and ground states. The spatial Center of Mass (CM) dynamics is ruled by the wave function \( \Psi(x,y) \). The scalar product in the last expression represents the overlapping of the two atoms.

The form of the initial state reflects two important physical characteristics of the system. On the one hand, the two dissociated atoms are identical bosons.
We must symmetrize the two-boson state. As initially they are very close, the spatial overlapping between them is large, and exchange effects can be important. On the other hand, the non-factorizable form of the spatial wave function indicates that the two atoms are entangled. Here, we follow the criterion introduced in [8] and [9] (see also [10]) to characterize entanglement in systems of identical particles: the state is separable if and only if $\Psi_0$ can be obtained by symmetrization of a factorized product of two orthogonal states or it is the product of the same state for the two particles. If $\Psi(x,y)$ is non-separable, the criterion shows that $\Psi_0$ is an entangled state. The fact that $\Psi(x,y)$ is non-separable can be easily understood from the physical point of view. At the classical level, the energy and momentum conservation lead to perfect correlations between the positions and velocities of the products of a decaying process. When we move to the quantum realm we expect that some (nonperfect) correlations will persist. The spatial state of the two atoms must be entangled (similarly for the momentum representation). See [11] for a simple discussion of the quantum correlations.

The complete two-atom and electromagnetic field initial state reads

$$|\Phi_0> = |\Psi_0> |0>_{EM}$$

with the field in the vacuum state.

The first photon emission leads to the new state

$$|\Phi_f> = |\Psi_f> |1>_{EM}$$

with the field changing from the vacuum to the one photon state, and the two atoms to

$$|\Psi_f> = \frac{1}{\sqrt{2}} (\psi(x)\phi(y)|g>_A |e>_B + \psi(y)\phi(x)|e>_A |g>_B)$$

This form of the atomic state is dictated by the disentanglement process associated with the emission. It is well-known [4, 5] that initially excited entangled states become disentangled after spontaneous emission. This result was obtained without explicitly taking into account the identical character of the emitters. Here, we assume that the result remains valid when the excited atoms are identical. For our initial state, where all the entanglement of the system is associated with the non-separability of the wave function, the disentanglement leads to the evolution $\Psi(x,y) \rightarrow \psi(x)\phi(y)$. According to the criterion in [8] and [9] state $\Psi_f$ is not entangled (the atoms separate after the photodissociation, leading to a negligible overlap, becoming $\psi$ and $\phi$ orthogonal).

Once we have determined the initial and first emission states, we can evaluate the probability of this transition. The associated matrix element is given by

$$\mathcal{M}_f = \langle \Phi_f | \hat{U}(t) | \Phi_0 >$$

5
with $\hat{U}$ the evolution operator, which can be expressed in the form $\hat{U} = \hat{U}_{EM} \otimes \hat{U}_{at}$, with the subscript $at$ referring to the atomic part. In addition, as the internal and CM parts are decoupled the atomic part can be written as $\hat{U}_{at} = \hat{U}_{CM} \otimes \hat{U}_{int}$. Moreover, as there is no interaction between the two atoms and they are identical (both internal operators are equal) we have $\hat{U}_{int} = \hat{U}_{int}^{(A)} \otimes \hat{U}_{int}^{(B)} = \hat{U}_{int}^{s} \otimes \hat{U}_{int}^{s}$, with $\hat{U}_{int}^{s}$ the single-atom internal-part operator.

Using all the above expressions we can write the matrix element as

$$M_f = \frac{N_0}{\sqrt{2}} <1|\hat{U}_{EM}|0>_{EM} <g|\hat{U}_{int}^{s}|e> <e|\hat{U}_{int}^{s}|e> \times 2(\psi(x)\phi(y)|\hat{U}_{CM}||\Psi(x,y) > + \psi(x)\phi(y)|\hat{U}_{CM}||\Psi(y,x) >)$$

(13)

The coefficient 2 in the second line of the above equation comes from the repetition of the terms after the interchange $x \leftrightarrow y$ ($A \leftrightarrow B$).

In order to continue our calculation we note the symmetric character of the initial wave function in our arrangement:

$$\Psi(x,y) = \Psi(y,x)$$

(14)

In effect, the Schrödinger equation for the system is symmetric with respect to the variables of the two particles. This is clear for the non-interaction terms. We also assume that the process of dissociation leading to two bosonic atoms must be ruled by a symmetric interaction. Thus, the initial wave function (previous to the symmetrization associated with the bosonic character of the particles) must be symmetric. The archetypical example of a symmetric wave function describing the decaying process is the Gaussian one $\Psi(x,y) \sim \exp(- (x+y)^2/X^2)$, with $X$ measuring the width of the distribution. This form assures the condition $x_{det} \approx -y_{det}$, representing the quantum correlations between position detections. More elaborate Gaussian distributions have been used in the study of dissociation. For instance, in [12] the authors resort to double Gaussian functions.

Relation (14) has two important consequences. On the one hand, the scalar product under the normalization condition reduces to $<\Psi(x,y)|\Psi(y,x) > = 1$ because we assume $\Psi$ to be normalized to unity. Then the normalization condition becomes $N_0 = 1/2$. On the other hand, the two spatial matrix elements in Eq. (13) are now equal.

The matrix element becomes $M_f = \sqrt{2}M_s = <\psi(x)\phi(y)|\hat{U}_{CM}||\Psi(x,y) >$. In this expression $M_s = <1|\hat{U}_{EM}|0>_{EM} <g|\hat{U}_{int}^{s}|e>$ is the single particle matrix element for the excited-ground atom transition times the vacuum-one-photon transition of the electromagnetic field. We have used the relation $<e|\hat{U}_{int}^{s}|e> = 1$ for the atom that does not undergo the transition.

From the matrix element we can derive the emission rates. In each repetition of the experiment the emission will take place in a random direction and we must take all these possible directions into account. We denote by $\{\psi, \phi\}$ the set of
all possible final wave functions. \( \mathcal{P} = \langle \psi | < \psi | \hat{U}_{CM} | \Psi > \) is the probability amplitude for the evolution \( \Psi \rightarrow \psi \phi \) of the initial wave function into each one of the members of the set. As the set is complete (the initial wave function must necessarily evolve into one of its members) we have \( \int_{\{\psi, \phi\}} D\psi D\phi |\mathcal{P}|^2 = 1 \), representing the functional integral the sum over all the possible final states. \( |\mathcal{P}|^2 \) and \( D\psi D\phi \) are respectively the probability density and the volume element in the space \( \{\psi, \phi\} \). Now, we can evaluate the emission rate which reads \( \Gamma_f = \int_{\{\psi, \phi\}} D\psi D\phi |M_f(\psi, \phi)|^2 \), where \( M_f(\psi, \phi) \) denotes the matrix element for each \( \psi, \phi \) in the set. Note that we add probabilities instead of amplitudes because the alternatives of transition to \( (\psi, \phi) \) and to any other pair of final wave functions, denoted \( (\psi_*, \phi_*) \), are distinguishable. We can understand this distinguishability by noting that every pair \( (\psi, \phi) \) defines a point in the functional wave-function space. Different points in this space can be distinguished. We can reach the same conclusion in a more technical way by comparison with the normal space. The states associated with different points in the normal space (in the spatial representation) are orthogonal. Similarly, the states representing different points in the functional space must be orthogonal and, consequently, distinguishable. Adding probabilities we easily obtain

\[
\Gamma_f = 2|M_s|^2 \int_{\{\psi, \phi\}} D\psi D\phi |\mathcal{P}|^2 = 2|M_s|^2 = 2\Gamma
\]  

(15)

The last step in this equation can be justified by translating the above two-atom argument to a single atom. Introducing the set of possible final wave functions for a single atom after spontaneous emission and repeating step by step the argument for two excited atoms, we have that \( \Gamma = |M_s|^2 \).

Two ingredients have entered into our derivation of this fundamental relation, entanglement and identity. The initial state \( \Psi_0 \) is entangled. It is simple to see that if the initial state is not entangled the value of \( \Gamma_f \) becomes different from \( 2\Gamma \). We demonstrate this property in the Appendix (Property 1). Thus, the initial presence of entanglement is a necessary condition to derive Eq. (5). The other ingredient is the identity of the two excited atoms. The initial and final states have been symmetrized because the atoms are identical. This symmetrization process is fundamental to obtain the results derived here. Without the symmetrization, that is, treating the atoms as distinguishable, we would obtain a different value for the first emission rate (Property 2 in the Appendix). Entanglement and bosonic identity are necessary conditions for the derivation.

It must be remarked that the disentanglement present in the spontaneous emission does not play a fundamental role at this stage. In order to justify this statement we show in the Appendix (Property 3) that Eq. (5) is also recovered when the final state is entangled.
4 Evaluation of the second photon emission rate

After the first emission the state of the system is $\Phi_f$. The atomic state after the second emission changes to

$$|\Psi_s> = N_s(\psi_s(x)\varphi(y) + \psi_s(y)\varphi(x))|g> |g>$$

with the normalization coefficient

$$N_s = \frac{1}{(2 + |\psi_t_s|\varphi>)^1/2}$$

where $<\psi_t_s|\varphi>$ is the overlap between the two atoms at $t_s$. The subscript $t_s$ denotes the time of the second emission.

During the interval between the first and the second emissions the wave functions evolve freely, changing from $\psi$ and $\phi$ to $\psi_{t_s}$ and $\phi_{t_s}$. After the second emission $\phi_{t_s}$ changes, due to the recoil, to $\varphi$.

The full atoms-electromagnetic-field state is

$$|\Phi_s> = |\Psi_s>|2>_{EM}$$

with two photons in the field.

The second emission probability can now be evaluated from the matrix element $M_s = <\Phi_s|\hat{U}|\Phi_f>$, whose explicit form is

$$M_s = \sqrt{2}N_sM_s(<\psi_{t_s}|\hat{U}^{s,fr}_{CM}|\psi>+<\psi_{t_s}|\hat{U}^{s,fr}_{CM}|\phi>+<\psi_{t_s}|\hat{U}^{s,fr}_{CM}|\varphi>)$$

with $\hat{U}^{s}_{CM}$ the single-particle CM evolution operator (the two atoms do not interact and the two-particle operator factorizes as $\hat{U}_{CM} = \hat{U}_{CM}^{s,fr}\hat{U}_{CM}^{s,re}$). The operators $\hat{U}_{CM}^{s,fr}$ describe the generic CM evolution of the atoms. In the case of the atom in the ground state it is a free evolution (the wave function only spreads) and we represent it as $\hat{U}^{s,fr}_{CM}$. For the excited atom the evolution operator describes two steps. Between $t_f$ and $t_s$ it evolves freely, $\hat{U}^{s,fr}_{CM}(t_f,t_s)$. Later, at $t_s$ it describes the recoil, $\hat{U}^{s,re}_{CM}(t_s)$. We incorporate both in the single expression $\hat{U}^{s,fr}_{CM} = \hat{U}^{s,re}_{CM}(t_s)\hat{U}^{s,fr}_{CM}(t_f,t_s)$. Using this notation we have $|\psi_{t_s}>=\hat{U}^{s,fr}_{CM}|\psi>$ and $|\varphi>=\hat{U}^{s,re}_{CM}|\phi>$. On the other hand, all the matrix elements associated with the internal variables and the electromagnetic field have been collected, as in the previous section, in $M_s$ (now with $<2|\hat{U}_{EM}|1>$). Combining all these expressions the matrix element reads

$$M_s = \sqrt{2}N_sM_s(1+<\psi_{t_s}|\varphi><\varphi|\psi_{t_s}>$$

To evaluate the overlap between the atoms at $t_s$ we recall that they evolve in almost-opposite directions, moving away from each other. In short times
they separate and the overlap is small. Quantitatively, according to [1], the separation is of the order of 100\(\mu\)m, justifying the above statement. Thus, we have \(\langle \psi_t | \varphi \rangle \approx 0\). Finally, we obtain
\[
M_s = M_s
\]
and we arrive at \(\Gamma_s = \Gamma\).

Two elements have determined this result, the disentanglement by spontaneous emission and the negligible overlap of the two atoms. With respect to the first element it must be stressed that if the state after the first emission were entangled, we would obtain a different result, as demonstrated by Property 4 in the Appendix. The second element is equivalent to neglecting the exchange effects at the time of the second emission, which in our case are represented by the second summand in Eq. (20).

The disentanglement after the first emission is also relevant for the experiment in [1]. After the first emission the state loses its initial entanglement and the coincidence time spectra cannot depend on it. We should have the same spectral distribution for any value of the initial entanglement of the excited hydrogen atoms. This is in marked contrast to the results in [1], where different spectra were obtained for different gas pressures (identified with different entanglement values in the sample). In the experiment in [3] the spectra were measured again for different pressure values, obtaining in all cases the same distribution (a decay ruled by the emission rate of single atoms) [7]. This result completely agrees with our approach and can be seen as a complementary verification of the disentanglement process.

5 Dependence on non-factorizability

We have shown that the condition \(\Gamma_f = 2\Gamma\) is strongly dependent on the non-separable (entangled and symmetrized) character of the initial state of the two atoms after the dissociation. In contrast, other properties of the system such as \(N_1\), \(N_2\), and \(N\) do not depend on it. This behavior is a novel manifestation of a property already described in entangled systems: nonfactorizability of the state can only manifest in two-particle properties (the values of the detection times of the two atoms are involved) of the system. If we consider one-particle properties (we do not need to consider both times jointly), we always obtain results compatible with product states.

Let us examine this point in detail. When we determine the distributions \(N_1\) and \(N_2\) we only care about their respective detectors. We do not need any information about the results in the other detector. In this case we cannot distinguish the two detection patterns from those obtained with \(2n_0\) single excited atoms (or \(n_0\) pairs of excited atoms in product states). In contrast, to determine which of the two photons in a single repetition of the experiment is the first one we must compare the two detection times. Now, the first and second detection
patterns are two-particle properties and differ from that obtained for a pair of independent excited atoms.

The correlation distribution between the two emissions analyzed in [1] is also a two-particle property as we must subtract the times determined at the two detectors ($t_1 - t_2$). However, in contrast with the first and second photon distributions discussed above, it does not show any dependence on the initial non-separable character of the states. This is so because the property we measure, $t_1 - t_2$, is only related to times subsequent to the first emission. After this emission the system is disentangled and the exchange effects are negligible. Consequently, during that period of time the system is in a product state. The nonseparability present in the system for times $t < t_1$ does not affect the subsequent behavior.

6 Discussion

Our analysis concludes, answering the initial question, that entanglement is actually a key ingredient in the explanation of experiments of the type in [1] and [3]. However, it is not the only physical ingredient involved in the problem. The identical nature of the two atoms and the disentanglement by spontaneous emission must also be taken into account to provide a complete picture of the underlying dynamics. These two elements were not considered in previous discussions. It is important to emphasize that these ingredients enter into the picture at different stages. Entanglement and identity act simultaneously before the first emission and disentanglement enters the scene at the time of that emission.

This work is a new example of the increasing interest in the role of entanglement in atomic and molecular systems. The characterization of entanglement is, in general, a difficult task [10]. Our approach has the advantage of easily identifying the type of entanglement present in the system. The form of the initial state, $\Psi_0$, indicates that the entanglement is associated with the spatial CM variables. Physically, this identification is a natural consequence of the strong correlations existing between the decaying fragments. This point of view contrasts with that expressed in [1], where the entanglement is between the different constituents of the two atoms (the two electrons and the two protons), taking into account the possible magnetic quantum numbers of the system (see Eq. (2) in [1]). The agreement of our results with those of the experiment in [3] shows that the relevant variables for the problem are the CM ones and that we do not need to be concerned about the complicated form of the connection between the different atomic constituents.

The process of emission by excited dissociation products is an illustrative example of entanglement and exchange effects acting at the same time. This example suggests that this process, and decaying phenomena in general, can be a powerful tool for the experimental study of the interplay between en-
tanglement and identity. The subject of entanglement in systems of identical particles has led to many discussions and still needs clarification. Measurement of the entanglement associated with the position and momentum correlations of the products of decaying processes seems to be a promising way to quantitatively study this question. In particular, if the products of the dissociation are fermions instead of bosons, we should expect different patterns. This extension to fermions could be an interesting line of research.

Finally, and this is the most important result from the practical point of view, we have shown that setups of the type in [1] and [3] provide an experimental verification of the process of disentanglement by spontaneous emission. As we indicated earlier it is, to the best of our knowledge, the first demonstration of the effect. This result widens the scope of the field of experimental disentanglement by including the case associated with spontaneous emission. It also opens the doors to explore other aspects of the field. For instance, one can study and test whether the process depends on the type of particles involved (distinguishable, bosonic or fermionic) or on the initial degree of entanglement.

Appendix

In this Appendix we demonstrate some properties used in the text.

**Property 1.** According to the criterion in [8] and [9] the form of a nonentangled initial state must be

$$|\Psi_{0}^{NE} > = N_{0}^{NE} (\chi(x)\xi(y) + \chi(y)\xi(x))|e >_A |e >_B$$

(22)

with $N_{0}^{NE} = (2 + 2 \langle \chi | \xi > |^2)^{-1/2}$, denoting by $\langle \chi | \xi >$ the overlap of the two one-particle wave functions. In addition, $\chi$ must be either orthogonal to $\xi$ ($N_{0}^{NE} = 1/\sqrt{2}$) or equal to it ($N_{0}^{NE} = 1/2$).

Repeating the steps used to derive $M_f$ we obtain

$$M_f^{NE} = \sqrt{2} N_{0}^{NE} M_s \times$$

$$\left( \langle \psi | \hat{U}_{CM} | \chi > < \phi | \hat{U}_{CM} | \xi > + \langle \psi | \hat{U}_{CM} | \xi > < \phi | \hat{U}_{CM} | \chi > \right)$$

(23)

with $\hat{U}_{CM}$ the single-particle CM evolution operator introduced in Sec. IV. When $\chi$ is orthogonal to $\xi$, in the probabilities derived from the amplitude $M_f^{NE}$ we have interference terms between $\langle \psi | \hat{U}_{CM} | \chi > < \phi | \hat{U}_{CM} | \xi >$ and $\langle \psi | \hat{U}_{CM} | \xi > < \phi | \hat{U}_{CM} | \chi >$. In general this amplitude probability does not lead to the relation $\Gamma_f = 2\Gamma$. Only when $\chi = \phi$ do we obtain the correct value for $\Gamma_f$. However, the condition of equality of the two one-particle wave functions leads to the relations $x_{det} \approx y_{det}$, which shows that this particular case is not relevant for our dissociation problem.

**Property 2.** If we do not symmetrize the initial state it reads $|\Psi_{0}^{NS} > = \Psi(x,y)|e >_a |e >_b$. Note that we use lowercase letters $a$ and $b$ to label
the atoms instead of capital ones in order to show that now it is possible in some way to distinguish the particles (if the particles were indistinguishable it would be necessary to symmetrize). After the first emission the state can be $|Ψ_f^{NS}⟩ = ψ(x)φ(y)|g >_a |e >_b$ or $|Ψ_f^{NS}⟩ = ψ(x)φ(y)|e >_a |g >_b$ with equal probabilities 1/2. The probability amplitude for the first channel is $M_{NS} = <Ψ_f^{NS}|\hat{U}|Ψ_0^{NS}>$, and the expression for the second channel is similar.

To obtain the transition probability we must sum both probabilities with equal weights 1/2: as we have assumed that the two atoms are somehow distinguishable, it is in principle possible to distinguish which one has emitted the photon and both emission alternatives become distinguishable; we must add probabilities instead of probability amplitudes. Now, using the same argument as in the text, we have $M^{NS} = M^{NS} = M_s$ and, finally, $Γ^{NS} = Γ$.

**Property 3.** When the final state is entangled we have

$$|Ψ_f⟩ = \frac{1}{\sqrt{2}}(|Ψ(x, y)⟩|g >_A |e >_B + |Ψ(y, x)⟩|e >_A |g >_B)$$

Repeating the steps in Sec. III and, in particular, invoking the symmetry condition of $Ψ$ we obtain

$$\tilde{M}_f = \sqrt{2}M_s <\tilde{Ψ}(x, y)|\hat{U}_{CM}|Ψ(x, y)>$$

Performing a summation over the continuous set of possible final states $Ψ$ we recover Eq. (5).

**Property 4.** We assume that the state after the first emission remains entangled and, consequently, is given by $Ψ_f$. If there is not disentanglement after the first emission, we can safely assume that there is also not disentanglement after the second one and the atomic state will remain entangled,

$$|Ψ_s⟩ = \tilde{N}_s(Ψ_s(x, y) + Ψ_s(y, x))|g >_1 |g >_2$$

with $\tilde{N}_s = (2 + 2Re(<Ψ_s(x, y)|Ψ_s(y, x)>) )^{-1/2}$.

The transition matrix element in this case is

$$<Ψ_s|\tilde{U}|Ψ_f> = \sqrt{2}\tilde{N}_sM_s × ( <Ψ_s(x, y)|\hat{U}_{CM}|Ψ(x, y)> + <Ψ_s(x, y)|\hat{U}_{CM}|Ψ(y, x)> )$$

When evaluating the probabilities from the matrix element we find interference terms between the spatial matrix elements in the above equation. Moreover, there is a dependence, through the normalization factor, on the final overlap. These two results prevent a relation of the type $|<Ψ_s|\hat{U}|Ψ_f>|^2 = Γ$. Another form of reaching the same conclusion is to consider the particular case of both wave functions, $Ψ$ and $Ψ_s$, symmetric. For this case we would obtain $|<Ψ_s|\hat{U}|Ψ_f>|^2 = 2Γ ≠ Γ$. 12
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