Dressed atom versus exciton polariton: From Rabi oscillations to the Fermi Golden rule

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Abstract

We rederive the dressed atom and the exciton polariton within the same framework to make clear that their difference only comes from the number of electrons available for photoexcitations. Using it, we analytically show how the time dependence of the photon number transforms from an oscillating behavior (at the stimulated or vacuum Rabi frequency) to an exponential decay (identical for atom and semiconductor) when the excited state lifetime decreases. Although the matter ground state is in both cases coupled by monochromatic photons not to a continuum but to a discrete state, this decay yet follows a kind of Fermi Golden rule. The energy conservation it contains, is however conceptually different.

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A very large amount of our physical understanding comes from matter-photon interaction, this matter being either a dilute set of atoms as in atomic physics, or a dense arrangement as in solid state physics. In all cases, the photons induce a coupling between electronic levels. It is known that for a single two-level atom, the electron Rabi oscillates \[1\] between its two possible levels while for solids, the photons are absorbed with a transition rate given by the Fermi Golden rule \[2\]. The reason invoked for this behavior change is the energy distribution of the states coupled by photons to the matter ground state: In usual solids, their energies are close to a continuum, so that Rabi oscillations are destroyed by interferences. These interferences in fact lead to an overall exponential decay controlled by excited states which have an energy close to the initial one. The energy conservation appears through a delta "function", \( \delta(\epsilon) = \sin(\epsilon t/2)/\pi \epsilon \), which has a width, \( t^{-1} \) (if \( \hbar = 1 \)), equal to \[3\] the characteristic energy of the Heisenberg uncertainty principle.

A case of special interest is however the interaction of photons with semiconductors, because the hole left in the valence band can form bound states with the photocreated electron, called excitons. In this case, the states coupled by photons to the semiconductor ground state do not form a continuum, as in usual solids, because the photocreated exciton has a well defined center of mass momentum, its components being the ones of photon along the exciton free directions. Consequently, the standard reason for having a photon absorption given by the Fermi Golden rule, can not be invoked in the case of exciton formation.

In addition, long ago, Hopfield has shown that photons and excitons form mixed states, called polaritons \[4\]. Being eigenstates of the coupled semiconductor-photon hamiltonian, they cannot decay, so that no photon absorption can result from the semiconductor-photon interaction alone, whatever the strength of this interaction is. According to Hopfield, the experimentally observed absorption is due to additional couplings of the exciton component of the polariton to external reservoirs \[5\].

Cases in which the polariton picture has to be used and cases in which photons are barely absorbed according to the Fermi Golden rule, are said to correspond to "strong" and "weak" couplings \[6, 7\]. Although the change from one regime to the other has been correctly related \[8\] to the strength of the semiconductor-photon interaction compared to
the exciton broadening induced by external couplings, to our best knowledge, no direct derivation of a photon absorption for transitions to a discrete state has been given. More precisely, how, starting from the Hopfield’s polariton, is constructed the exponential decay corresponding to a photon absorption when the ground state is coupled to one state only? In particular, is the characteristic time of this decay really given by the Fermi Golden rule, as implicitly assumed by everyone, since the initial state is not coupled to a continuum, so that the energy conservation appearing in this rule cannot be due to destructive interferences between final states close in energy?

We wish to stress that the problems linked to strong versus weak couplings are usually discussed in the context of photon emission from excitons in a quantum well, either in free space or in a microcavity [9, 10]. The physics is then totally different. The possible momenta of photons emitted by an exciton with a momentum \( \mathbf{Q}_{//} \) along the well, are \( \mathbf{Q}_{//} + \mathbf{q}_z \) with \( E_x + \hbar^2 \mathbf{Q}_{//}^2/2M_x = \hbar \nu |\mathbf{Q}_{//} + \mathbf{q}_z| \). For a quantum well in vacuum, there is a continuous set of \( \mathbf{q}_z \) which fulfill this relation. The exciton being then coupled to a continuum of photons, the photon emission transition rate has to be given by the Fermi Golden rule. Because, in the Fermi Golden rule, the exciton-photon interaction enters at lowest order only, these excitons seem to have a ”weak coupling” with photons. On the opposite, when the well is in a microcavity, the possible cavity modes \( \mathbf{q}_z \) are discrete. The exciton being no more coupled to a continuum, there is no reason for the photon emission to follow the Fermi Golden rule. And indeed, in order to explain the observed results, the exciton-photon coupling has be treated exactly, through the polariton, so that these excitons seem to have a ”strong coupling” with photons [11]. Such a photon emission has to be contrasted with the photon absorption considered here, in which one photon \( \mathbf{q} = \mathbf{q}_{//} + \mathbf{q}_z \) is coupled to one exciton only, its momentum being \( \mathbf{q}_{//} \); As the final state is then discrete, the reason for a possible regime in which the Fermi Golden rule is valid, is a priori not obvious.

In this letter, we address this quite fundamental question of semiconductor physics: why can we use the Fermi Golden rule for photon absorption in a semiconductor since the final state is discrete.

In order to tackle this question, we follow a dressed atom approach [12, 13]: we consider \( N \) photons interacting either with a single atom or a semiconductor, initially in their
ground state. We look for the time evolution of the photon number, $N(t)$, in terms of the matter-photon coupling, the excited state detuning and its broadening. The problem is actually more tricky for semiconductors than for atoms, because, the valence band having very many electrons, all photons can a priori be transformed into excitations so that the dimension of the coupled subspace is not (1+1), as for a dressed atom, but (1+N). By using generalized polariton operators dressed by the exciton external couplings, it is however possible to obtain an analytical expression of the photon number time evolution, also in the semiconductor case.

We recover that, when the excited state broadening $\gamma$ is small, $N(t)$ oscillates with the vacuum Rabi frequency, $\Omega_1$, in the case of semiconductor and the stimulated one, $\Omega_N=\Omega_1\sqrt{N}$, in the case of atom. On the opposite, when $\gamma$ is large, $N(t)$ for semiconductor and atom exhibits the same exponential decay, with a characteristic time given by a kind of Fermi Golden rule. This decay, which cannot be due to standard destructive interferences between Rabi oscillations of quasi-continuous states, has a quite different origin: The energy conservation it contains, is not the usual $\delta_t(\epsilon)$, but $\tilde{\delta}_\gamma(\epsilon) = \gamma/\pi(\epsilon^2+\gamma^2)$. This other delta ”function” has a $t$ independent width, $\gamma$. For large $t$, more precisely for $\gamma t \gg 1$, its width is in fact larger than the width, $t^{-1}$, of the standard rule, as fully reasonable since the energy uncertainty due to the excited state broadening $\gamma$ is then larger the one of the Heisenberg principle, the energy conservation being possible to enforce at the larger of the two.

1 Two-level atom dressed by photons

The presentation we give here of this quite well known problem [12, 13], allows to enlighten the similarities and differences between atom and exciton.

In a two-level atom, the photon transfers the electron from level 0 to level 1, leaving a hole in the level 0 (see fig.(1a)). Once in level 1, the electron can return to level 0 either by emitting the same photon or by relaxation such as the emission of fluorescent photons. The coupled atom-photon hamiltonian can be written as

$$H = \omega_p A^+ A + (\omega_x - i\gamma) B^+ B + (\mu^* B^+ A + h.c.)$$  \hspace{1cm} (1.1)

where $\omega_p$ is the photon energy, $\omega_x$ the energy difference between levels 1 and 0 and $\gamma =$
the broadening of level 1 induced by its relaxation. $\mu^*$ is the matrix element for the transformation of one photon into one excitation. $A^+$ creates one photon while $B^+$ creates one excitation. In the case of a single two-level atom, it is such that $B^+ \mid X^0\rangle = \mid X^1\rangle$, $B \mid X^1\rangle = \mid X^0\rangle$, while $B^+ \mid X^1\rangle = B \mid X^0\rangle = 0$ where $\mid X^1\rangle$ and $\mid X^0\rangle$ correspond to the electron in the levels 1 and 0, respectively.

Let us consider $\mid \psi_N(t = 0)\rangle = \mid N, X^0\rangle$ as initial state: the system has N photons, the atom being in its ground state. The atom-photon interaction couples this state to $\mid N-p, X^p\rangle$, within an irrelevant phase factor $e^{-i(N\omega_p+\Delta)t/2}$. The prefactors $a_N^{(p)}(t)$ are given by

$$a_N^{(0)}(t) = e^{-\gamma t/2} \left[ \cos(\tilde{\Omega}_N t/2) + i(\tilde{\Delta}/\tilde{\Omega}_N) \sin(\tilde{\Omega}_N t/2) \right] \quad (1.3)$$

$$a_N^{(1)}(t) = -ie^{-\gamma t/2} (2\mu^*/\sqrt{N}/\tilde{\Omega}_N) \sin(\tilde{\Omega}_N t/2) \quad (1.4)$$

in agreement with textbooks [13].

The photon number, equal to $\langle \psi_N(t) \mid A^+ A \mid \psi_N(t)\rangle$, thus reads for a two-level atom initially in its ground state

$$N_a(t) = N[P_N^{(0)}(t) + P_N^{(1)}(t)] - P_N^{(1)}(t) \quad (1.5)$$

where $P_N^{(p)}(t) = |a_N^{(p)}(t)|^2$ are the probabilities for the atom to have no excitation (p=0) or one excitation (p=1). Using eqs.(1.3,1.4), these probabilities are

$$P_N^{(1)}(t) = \frac{e^{-\gamma t}}{2} \frac{\Omega_N^2}{|\Omega_N^2|} \left[ ch(tO_N') - \cos(tO_N) \right] \quad (1.6)$$

$$P_N^{(0)}(t) = e^{-\gamma t} \left[ I_N^+ \cdot ch(tO_N') - D_N \cdot sh(tO_N') + \bar{I}_N \cdot \cos(tO_N) - D' \cdot \sin(tO_N) \right] \quad (1.7)$$

where we have set $\tilde{\Omega}_N = O_N + iO'_N$, $\tilde{\Delta}/\tilde{\Omega}_N = D_N + iD'_N$ and $I_N^{(\pm)} = (1 \pm |\tilde{\Delta}^2/\tilde{\Omega}_N^2|)/2$. 

$1/2\tau$
Figure 1: Possible couplings of a two-level atom, (a), or a semiconductor, (b), induced by the presence of N photons, when the excited states have external relaxation (wavy lines).

2 Semiconductor polariton

The problem of photon interaction with a semiconductor has similarities with the one of a two-level atom except that, after the transformation of one photon into one exciton, it is a priori possible to excite another electron from the valence band, and, again, another one, until all the photons are transformed into excitations. This of course implies that the photon number N is small compared to the total number of valence electrons. In order to possibly describe this phenomena in a simple way, this also implies that N is not too large to end with a set of N excitons, which can be considered as non-interacting, i.e., all at the same energy. This is valid if the Coulomb and Pauli interactions between N excitons are small, i.e., if $N(a_x/L)^d \ll 1$, $a_x$ and $L$ being the exciton and sample size, while $d$ is the space dimension. In this limit, the excitons can be taken as non-interacting bosons. The coupled semiconductor-photon hamiltonian then reads as eq.(1.1), where $B^+$ is now the ground state exciton creation operator. It is such that $B^+ | X^n \rangle = \sqrt{n+1} | X^{n+1} \rangle$ for any $n \geq 0$, with $| X^n \rangle = (n!)^{-1/2} (B^+)^n | v \rangle$ being the normalized semiconductor state with $n$ boson-excitons.

If we consider an initial state similar to the one of a two-level atom, namely $| \psi_N(t = 0) \rangle = | N, X^0 \rangle$, with N photons and the semiconductor in its ground state, the hamiltonian couples it to $| N - 1, X^1 \rangle, \ldots, | 0, X^N \rangle$, with possible relaxation to states like $| N - 1, X^0 \rangle$ if the exciton has external couplings, i.e., if $\gamma \neq 0$ (see fig. (1b)).
In order to obtain the time evolution of $|\psi_N(t)\rangle$, we can either diagonalize $H$ in this $(N+1)$ subspace or better, rewrite $H$ in a diagonal form. As for $\gamma \neq 0$, $H$ is not hermitian, this diagonalization is less trivial than the one for textbook polaritons because, $H$ and $H^+$ being different, they have different eigenstates. It is however easy to check, just by replacing, that $H$ given in eq.(1.1), also reads
\[ H = E^{(-)}_1 \alpha^+_k \alpha_b + E^{(+)}_1 \beta^+_k \beta_b \] (2.1)
where $E^{(\pm)}_1$ are the eigenenergies for $N=1$ photon, given in eq.(1.2). The operators $\alpha^+_k$ and $\beta^+_k$ are defined as
\[ \alpha^+_k = \frac{(\bar{\Delta} + \bar{\Omega}_1)A^+ - 2\mu^*B^+}{\sqrt{(\bar{\Delta} + \bar{\Omega}_1)^2 + \Omega_1^2}} \quad \beta^+_k = \frac{(\bar{\Delta} + \bar{\Omega}_1)B^+ + 2\mu A^+}{\sqrt{(\bar{\Delta} + \bar{\Omega}_1)^2 + \Omega_1^2}} \] (2.2)
while $\alpha_b$ and $\beta_b$ read as $\alpha_k$ and $\beta_k$ with $(\bar{\Delta}^* + \bar{\Omega}_1^*)$ replaced by $(\bar{\Delta} + \bar{\Omega}_1)$. For a finite exciton broadening, i.e., $\bar{\Delta} \neq \bar{\Delta}^*$, the operators $(\alpha^+_k, \alpha_b)$ and $(\beta^+_k, \beta_b)$ appearing in $H$ are not conjugate. They however are the ones which fulfill the commutation relations
\[ [\alpha_b, \alpha^+_k] = 1 = [\beta_b, \beta^+_k] \quad [\alpha_b, \beta^+_k] = 0 = [\beta_b, \alpha^+_k] \] (2.3)
so that $(H - E^{(-)}_1)\alpha^+_k|v> = 0$ while $<v|\alpha_b(H - E^{(-)}_1)|v> = 0$ and similarly for $\beta^+_k$ and $\beta_b$. The operators $\alpha^+_k$ and $\beta^+_k$ have thus to be seen as the creation operators for polaritons dressed by exciton relaxation in the ket space, while $\alpha_b$ and $\beta_b$ are the ones in the bra space.

To our best knowledge, the diagonal form of the coupled photon-exciton hamiltonian written in eq.(2.1), has not been given before. The generalized polariton operators in the bra and ket space it contains, are however of conceptual importance to possibly describe photon-exciton interaction with relaxation.

By writing the photon creation operator $A^+$ in terms of $\alpha^+_k$ and $\beta^+_k$, we can show, from the commutation relations (2.3) and the expression of the hamiltonian given in eq.(2.1), that the initial state $|N, X^0\rangle = (N!)^{-1/2}(A^+)^N|v\rangle$, with $|v\rangle$ being the exciton and photon vacuum, transforms into $|\psi_N(t)\rangle = |\Phi_N(1,1,t)\rangle$ with
\[ |\Phi_N(x,y;t)\rangle = (N!)^{-1/2}\left( A_{x,y,t}^+ \right)^N|v\rangle \] (2.4)
\[ A_{x,y,t}^+ = xa_1^{(0)}(t)A^+ + ya_1^{(1)}(t)B^+ \] (2.5)
within an irrelevant phase factor $e^{-i(\omega_p+\Delta/2)t}$. $a^{(0)}_1(t)$ and $a^{(1)}_1(t)$ are the prefactors appearing in the time evolution of a two-level atom in the presence of $N=1$ photon, as given in eqs.\textsuperscript{[1,3,14]}.

From this compact expression of $|\psi_N(t)\rangle$, it is easy to obtain the time evolution of the photon number $\langle \psi_N(t) | A^+ A | \psi_N(t)\rangle$. Indeed, as $A^+ A | \psi_N(t)\rangle$ is just $\varphi_x | \Phi_N(x,y;t)\rangle$ taken for $x=y=1$, this photon number is nothing but $(1/2)\partial_x \langle \Phi_N(x,y;t) | \Phi_N(x,y;t)\rangle |_{x=y=1}$. In order to get the norm of $|\Phi_N(x,y;t)\rangle$, we can note that

$$A_{x,y,t} \left( A^+_{x,y,t} \right)^N | v \rangle = N b_{x,y,t} \left( A^+_{x,y,t} \right)^{N-1} | v \rangle \quad (2.6)$$

where we have set

$$b_{x,y,t} = [A_{x,y,t}, A^+_{x,y,t}] = x^2 P_{1}^{(0)}(t) + y^2 P_{1}^{(1)}(t) \quad (2.7)$$

$P_{1}^{(0,1)}(t)$ being the probabilities given in eqs.\textsuperscript{[1,6,17]}, for $N=1$ photon. This shows that the norm of $|\Phi_N(x,y;t)\rangle$ is nothing but $b^{-N}_{x,y,t}$, so that the photon number for a semiconductor initially in its ground state, finally reads

$$N_{sc}(t) = (N/2) \left( b_{x,y,t} \right)^{N-1} \partial_x b_{x,y,t} |_{x=y=1} = N P_{1}^{(0)}(t) \left( P_{1}^{(1)}(t) + P_{1}^{(0)}(t) \right)^{N-1} \quad (2.8)$$

This nicely compact analytical result is valid for any time, any photon number, any detuning, any exciton broadening, and any photon-semiconductor coupling.

## 3 Time evolution of the photon number

The time evolution of the photon number for an atom or a semiconductor initially in their ground state are given by eqs.\textsuperscript{[1,3,2,8]}. These analytical expressions can be used to obtain the photon number for any given set of experimental conditions. We are just going here to discuss the limiting cases of importance for physical understanding.

(i) *In the absence of matter-photon coupling*, i.e., for $\Omega_1 = 0$, we have $P_{N}^{(1)}(t) = 0$ while $P_{N}^{(0)}(t) = 1$: the matter stays in its ground state, and the photon number stays equal to $N$, as expected.

(ii) *In the absence of excited state relaxation*, i.e., for $\gamma = 0$, the sum $P_{N}^{(1)}(t) + P_{N}^{(0)}(t)$ stays equal to 1. The photon number change $n(t) = N - \mathcal{N}(t)$ reduces to $P_{N}^{(1)}(t) = (\Omega_{N}^2/\omega_N^2)\sin^2(\omega_N t/2)$ in the case of atom and to $\mathcal{N}_{1}(t) = N(\Omega_1^2/\omega_1^2)\sin^2(\omega_1 t/2)$ in the
case of semiconductor, with $\omega_N^2$ being $\Omega_N^2 + \Delta^2$. At resonance, i.e., $\Delta=0$, we recover that for a dressed atom, the photon number oscillates, between $N$ and $(N-1)$, at the stimulated Rabi frequency $\Omega_N$. For a semiconductor, it oscillates, at the vacuum Rabi frequency $\Omega_1$, between $N$ and 0, so that all the photons can possibly be transformed into excitons. When the detuning increases, the frequency of these oscillations increases, while the photon number stays closer to $N$.

(iii) When the excited state relaxation is included, i.e., for $\gamma \neq 0$, the sum $P_N^{(1)}(t) + P_N^{(0)}(t)$ decreases with time. For $\gamma$ small, the Rabi oscillations are essentially damped, while with increasing $\gamma$, the decrease of $P_N^{(1)}(t) + P_N^{(0)}(t)$ ends by controlling the photon number change. For $\gamma \gg \Omega_N$, the expansion of eqs. (1.6, 1.7) in $\eta_N^2 = \Omega_N^2/(\gamma^2 + \Delta^2)$, shows that, as $\tilde{\Omega}_N \simeq \tilde{\Delta} + \tilde{\Delta}^* \eta_N^2/2$

$$P_N^{(1)}(t) \simeq (\eta_N^2/4)e^{-\gamma \eta_N^2 t/2} \quad P_N^{(0)}(t) \simeq \left(1 + \frac{\eta_N^2 \gamma^2 - \Delta^2}{2 \gamma^2 + \Delta^2}\right)e^{-\gamma \eta_N^2 t/2} \quad (3.1)$$

for $\gamma t \gg 1$. Using the above equations, we find that, for $\gamma \gg (t^{-1}, \Omega_N)$, the photon number of a two-level atom, given in eq. (1.5), tends to $N_a(t) \simeq Ne^{-\gamma \eta_N^2 t/2}$ while the one for a semiconductor, given in eq. (2.8), tends to $N_{sc}(t) \simeq Ne^{-N \eta_1^2 t/2}$. Since $\eta_N^2 = N \eta_1^2$, this shows that, in this limit, $N_a(t) \simeq N_{sc}(t) \simeq Ne^{-t/T}$ with $T$ given by

$$\frac{1}{T} = \frac{N \gamma \eta_N^2}{2} = 2\pi |\mu\sqrt{N}|^2 \delta_\gamma(\Delta) \quad (3.2)$$

$\delta_\gamma(\Delta) = \gamma/\pi(\Delta^2 + \gamma^2)$ being a delta "function" of width $\gamma$.

We see that, for $\gamma$ large compared to $t^{-1}$ and $\Omega_N$, the photon number for a two-level atom and a semiconductor, both behave as $Ne^{-t/T}$, the characteristic time of this exponential decay following a kind of Fermi Golden rule: $\mu^*\sqrt{N}$ is indeed the matrix element of the matter-photon coupling between the initial state, $N$ photons and the matter in its ground state, and the excited state, $(N-1)$ photon and one excitation. The energy conservation it contains is however conceptually different from the standard one: It is linked to the energy uncertainty induced by the excited state broadening, not the one coming from the Heisenberg principle, as in the usual Fermi Golden rule. The ground state of an atom or a semiconductor being not coupled to a continuum but to a discrete state, the exponential decay of the photon number does not come from destructive interferences between Rabi oscillations, but from the broadening of the excited state itself, in a non trivial way: It is not a bare $e^{-\gamma t}$, but results from the interplay between $\gamma$ and $\Omega_N$. This
decay only exists when the excited state energy uncertainty $\gamma$ is much larger than the strength of the stimulated matter-photon coupling. If we see the broadened excited state as a continuous set of states, all at the same energy, we can say, in a very crude way, that the delta "function" $\hat{\delta}_\gamma$, which appears as a multlicative factor in the decay rate, "selects" a part of this broadened excited state "continuum", in the same way as the delta "function", $\delta$, of the usual Fermi Golden rule, "selects" the part of the continuum coupled to the ground state.

Conclusion

We have derived the time evolution of the photon number in the presence of an atom or semiconductor in their ground state, using the same framework. This led us to introduce generalized polariton operators dressed by the exciton finite lifetime, which differ in the bra and ket spaces. They actually are of conceptual importance for the study of photon-semiconductor interaction in the presence of relaxation.

When the photons couple the ground state to a discrete excited state which has an infinite lifetime, we recover that not only a single two-level atom, but also a macroscopic semiconductor have Rabi oscillations, at the stimulated frequency, $\Omega_1\sqrt{N}$, for atoms, and at the vacuum one, $\Omega_1$, for semiconductors.

On the opposite, when the excited state lifetime is small, the photon number has the same exponential decay for atom and semiconductor, with a characteristic time given by the Fermi Golden rule. This rule can indeed be extended to transitions to a discrete state, provided that the discrete state is highly broadened compared to the stimulated matter-photon coupling, and that the "energy conservation" this Fermi Golden rule contains, is enforced at the scale of the excited state broadening.

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