Incoherent Mollow triplet

E. del Valle and F. P. Laussy

1 School of Physics and Astronomy, University of Southampton, SO17 1BJ, Southampton, United Kingdom
2 Walter Schottky Institut, Technische Universität München, Am Coulombwall 3, 85748 Garching, Germany

(Applied: July 12, 2010)

A counterpart of the Mollow triplet (luminescence lineshape of a two-level system under coherent excitation) is obtained for the case of incoherent excitation in a cavity. Its analytical expression, in excellent agreement with numerical results, pinpoints analogies and differences between the conventional resonance fluorescence spectrum and its cavity QED analogue under incoherent excitation.

Mollow [1] discovered a striking type of spectral shape in the resonance fluorescence problem, where an atom is irradiated by a strong laser beam. The celebrated Mollow triplet [2], that results from transitions between atomic states that are dressed by the coherent light field, has since been a testbed of nonlinear optics. It stands as one of the fundamental spectral shapes of light-matter interaction, maybe second only to the Rabi doublet.

Although the Mollow triplet is rooted in quantum physics and bears much quantum features itself, it arises from a fully classical light field. Its Hamiltonian, in the rotating frame of the laser and at resonance, simply reads $H_L = \Omega_L (\sigma + \sigma^\dagger)$, with $\Omega_L$ the laser intensity and $\sigma$ the only quantum operator, namely, the two-level system annihilation operator. Including the spontaneous decay of the emitter, in the Lindblad form $\mathcal{L}_\sigma(\rho) = (2\sigma \rho \sigma^\dagger - \sigma^\dagger \sigma \rho - \rho \sigma \sigma^\dagger)$, leads to a master equation $\partial_t \rho = i[\rho, H_L] + \frac{\gamma}{2} \mathcal{L}_\sigma(\rho)$ from which one obtains the famous Mollow triplet lineshape:

$$S_{\text{coh}}(\omega) = \frac{\gamma^2}{8\Omega^2_L} \left( \frac{\gamma^2}{\pi} \frac{\omega^2}{\delta^2} \right) \left( 1 + \frac{\gamma^2}{2\pi \delta^2} \right) + \frac{\gamma^2}{\pi^2} \left( \frac{\gamma^2}{\delta^2} + 4(\omega^2 - \Omega_L^2)^2 + \gamma^2 \Delta^2 \right).$$

(1)

It is composed of an elastic scattering peak, the Dirac $\delta$ function, and the triplet itself, with a central Lorentzian peak of full width at half maximum (FWHM) $\gamma$ and two satellite peaks at the symmetric positions $\pm \delta(\Omega_L)$ with Mollow splitting $R_L = \sqrt{(2\Omega^2_L)^2 - (\gamma/4)^2}$ and FWHMs $3\gamma/2$. This structure was observed a long time ago with atoms [3] and more recently also in a variety of solid state systems [4-8], with, as befits the above description, coherent excitation.

In this text we consider a close counterpart of this fundamental system, where the light field is initially fully quantized, and becomes continuous as a result of an incoherent and continuous pumping that feeds the system with a very large number of photons. This situation is realized—as for quantization of the light field—in cavity QED [9], where quanta of a trapped standing waves (the photons) can be brought to interact with an isolated emitter, and—as for the incoherent pumping—with semiconductor microcavities [10], where excitations are continuously poured into the system with no external coherence fed in by a driving field. The role of the emitter is, in this case, played by a quantum dot placed in the antinode of the microcavity field. In the cavity QED version of the resonance fluorescence physics, the system is described by the Jaynes-Cummings Hamiltonian (still at resonance): $H = g(a^\dagger \sigma + a \sigma^\dagger)$ with the cavity mode also quantized through the boson operator $a$. Cavity decay $\gamma_\sigma$ as well as incoherent pumping $P_\sigma$ are described like before with a master equation $\partial_t \rho = i[\rho, H_L] + \frac{\gamma_\sigma}{2} \mathcal{L}_\sigma(\rho)$ from which one obtains the following expression for the cavity fluorescence spectrum:

$$S_{\text{coh}}(\omega) = \frac{\gamma_\sigma^2}{8\Omega^2_L} \left( \frac{\gamma^2}{\pi} \frac{\omega^2}{\delta^2} \right) \left( 1 + \frac{\gamma^2}{2\pi \delta^2} \right) + \frac{\gamma^2}{\pi^2} \left( \frac{\gamma^2}{\delta^2} + 4(\omega^2 - \Omega^2_L)^2 + \gamma^2 \Delta^2 \right).$$

This expression is valid for low excitation regimes, when the classical oscillator term can be dropped and $g \gg \gamma_\sigma$. For higher order of amplification, other mechanisms can also result in a similar behaviour of Rabi splitting collapse without entering the quantum nonlinear regime [13]. Quantum features are generally better observed when probing the quantum emitter, rather than the cavity, whose close connections with the classical oscillator tend to surface rapidly and dominate strongly. The theoretical description, which is straightforward in the low excitation regime even when solving the system exactly [12], becomes computationally demanding when the lasing regime is approached, but good approximations can be sought [14].
The expressions Eqs. (3) for the populations have a clear physical meaning, that can be discussed in terms of two parameters: the “cavity feeding”, $F_a = P_σ/(2\gamma_a)$, and the “dot feeding”, $F_σ = (P_σ - \gamma_a)/κ_σ$, efficiencies. At low pump, but still enough to be beyond the quantum regime \[12\], i.e., $\gamma_a \ll P_σ \ll κ_σ$, the cavity population increases linearly with pumping, with a half occupied dot. This is the most effective region for accumulation of photons in the cavity (the so-called one-atom laser \[13\]), with little disruption from incoherent processes. At smaller efficiency $F_σ$, the dot occupation also increases linearly with pumping, quenching the linear increase of the cavity population. $F_σ$ represents, therefore, the degree to which the dot pumping succeeds in populating the dot itself, against the coherent exchange of population that feeds the cavity with efficiency $F_σ$. These expressions are thus valid until the dot population is fully inverted, at $P_{\text{max}} \approx κ_σ$, then, the self-quenching dominates the dynamics, emptying the cavity that goes to a thermal state. The maximum population of the cavity, max($n_a$) $\approx g^2/(2\gamma_a^2)$, is reached at the intermediate rate $P_σ \approx κ_σ/2$. This identifies the regime of interest for the observation of the Mollow triplet, where the cavity field is intense ($n_a \gg 1$) and coherent (with a Poissonian photon distribution, $T[n] = e^{-n_a}n_a^n/n!$ and $g^2(1)$):

$$γ_a \ll g < P_σ < κ_σ.$$  (4)

Now that we have a good and analytical description of the populations, we turn to the optical emission spectrum, that we show can be obtained in equally good approximations. The dot emission reads $n_σπS_{\text{inc}}(ω) ≡ \Re \int_0^∞ (σ(0)|σ(τ)|e^{iωτ}dτ$. We compute the two-time correlator $⟨σ(0)|σ(τ)⟩$ in two steps: first, we solve the master equation in the steady state, finding the density matrix elements $ρ_{m,i,n,j}$ (for $m, n ∈ N$ and $i, j \in \{0,1\}$, photon and exciton indexes, respectively). For the range of parameters of interest, we show that they can be analytically expressed only in terms of the photon distribution $T[n]$. Second, we apply the quantum regression formula.

### 1. Steady state density matrix

We consider only elements that are nonzero in the steady state: the populations $p_i[n] = ρ_{m,i,n,j}$ with $i = 0,1$, corresponding to the probability to have $n$ photons with $(p_1)$ or without $(p_0)$ exciton, and the off-diagonal terms $q_i[n] = 3(ρ_{n,0,n-1,1})$, corresponding to the coherence between the states $|n,0⟩$ and $|n-1,1⟩$. The master equation now reads:

$$\partial_t p_i[n+1] = D_{\text{phot}} \{p_i[n+1]\}$$

$$- P_σ p_i[n+1] - 2g√n + 1q_i[n+1],$$

$$\partial_t q_i[n+1] = D_{\text{phot}} \{q_i[n+1]\}$$

$$- P_σ \{T[n] - p_1[n]\} + 2g√n + 1q_i[n+1],$$

where we have separated the photonics dynamics into a superoperator $D_{\text{phot}}$. Given that it is much slower than the dot dynamics, one can solve the steady state ignoring $D_{\text{phot}}$ \[10\]. The photon distribution, $T[n] = p_0[n] + p_1[n]$. 

Through the strong-coupling with the dot, resulting in striking variations from the case where it is provided by an external laser. We now describe them analytically.

**Mollow regime** — Whereas only one parameter (intensity) fully describes the light in Mollow’s description, the Jaynes-Cummings picture requires from the start to take into account an infinite number of correlators between the fields, that we can however relate to each other \[12\]: $⟨a^†n a^n⟩ = \frac{i^{2n}}{2^n} (2n)! (a^n a^n)$ and $⟨a^†n−1 a^n−1 σ|σ⟩ = [P_σ(a^n−1 a^n−1) − γ_0(a^n a^n)]/[P_σ + γ_0(n−1)]$ (all others being zero in the steady state). From this follows a first relation for the populations of the modes, $n_σ = ⟨σ|σ⟩$ and $n_a = ⟨a^†a⟩$, namely $n_σ = (P_σ − \gamma_a n_a)/P_σ$. This also allows to obtain a self-contained equation for $⟨a^{1n}⟩$:

$$⟨a^{1n}⟩ = \frac{P_σ P_{n−1/2} (a^n a^n − 2γ_0 a^n a^n)}{1 + 2P_σ/(2σ − 2γ_0)} + \frac{2P_σ n_σ + n_{n−1}}{P_σ + κ_σ},$$  (2)

where $κ_σ = 4g^2/γ_0$ is the Purcell rate of transfer of population from the dot to the cavity mode. This recurrence equation allows to compute $⟨a^{1n}⟩$ for all $n$ as a function of $n_a$ only. The solution for $n = 0$ gives a good approximation for the region where the cavity field behaves classically:

$$n_a \approx \frac{P_σ}{2γ_0} \left(1 - \frac{P_σ - γ_0}{κ_σ}\right)$$

$$n_σ \approx \frac{1}{2} \left(1 + \frac{P_σ - γ_0}{κ_σ}\right).$$  (3)

The quality of this approximation is seen in Fig. 1 where it is compared with the exact solution, computed numerically \[12\]. The second order coherence function $g^2(1)$ also admits a closed-form expression (not given here but plotted in Fig. 1) which is unity in good approximation. The expressions Eqs. (3) for the populations have a clear physical meaning, that can be discussed in terms of two parameters: the “cavity feeding”, $F_a = P_σ/(2γ_0)$, and the “dot feeding”, $F_σ = (P_σ - γ_0)/κ_σ$, efficiencies. At

![FIG. 1: (Color online) Exact numerical solution (points) and their analytical approximation, Eqs. (3) (lines), for $n_a$ (blue/circles), $n_σ$ (brown/squares) and $g^2(1)$ (purple/triangles), for $γ_0 = 0.1g$, as a function of pumping $P_σ/g$. The analytical solutions become unphysical when $P_σ = κ_σ$ (here at 40g), where $n_a = 0, n_σ = 1$ and $g^2(1)$ diverges. They are very good approximations in the region of interest, Eq. (3).](image-url)
remains unperturbed during the excitation and interaction with the dot. Equations \(5\) then admit solutions in terms of \(T[n]\), i.e., \(p_0[n] \approx \frac{\kappa_a(n+1)}{P_s + 2\kappa_a(n+1)} T[n]\), and \(q_i[n] \approx \frac{-2\sqrt{n}}{P_s + 2\kappa_a(n+1)} T[n-1]\) where \(\kappa_a = 4g^2/P_s\) is the Purcell rate of transfer of population from the cavity to the dot. Our approximations of large intensities imply \(n + 1 \approx n\).

2. Two-time correlator and spectrum — The two-time correlator can be expressed as a sum \(\langle \sigma^1(0)\sigma(\tau) \rangle = \sum_{\tau=0}^{\infty} Q[n](\tau)\), where \(Q[n]\) and other functions are defined through the quantum regression formula by coupled differential equations (\(n \geq 0\)):

\[
\partial_\tau Q[n] = D_{\text{photon}}\{Q[n]\} - \frac{P_s}{2} Q[n] + ig(\sqrt{n}S_1[n] - \sqrt{n+1}S_0[n+1]),
\]

\[
\partial_\tau S_0[n+1] = D_{\text{photon}}\{S_0[n+1]\} - P_s S_0[n+1] + ig(\sqrt{n+1}V[n+1] - \sqrt{n}Q[n]),
\]

\[
\partial_\tau S_1[n] = D_{\text{photon}}\{S_1[n]\} + P_s \{X[n] - S_1[n]\} - ig(\sqrt{n+1}V[n+1] - \sqrt{n}Q[n]),
\]

\[
\partial_\tau V[n+1] = D_{\text{photon}}\{V[n+1]\} - \frac{P_s}{2} V[n+1] + ig(\sqrt{n}S_0[n+1] - \sqrt{n+1}S_1[n]).
\]

They are, like for the single-time dynamics, separated into a slow photonic dynamics embedded in a superoperator \(P_{\text{photon}}\) that is \(\tau\)-independent in good approximation, and a fast exciton and coupling dynamics that we can solve analytically. Moreover, we have introduced the steady state function \(X[n] = S_0[n](0) + S_1[n](0)\), in analogy with \(T[n]\). The initial conditions in Eq. [6], are the steady state values \(S_0[n+1](0) = iq[n+1], S_1[n](0) = 0, Q[n](0) = p_1[n]\) and \(V[n+1](0) = 0\) (therefore, \(X[n] = iq[n]\)). After some long, but straightforward algebra, we can find the expression for \(Q[n](\tau)\) in terms of \(p_0, q_1\) and \(q_i\), which, in turn, are expressed in terms of the statistics \(T[n]\) (as shown previously). This allows to compute a closed-form solution for \(\langle \sigma^1(0)\sigma(\tau) \rangle\), which is however lengthy and not worth writing here. Its main physical features are to reveal that each term in the sum over \(n\), accounts for the 4 possible transitions between the dressed states in the Jaynes-Cummings rungs \(n + 1\) and \(n\), as in the spontaneous emission embedded [12]. The first rung, or linear regime, is given by \(n = 0\) and consists of only the two transitions of the Rabi doublet. Other rungs give rise to a generalization of the Rabi frequency in the nonlinear regime, the \(n\)th rung inner and outer Rabi frequencies: \(R_{O,I}[n] = \sqrt{g^2(\sqrt{n+1} \pm \sqrt{n})^2 - (P_s/4)^2}\).

In the Mollow triplet regime \(P_s > g\), all the peaks positioned at the inner frequencies collapse at the centre (including the Rabi doublet) giving rise to a single central peak. Outer peaks remain split at frequencies \(\pm R_O[n] \approx \pm 4g^2n -(P_s/4)^2\).

The spectrum obtained with the previous derivations can be further simplified for the range of parameters in Eq. [6], to give a compact analytical expression. First, one considers only the coefficients with leading terms in \(n\), making use again of \(n + 1 \approx n\). Furthermore, due to the Poissonian statistics, only rungs with \(n\) close to \(n_a\) contribute significantly to the spectra allowing the substitution \(n \rightarrow n_a\) in \(Q[n]\). The sum over \(n\) simplifies thanks to the normalization of the distribution function: \(\sum_n T[n] = 1\). Finally, we neglect terms related to \(\gamma_a\) before those related to much larger rates, \(P_s\) and \(\kappa_a\), i.e., we write the spectrum for these three rates only, through the substitution \(g^2 = \kappa_a\gamma_a/4\), and then simply set \(\gamma_a \rightarrow 0\). This results in the expression for \(S_{\text{inc}}(\omega)\) in terms of \(P_s\) and \(\kappa_s\) only:

\[
S_{\text{inc}}(\omega) = \frac{P_s}{\kappa_s} \frac{P_s}{\kappa_s + P_s} \delta(\omega) + \frac{1}{2\pi} \frac{P_s}{(P_s/2)^2 + \omega^2} + \frac{1}{\pi} \frac{P_s}{\kappa_s + P_s} (3\kappa_s - P_s) \omega^2 - (\kappa_s - 3P_s)P_s^2 A(\omega) + \frac{4\omega^4 - P_s(4\kappa_s - 9P_s)\omega^2 + \kappa_s^2P_s^2}{4\kappa_s + P_s}. (7)
\]

This is our main result. The structure of the lineshape is the same as that of its coherent counterpart, Eq. [1]: a Dirac \(\delta\) function from the elastically scattered laser light superimposed to a triplet. The central Lorentzian peak has the same weight 1/2 but with FWHM given by the pump, \(P_s\). The two satellite peaks sit at \pm R\(_O\) with Mollow splitting:

\[
R_O = \frac{P_s}{4} \sqrt{8\kappa_s/P_s - 9} (8)
\]

and FWHM 3\(P_s/2\). The excellent agreement of our formula with exact numerical results [12] [17] is shown in Fig. 2(a), where we superimpose in dashed red the numerical computation to, in solid red, the analytical expression Eq. [7]. Note the elastic peak in the numeric as a very narrow central line.

Despite their similar structure, the lineshapes are intrinsically of a different nature, as can be seen mathematically when reduced to their simplest, dimensionless expression, where they depend only on one parameter.
two types of triplets: that shows the fundamental discrepancies between the theory of the coherent one. We obtain an expression attempting the description of the incoherent system with broadening of the dot includes other factors such as the dot population, which under malizes the rungs of the Jaynes-Cummings ladder, and affected by the effect of incoherent pumping, that renor- the satellite lineshapes differ considerably, being strongly underlying structures bear much similarities. However, indeed, that the central peak is the same in both cases, and when the broadening of the dot includes $P_\sigma$. In this way, we attempt the description of the incoherent system with the theory of the coherent one. We obtain an expression that shows the fundamental discrepancies between the two types of triplets:

$$S_{\text{coh}}(\omega) = \frac{P_\sigma}{\kappa_\sigma} \delta(\omega) + \frac{1}{2\pi} \frac{P_\sigma}{\left(\frac{P_\sigma}{\kappa_\sigma}\right)^2 + \omega^2}$$

$$- \frac{1}{\pi} \frac{P_\sigma \omega^2 - (2\kappa_\sigma - 3P_\sigma)P_\sigma^2}{4\omega^4 - P_\sigma(4\kappa_\sigma - 9P_\sigma)\omega^2 + \kappa_\sigma^2 P_\sigma^2}. \quad (9)$$

Comparing this expression with Eq. (7), one can see, indeed, that the central peak is the same in both cases, as well as the position and broadening of the satellite peaks (third terms have the same denominator), so the underlying structures bear much similarities. However, the satellite lineshapes differ considerably, being strongly affected by the effect of incoherent pumping, that renor- the rungs of the Jaynes-Cummings ladder, and other factors such as the dot population, which under coherent excitation shows opposite behaviour to that of Eq. (3): $n_{\text{coh}}^\sigma \approx \frac{1}{2}(1 - P_\sigma/\kappa_\sigma)$. The shapes of these peaks are shown in more details in Fig. (b), where they are plotted (in dashed) together with the whole triplets, in the coherent (thin black) and the incoherent (thick red) cases.

Finally, Fig. (3) shows the natural experimental configuration to demonstrate the new character of nonlinear spectroscopy in microcavities under incoherent pumping, and to contrast it with its coherent counterpart. Increasing pumping, one sees that in the coherent case (upper panel), the triplet is better resolved, with a larger splitting, while in the incoherent case (lower panel), the satellites overlap with the central line as a result from pumping that splits them sublinearly, Eq. (9), and also increases their broadening. The two phenomenologies, despite their deep interconnections and common features, are strikingly different and the evidence of the new one should pose no problem even on qualitative grounds.