Two-photon correlations in detuned resonance fluorescence

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Abstract

We discuss two-photon correlations from the side peaks that are formed when a two-level system is driven coherently, with a detuning between the driving source and the emitter (quasi-resonance fluorescence). We do so in the context of the theories of frequency-resolved photon correlations and homodyning, showing that their combination leads to a neat picture compatible with perturbative two-photon scattering that was popular in the early days of quantum electrodynamics. This should help to control, enhance and open new regimes of multiphoton emission. We also highlight a way to evidence the quantum coherent nature of the process from photoluminescence only, through the observation of a collapse of the symmetry of the lineshape accompanied by a surge of its intensity of emission. We discuss several of our results in the light of recent experimental works.

1. Introduction: historical developments of resonance fluorescence

Resonance fluorescence is the ‘drosophila’ of quantum optics. It is the simplest yet rich enough problem to capture many of the key considerations on light–matter interaction, from quantization of the light field up to the riddle of measurements and observations in quantum mechanics. It consists of driving optically a two-level system (e.g. an atomic transition, a spin, a semiconductor exciton, a superconducting qubit, etc) with a coherent wave that has the same or a close frequency than that of the spontaneous emission of the emitter. The platform is both of great fundamental interest for the understanding of basic aspects of quantum theory as well as from a technological perspective for its prospects as a quantum emitter, not only as a single-photon source but also in an unsuspected regime of multiphoton emission.

The multiphoton problematic turns out to have been central to theoretical modeling since the early days. As a basic problem of light–matter interaction, the origin of resonance fluorescence goes back to the dawn of quantum electrodynamics (which itself can be dated with Dirac [1]). Pioneering contributions include those of Weisskopf [2, 3], for scattering off the ground and excited state of an atom, respectively. A major and recurrent work still relevant today, in the low-driving regime, is that of Heitler [4] who reported his analysis directly in the 3rd edition of his textbook ‘the Quantum Theory of Radiation’, in a chapter (absent in previous editions) titled ‘resonance fluorescence’ (§20), where he shows that the lineshape of radiation is provided by the driving source itself as opposed to the natural lineshape of the emitter. His analysis follows in essence from the conservation of energy $\delta(\omega - \omega_L)$ of the scattering process so that each photon from the source gets scattered at the energy with which it impinged on the atom, whence the result. The process is actually not as trivial as it looks, with Heitler already observing that the radiation occurs ‘as if two independent processes, an absorption and a subsequent emission, took place’, with the atom ‘remembering’ (his term) ‘before the emission which quantum it has absorbed’. Seen in this way, it is less obvious why the spontaneous emission character of the emitter plays no role. It also brings forward that a two-photon process is involved in this scattering. In fact, as we discuss further below, it turns out to be of central importance for the photon statistics of resonance fluorescence in this low-driving regime [5]: the $\delta$-shaped scattered light itself is uncorrelated and emission becomes antibunched only if also detecting the weak—but at the two-photon level, essential—incoherent part of the spectrum, that is indeed
spread spectrally and originating from multiphoton events. This incoherent part is however very small in intensity as compared to the Rayleigh peak. This led to some confusion in the literature [6, 7] that we hope to have clarified [8] (see also [9]): although the incoherent peak vanishes at low intensities in one-photon observables, its contribution rules the photon-statistics (a two-photon observable).

Multiphotons are even more prominent at higher driving. In a nonlinear quantum-optics framework, several degenerate photons from the driving source (which we shall from now on refer to as the ‘laser’) can be redistributed by the emitter at different energies so as to produce a more complex spectral shape, known today to be a triplet with ratio of peak heights 1:3:1 and with a splitting given by the laser intensity. The problem was initially regarded as that of the competition between spontaneous and stimulated emission, with a feeling shared by many theorists of the time that spontaneous emission required quantization of the field for a correct treatment. The exact nature of this spectral shape was the topic of some controversy, in particular it took part in the debates initiated by Jaynes according to whom the light field should not be quantized and his neoclassical theory (relying on a nonlinear feedback from the radiation field back to the emitter) should be used instead. The neoclassicists were also experts in solving the quantized version of a problem to provide what they assumed were the wrong QED predictions, which is how, famously, the Jaynes–Cummings model [10] arose. In this framework, the quantized version of resonance fluorescence by Stroud [11] (part of Jaynes’ team) but at the one-photon level, led to incorrect results, such as a 1:2:1 ratio of the peaks, in contrast to a semiclassical treatment by Mollow [12] which was not quantizing the light field but obtained, for the first time, the correct lineshape. For this reason, this characteristic result, which was originally referred to as the AC stark effect, became known as the ‘Mollow triplet’ (it seems that Zoller [13] is the first to have used this denomination). Stroud et al mention in their conclusions that their analysis is ‘incomplete in one important aspect’: the truncation to one-photon emission. While they recognize that ‘in the real physical case there will be a cascade emitting many spontaneous-emission photons’, they believed that antibunching would make such successive emissions from a quantized model justifying their approximation. Further support for Stroud et al’s view came from Smithers and Freedhoff [14] who claimed to have included multiphoton effects and yet still arrive at the same (incorrect) result as Stroud et al, but this was disputed by Carmichael and Walls [15] who observed that in their treatment, ‘Smithers and Freedhoff have not managed to include true photon cascades but have simply followed a series of sequential one-photon emissions’. Convincingly, by truncating their quantized version to single-photon transitions, Carmichael and Walls showed how they downgraded Mollow’s spectrum to one with the same attributes as Stroud’s. Mollow’s result, it must be emphasized, although not quantizing the light field, is not part of the neoclassical theory, which treats spontaneous emission as a continuous process as opposed to quantum jumps, leading to still further departures between the various models. The reason why Mollow got the correct result is interesting: ironically, it turns out that the semiclassical model is equivalent to a multiphoton quantized model, and that multiphoton effects are responsible for the lineshape, although luminescence is a single-photon observable. This has been recognized and commented by various people at the time but the most insightful discussion seems to be that of Mollow himself, in his 1975 follow-up paper [16]. He carried on such a fully-quantum treatment, including multiphoton contributions of all orders, and showed that the c-number description of the laser does not spoil the fully-quantum nature of the problem, as long as multiphoton effects are included. These correspond to the back-reaction in Jaynes’ neoclassical theory and to what a modern treatment would qualify as virtual photons, i.e. the atom re-absorbing photons that it has just emitted. In this context, the problem can be understood as a scattering one. In the words of Mollow [16] ‘the individual multiphoton scattering processes [...] are concealed from view, with only their accumulated effect exhibited’. We will come back to this important observation later on. Another key contribution to that approach of resonance fluorescence comes from Cohen–Tannoudji [17] and his co-workers [18–21], who provided both the so-called dressed-at and a perturbative scattering pictures. From this viewpoint, spontaneous emission is not deemed central but is relegated to a secondary plane. Instead, dressing the atom is considered first, yielding new eigenstates for the system with an exact (all-order) treatment of the light–matter coupling. Then spontaneous emission is brought back to the dressed atom cascade down its energy diagram and in this process replace photons from the laser with fluorescence photons from the atom. This remains the most picturesque way to understand the spectral shape of the Mollow triplet and can also account for a lot, although not all, of the phenomenology of correlations between the peaks.

We now turn to the detuned resonance fluorescence, i.e. when the driving laser is close to but not right at the energy of the two-level system. In the earlier treatments, such as from Heitler, exact resonance implied divergence and one of Heitler’s inputs was precisely to dampen the system [22] so as to arrive at a physical response for exact resonance. In a modern quantum-optical, master equation approach, resonance is actually simpler while off-resonance comes with additional subtleties but also with several advantages. Not least is the fact that detuning helps the splitting of what always remains a triplet. In fact, the first neatly resolved Mollow triplet was out of resonance [23] and if one would stick to resonance, it would then be apparently the improved setup of Walthers that has reported the first resonant Mollow triplet, albeit in a conference proceedings [24] (the
first report of an even better triplet in a leading journal came however only a few months later [25]). Detuning also weakens the efficiency of the coupling and what determines whether one is in the Heitler (low-driving) or Mollow (high-driving) regime in this context is an interesting question that we address elsewhere [26]. As was already described by Mollow in his magnum opus [12], when the two-level system is detuned from the laser, one gets at low driving a doublet with a peak centered on the atom and the other peak shifted by twice the detuning, with the Rayleigh-scattered laser sitting in between, that is to say, one always has a triplet in the non-detuned case. This is shown in figure 1(e). Here it must be appreciated that the two side peaks are vanishing with \( \Omega_\sigma \to 0 \) as compared to the coherent peak in the center, with a ratio \( 8\Delta_\sigma^2/\left(\gamma_\sigma^2 + 4\Delta_\sigma^2\right) \) for their respective intensities, i.e. most of the emission comes from the central peak, just as the case of resonance where the Lorentzian foothill is dwarfed by the Rayleigh peak. Here too, however, two-photon observables, such as photon statistics, are ruled by the interplay of the coherent and incoherent emission [27], regardless of their relative intensities. The only, but striking, difference is that this central incoherent peak has now split in two. With increasing driving, a central incoherent peak grows at the laser position, becoming of identical height with the side peaks when \( \Omega_\sigma \approx \Delta_\sigma / \sqrt{2} \) (exactly so in the limit \( \gamma_\sigma \ll \Delta_\sigma, \Omega_\sigma \)) and for higher-still driving, converging towards a resonant Mollow triplet, since detuning now becomes negligible as compared to driving. There is therefore a smooth transition between the various cases [26].

A triplet structure comes with an obvious opportunity to correlate photons from the various peaks. This was highlighted by Cohen–Tannoudji and Reynaud [19] and implemented by Aspect et al [28]. At low-enough driving but with detuning to maintain a multi-peak structure, the perturbative treatment is appealing and provides a remarkable and compelling picture for the triplet, that is shown in figures 1(a)–(d). Here again, scatterings between the states of the system give the best phenomenological explanation for the observed doublet (on the one hand) surrounding the central peak (on the other hand). This was discussed in detail by Dalibard and Reynaud [20]. In addition to the Rayleigh peak, which elastically scatters the laser photon and thus pins the peak at \( \omega_\sigma \), the scheme also involves two virtual states and a two-photon transition. One of the photons originates from the excited-to-ground state transition with energy \( \omega_\sigma \), but the other, not expected \emph{a priori}, originates from the virtual states created by the detuned laser which needs them to close the two-photon emission process. This is at energy \( \omega_\nu = 2\omega_\lambda - \omega_\sigma \). Note that, although the states are virtual, both photons are real. We will clarify this statement in the following. The prediction from such a picture, indeed confirmed experimentally [28], is that such a two-photon emission comes as a cascade: first the virtual-state’s photon then the emitter’s. While such considerations have been made and confirmed a long time ago, recent reports by Masters et al [29] of the observation of bunching from the incoherent part of the spectrum of the low-driving detuned resonance fluorescence, and by Long Ng et al [30] of cross-correlations from the high-driving but also detuned Mollow triplet, bring back such important questions in the limelight of modern setups and the

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**Figure 1.** Scattering picture of detuned resonance fluorescence, in the perturbative treatment of Dalibard and Reynaud [20]. Regardless of detuning, the spectral shape is the same: the central peak is the Rayleigh peak that elastically scatters the laser photon (depicted as a thin blue line topped by the \( \circ \) symbol). The detuning places the emitter on one side (upper row) or the other (lower row), while two-photon energy conservation places a copycat peak on the other side of the laser as a result of the virtual states created to fill in the loop (second column). The spectral shape is shown, in (e), for the Heitler regime of vanishing driving \( \Omega_\sigma \to 0 \) and, in (f), at nonzero driving, producing an emerging fluorescence peak at the center of the triplet and slightly increasing the triplet splitting.
improved accuracy affordable today. In particular, modern authors envision quantum-optical technological prospects, which were not at the core of the preoccupations of the founding fathers who were worrying, instead, on experimental validations of one or the other model of the theory of light–matter interactions, from Dirac’s quantum electrodynamics to dissipative quantum optics. Such recent works can also participate to the experimental validation of, or discrimination between, the more refined theories available today, e.g. compare figure 5 of [30] with figure 4(a) of [31] that itself includes, in addition to the assumed exact cross-correlations between the peaks, results from earlier works [32]. More importantly, they also allow testing more recent and new proposals regarding multiphoton emission from resonance fluorescence. In the following, we discuss our own input to this problem, which starts with the theory of frequency-resolved photon correlations [33]. We articulate our discussion along the experimental findings of Long Ng, Masters et alii.

2. Frequency-resolved photon-correlation and homodyning

Correlating the peaks in figure 1 can be done by filtering them first and directing the respective outputs to a correlator (e.g. an Hanbury Brown–Twiss setup). This was already discussed in precisely these terms by Cohen-Tannoudji. Masters et al chose a different strategy of filtering out the central Rayleigh peak and auto-correlating the output. We will come back to their approach but discussing first the more traditional one that has been considered several times [28, 34], we must highlight the input by Schrama et al [32]. They implement the photodetection theory [35], though at, or close to, resonance and in the high (Mollow) driving regime, but more importantly, with some approximations in the model to undertake the complex calculations involved in the way the theory was then formulated (as nested, time-ordered, high-dimensional integrals). We provided an alternative, numerically exact as well as efficient, formulation of the problem that allows us to compute faithfully frequency-resolved n-photon correlations [33], in some cases even analytically. This consists in enlarging the original problem (in this case resonance fluorescence) to n two-level systems $\xi_i$ (in this case, $n = 2$) that are coupled both through an Hamiltonian $H_i$ and Liouvillian $\mathcal{L}_n$ to the system (in units of $\hbar = 1$):

$$\partial_t \rho = -i[\omega_i, \sigma^+ \sigma^-] + \Omega(\sigma^+ + \sigma^-), \quad \rho + \frac{2\omega}{2} \mathcal{L}_n \rho$$

where the first line in equation (1) is the resonance fluorescence problem in its simplest possible and modern quantum-optical formulation, while the second line implements the sensor formalism where $\omega_i$ set the frequencies which are to be correlated while $\Gamma_i$ set the filters’ bandwidths. One can then (easily) solve this master equation without worrying about the complicated frequency variables that otherwise enter at the level of parameters in complex integrals, and which have been upgraded here to operators. This makes the original evaluations in terms of folded integrals turn to standard intensity–intensity correlations:

$$g_{\omega_1\omega_2}(\omega_1, t_1, \omega_2, t_2) = \lim_{\epsilon \to 0} \frac{\langle \mathcal{O}(t_1) \mathcal{O}(t_2) \rangle}{\langle \mathcal{O}(t_1) \rangle} \langle \mathcal{O}(t_2) \rangle$$

where the limit $\epsilon \to 0$ is to be taken, providing finite values for Glauber’s correlators since both numerators and denominator are of the same order. Given that we will deal with steady states only, we will compute $g_{\omega_1\omega_2}(\omega_1, \omega_2; \tau)$ with $\tau \equiv t_2 - t_1$ the time delay between the photons, which can be done with the quantum regression theorem. We need not elaborate further here on the method itself, only emphasize again that it provides the exact (according to the established theory of photodetection) correlations between the filtered photons, by detectors with the respective bandwidths at the given time delay. Note that if $\omega_1 = \omega_2$, this describes filtered auto-correlations. In the following, we shall consider that $\Gamma_1 = \Gamma_2$, we discuss the results in next section. We need however to first introduce another idea that relates to the contribution of the Rayleigh peak.

The nature of antibunching in resonance fluorescence is very different in the low-driving (Heitler) and high-driving (Mollow) regimes [8]. In the latter, it follows from the more straightforward and popular picture of the two-level system being with some probability in its excited state and, in its transition back to the ground state, releasing a single photon. The density matrix in the limit of infinite driving is $\rho = \frac{1}{2} |0\rangle \langle 0 | + |1\rangle \langle 1 |$. In stark contrast, in the low-driving regime, antibunching arises from an interference between the displaced squeezed state in which the two-level system is driven to leading order, and the coherent state that is imprinted by the driving laser [27]. The presence of squeezing as well as a strong Poisson content from the laser means that the antibunched emission is of a more subtle multiphoton character in this case. Photodetection tampers with the squeezing in a way that is fundamentally equivalent to frequency filtering. This disturbs the interference that otherwise yields a perfect antibunching. We have shown with López Carreño how one can, however, restore perfect antibunching by correcting for the excess coherence from the laser as a result of filtering the tails of the
incoherent spectrum [5]. We later addressed the case of detuned resonance fluorescence in the context of filtered-homodyned correlations [36], but we did so for antibunching. In the next section, we will instead phase-shift our focus to consider the case of multiphoton emission, which is the one revived by Masters et al [29]. The formalism is exactly the same, the regime and interpretations however deserve considerations of their own. We also later generalized the scheme to a large range of platforms where coherence is involved in some form [37], in which case we have shown that such multiphoton interferences are key to explaining the structure and type (conventional or unconventional) of a wide range of correlations, from antibunching to superbunching. As a general statement, one can in the framework that we have just laid down, seize additional control of the system by homodyning, i.e. externally changing the nature (constructive or destructive) of the interference and further selecting auto or cross-correlations to characterize the system, resulting in a much-enhanced versatility and performance of its emission. We do this in the following in the case of two-photon emission in detuned resonance fluorescence.

3. Removing the central peak

We now turn to the exact computation of correlations in the case where the laser is (with no loss of generality, since solutions are symmetric) red-detuned at \( \omega_\ell \) as compared to the atom at \( \omega_\ell = \omega_\ell + \Delta_\ell \) (cf. figure 1 (c), (e)). The main question addressed by Masters et al is whether the side peaks are bunched, what they interpret as simultaneous two-photon emission. We come back to qualify further this interpretation but first consider the question itself. In their case, they used a narrowband notch filter to attenuate the central peak, and let otherwise pass the other photons, i.e. from the side peaks, which they autocorrelate. The ideal version of this experiment is precisely our homodyning scheme, unfiltered, where we remove the central peak by destructive interference. Indeed, we have checked that the two-photon coincidence for the notch filter of width \( \Gamma \) and centered at \( \omega_\ell \) when filtering out the coherent Rayleigh peak, agrees in the limit \( \Omega_\ell \to 0 \) with the full-homodyned zero-delay coincidence, that is given by \( g_\ell^{(2)}(0) = (\gamma_\ell^2 + 4\Delta_\ell^2)(\gamma_\ell^2 + 4[8\Omega_\ell^2 + \Delta_\ell^2])/(64\Omega_\ell^4) \). The general and time-resolved case is shown in figure 2 from, bottom, no homodyning (corresponding to \( F = 0 \)), up to, top, full-destructive homodyning (\( F = 1 \)) that completely removes the laser contribution. In the first case, we are looking there at the full autocorrelation of the light itself. This is a well-known result: the antibunching is perfect but oscillates strongly, which is usually (and correctly) interpreted as the effect of detuning. As the coherent peak is removed, one can see how antibunching is gradually lost, along with the oscillations and with decay rate \( \Gamma \), i.e. emitted before the triggering photon, with probability \( 1 - p \) and decay rate \( \gamma_\ell \). In this case, we find that:

4. Cross-correlations of the side peaks

So far, we have not actually filtered the emission. Instead, we used homodyning to remove a particular part of the spectrum, namely, the central scattering peak. We now bring filtering to similarly remove this central peak, by placing two detectors (or filters) on the side peaks, although what we actually gain in this way is the possibility to turn to cross-correlations. This can tell us about the cascaded emission, i.e. whether one photon comes before the other, which was the actual prediction from the pioneering papers of the two-photon character in this case, as opposed to simultaneous emission. We first provide the ideal two-photon cascade cross-correlation \( g_\ell^{(2)}(\tau) \), defined as an uncorrelated (Poisson) stream (1) of photons with emission rate \( \gamma_\ell \), each photon of which triggers the emission of another photon in another stream (2), in the good time order, i.e. emitted at a later time, with probability \( p \) and with decay rate \( \gamma_2 \), or in the wrong time-order, i.e. emitted before the triggering photon, with probability \( 1 - p \) and decay rate \( \gamma_2 \). In this case, we find that:
Note that there is a discontinuity at $\tau = 0$ which prevents to define $g^{(2)}(0)$, that is however of no concern since this occurs at a single point (in a practical context, one can take either limit $\pm \tau \to 0$ or an average). A physical mechanism approaching this ideal scenario would besides connect smoothly the two domains. In our case, the exact numerical results for the filtered two-photon correlations of resonance fluorescence are shown in figure 3 for various critical parameters being varied one at a time. Taken in turns, we find that:

(a) With no (or very broad, $\Gamma \gg \gamma_0$) filtering, there is no asymmetry and there is a strong antibunching, recovering in this case the full correlation of the complete spectrum. As the filtering tightens around the peaks (i.e. $\Gamma$ decreases), an asymmetry develops in the form that is typical of a two-photon cascade, cf. equation (3), with a transition to bunching that is maximum at positive $\tau$. The correlations also exhibit strong oscillations. For smaller still values of $\Gamma$ (narrow filters), correlations increase but the asymmetry reduces. In the limit of $\Gamma \to 0$, correlations diverge and become $\tau$-independent (flat).

(b) As a function of detuning $\Delta_\sigma$, one goes from antibunching at resonance ($\Delta_\sigma = 0$) to a growth of the asymmetry and increase of the correlations. In this case, the trend is consistent and both the asymmetry and correlations increase to realize the case of an increasingly better two-photon cascade.

(c) As a function of driving $\Omega_\sigma$, one goes from the Heitler ($\Omega_\sigma \to 0$) to the detuned Mollow (growing an incoherent central peak) and ultimately to the resonant Mollow triplet $\Omega_\sigma \gg \gamma_0$ as detuning becomes negligible. In this case, the cascade asymmetry remains constant while the strength of the correlations decreases and the oscillations dampen.

(d) Finally, as a function of pure dephasing $\gamma_f$, the asymmetry also remains constant as the correlations weaken, although in this case better retaining the oscillations and also recovering some antibunching for very large dephasing, due to spectral overlap induced by the line broadening.
These observations are informative regarding the general character of the two-photon emission, which we can summarize and understand as follows: there is a clear cascade, i.e. one photon comes before the other, as is consistent with the two-photon scattering picture; this is better resolved when photons are adequately detected, i.e. there is an optimum filter width to optimize the effect with too narrow filters randomly delaying photons and too broad ones making them indistinguishable; the cascade gets better with increasing detuning, forcing the system into the perturbative two-photon limit; correlations are stronger for weaker driving, i.e. the Heitler regime is more correlated than the Mollow one but oscillations are also more pronounced; dephasing damages the process by acting as erasing, and even reverts to the opposite regime of single-photon emission. Also, it should be emphasized once more that these results are supposedly exact, since they have been obtained with no approximations from the filtered and photo-detection side once given the model (which is that from Mollow). It becomes particularly interesting, however, to now combine homodyning and cross-correlation filtering, which we do in the next section.

5. Cross-correlations of the homodyned side peaks

We now repeat the same procedure as previously, cross-filtering the two peaks, but removing the central peak as well. We do that by homodyning, since this is the ideal scenario, but this could experimentally be approached by cross-correlating the output of Masters et al’s narrowband notch filter. The results are shown in figure 4 and are to be contrasted one-to-one with those of figure 3, which was including the central coherent peak. While one could assume that cross-correlating the side peaks is tantamount in the first place to remove the central peak, interference filters have tails and their suppression is not perfect, therefore some residual physics of interference leaks through and impact the results. This is why figure 2 differs from Masters et al’s filter approach: their suppression of the coherent peak in this way is partial only. The impact turns out to be, maybe surprisingly, momentous. One needs only compare the strong—both qualitative and quantitatively—departures between the non-homodyned and homodyned versions to appreciate the importance and value of this modus operandi.

First, notice that the scale up to $\approx 10^6$ in figure 3 needs be inflated to over $10^{10}$ in figure 4: correlations are considerably stronger. Also, the various cases split neatly the ones from the others, with the exception of filtering, which tends to now feature more similar magnitudes of correlations, while it retains an optimum filter width to maximize the asymmetry. Finally, oscillations are very much, sometimes completely suppressed. Understanding these oscillations as a way for the system to provide one-photon emission from a two-photon mechanism by synchronizing them, this means that, by removing the central peak, we manage to enter more deeply into a more
genuine two-photon physics. The shapes observed in this case are very close to the ideal two-photon cascade, equation (3), with identical slopes and thus decay rates \( \gamma_2 = \gamma_1 \) for the ordered and out-of-order photons, as expected for a biphoton. The rate is
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\frac{2}{2} \bar{\gamma}_g = \frac{2}{2} \bar{\gamma}_g = W + D \gg \text{in the Heitler regime while the fast-rising slope bridging the gap is given by } \Gamma, \text{ the filters' width. For } \gamma_2 = \gamma_1, \text{ this gap allows from equation (3) to estimate both the ratio } \gamma_2/\gamma_1 \text{ (absolute gap obtained by the differences between the } \pm \tau \to 0 \text{ limits) and the probability } p \text{ (relative gap obtained by their ratio for high enough correlations), from which one can see that the time-ordering is very good, with the } \omega_\upsilon \text{ photon arriving with high probability before the } \omega_\sigma \text{ one. It should be noted, however, that two-photon emission is not fully captured by the two-photon correlations alone. For instance, the probability that the emission of one photon successfully heralds the other is not captured by a } g^{(2)} \text{ measurement, which is not affected by photon losses, although the symmetry of the peaks in luminescence suggests that this also is very high. Still, a thorough description of the two-photon emission in this regime is a separate problem. Nevertheless, all these facts together confirm that an ideal two-photon cascade can be approached in detuned resonance fluorescence, at large detunings, for filter widths commensurable with the width of the peaks and when suppressing the coherent Rayleigh peak.}

We conclude with an interesting result. While the process is more efficient without dephasing, its correlations are not seriously affected by it, but the impact of dephasing on the signal itself is dramatic. Two-photon emission from the side peaks of detuned resonance fluorescence is very weak in intensity, being second-order in the driving. Most of the emission originates from the central peak, and removing it, naturally leaves only little emission. A tiny amount of pure dephasing changes considerably both the spectral shape and the amount of incoherent emission: the perfectly symmetric spectral shape collapses with very small dephasing, at the same time as the intensity grows considerably. The symmetric peak at \( \omega_\upsilon \) remains the same (i.e. remains very small), but a channel is now opened for bare, incoherent emission from the real transition at \( \omega_\sigma \). This means that the two-photon diagrammatic picture gets substituted by a more straightforward one of a classical oscillator driven out of resonance and emitting at its natural frequency, which is a first-order process, and therefore of considerably higher intensity than the two-photon emission. The effect was reported by several authors, with dephasing due to atomic collisions in a gas \[38\] or to fluctuations from the driving laser for a single atom \[39\], and investigated under further various variations of the same theme of an asymmetry induced by decoherence \[40–43\]. In our case, it achieves the demonstration that two-photon emission is a fragile quantum coherent process, of weak intensity. The dwarfing of the satellite peak on the other side from the emitter is also more dramatic as \( \Omega_c \) goes to zero, showing that this is a Heitler, two-photon effect. Experimentally, it is likely that some amount of dephasing is present but is counteracted by some amount of driving. Observing a collapse of the satellite peak—the more abruptly and with a greater gain of intensity from the other (emitter) peak, the lower the

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**Figure 4.** Same as figure 3 but without the coherent peak, i.e. with homodyning \( \mathcal{F} = 1 \) to suppress it. This results in a considerable improvement in the two-photon cascade character of the emission.
driving—would be a measure of how deep one is in the Heitler regime as well as an experimental demonstration of two-photon physics directly at the level of photoluminescence.

6. Summary and conclusions

We discussed the two-photon correlations in detuned resonance fluorescence, whose spectral shape—two-side peaks sitting around a central Rayleigh scattered peak (at low enough driving)—lends itself to a natural interpretation in terms of two-photon scattering, as presented by Cohen–Tannoudji, Reynaud, Dalibard and others. In particular, we contrasted our theoretical results to those aimed by Masters et al of suppressing the coherent peak. They achieved this by filtering it out and otherwise auto-correlating the emission that passes through, finding a clear transition from antibunching with strong oscillations to bunching with reduced—but always present—oscillations. In our case, we can completely remove the coherent peak by perfect destructive interferences with an external (homodyning) laser. We find essentially the same result as Masters et al of a transition from perfect antibunching with strong oscillations, when the coherent peak is present, to bunching when removing it, although in our case there is also a disappearance of the oscillations. We then turn to cross-correlations of the peaks, showing how they bear strong features of a two-photon cascade indeed, that get considerably improved by removing the coherent peak also in this case. This shows that homodyning is a valuable additional concept, rather than a supplement, to frequency filtering, and can lead, in the case of resonance fluorescence, to a basically ideal cascaded two-photon emission. These considerations are only a small part of the general picture of multiphoton correlations and emission from resonance fluorescence, which generalizes to higher photon numbers, involving other types of scattering processes (prominently, leapfrog processes [44, 45]) and calling for further investigation of their fundamental character as well as relevance for technology. The interpretations of the underlying physics are interesting but intrinsically limited by one’s mental picture(s). The two-photon diagrams in figure 1 provide a compelling mechanism of a coherent quantum character, but they do not include the subtle multiphoton interferences with the Rayleigh peak and the detection process, which are crucial for a comprehensive description. As befits quantum mechanics, the best one can do is to compute observables. All interpretations will be constrained in one way or another, although useful applications could derive from them. In that regard, we have provided a formalism to achieve exact results. It is remarkable that the simplest treatment of the simplest system, already at the time of Mollow, yields the best results while more sophisticated techniques, more suitable for more complex systems, come with some approximations. We ourselves suspect that the driven two-level system could be the richest and most fundamental source of multiphoton physics and that more complex systems could, instead of bringing additional physics, result in imperfect limits of this ultimate realization. There are many platforms to implement this physics, from atoms [46] to ions [47] and molecules [48] passing by semiconductors (quantum dots [49], spin [50], NV centers [51], etc), superconducting qubits [52] and circuits [53], and still others [54–56]. While the interest for basic and fundamental multiphoton emission originates from the early days of quantum electrodynamics, recent progress from both material, technological and theoretical sides makes it a particularly burgeoning field of study.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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